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RADIOACTIVE WASTE DISPOSAL

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CHAPTER 1

INTRODUCTION

Waste disposal problems are as old as civilization itself. The more complex civilization becomes, the more complex the problems become. The advent of the nuclear energy industry presented a whole new spectrum of problems. Fortunately, this industry started in an age when an awareness of waste disposal problems had begun to develop. The industry began in earnest during a war when all emphasis was directed toward the primary goal of producing fissionable material and converting it into a weapon. Treatment of wastes was something to be postponed until the main job was accomplished. Fortunately, the postponement was done in such a manner as to minimize the insult to the environment. It consisted of venting gases to the atmosphere through tall stacks, burying solids in waste-land areas, and holding liquid wastes in underground retention tanks. These temporary solutions worked well; so well, in fact, that all are still in use and in all probability each will have some place in the systems finally adopted for handling wastes from a nuclear power economy.

In the intervening years there has been a great deal of work done on the treatment of wastes, and the waste processor now has at his disposal a very considerable arsenal of information and available processes In this discussion there will be considered the nature of radioactive wastes, the philosophy of their handling, the origin of wastes in the nuclear fuel cycle, waste management as it is practiced today treatment of gaseous, solid, and liquid wastes, the question of ultimate disposal, and some estimates of what this problem may become in the future

Nature of Radioactive Wastes

Wastes from the nuclear energy industry are unique in at least three respects·

1) Their radioactivities cannot be detected by any of the human senses. However, with appropriate instrumentation the nature, character, and concentration of the wastes may be determined with great accuracy.

2) The toxicity of some of them is greater than that of any hitherto known industrial waste.

3) There is no known practical process by which they can be destroyed other than to allow them to decay to innocuousness by their own natural disintegration. Some of them (those with short half-lives) do this quite rapidly; but many of them have very long half-lives and these take centuries to disappear.

Our responsibilities to future generations demand that such wastes be guaranteed supervision and control for very long periods of time.

The radioactivity which finds its way into waste streams may occur naturally, or be produced by fission or by neutron activation in nuclear reactors. For substantial amounts of a radionuclide to have survived the interval since the elements were created (estimated at $\sim 6 \times 10^9$ years) the nuclide must have a half-life of at least 3×10^8 years. While a few lighter elements have naturally occurring radioactive isotopes (potassium-40 is a notable example), uranium and thorium and their respective daughter products are the important radionuclides of this class. These, together with some of their properties, are shown in Table 1.

Uranium-235, uranium-233, and plutonium-239 may undergo fission, thereby producing a variety of fission products comprising the whole middle portion of the periodic table starting with element no. 30 (Zn^{72}) and ending with element no. 66 (Dy^{161}). From the standpoint of waste disposal only a few of these, those with long half-lives and high hazard potential to man, are significant. A dozen of the more important of these are listed in Table 2 together with their half lives, fission yields, and maximum allowable concentrations in the body and in water.

As born in fission, the fission products are distributed according to the familiar saddle-shaped curve of yield vs mass number. But each fission product immediately begins to decay with its characteristic half-life. Since these half-lives vary from seconds to centuries, the composition of mixed fission products in a reactor after startup and in spent fuel or waste streams after discharge from the reactor is a constantly changing function of time.

In an operating reactor the accumulation of any particular fission product over a differential time element dt can be expressed by the equation:

$$\frac{dn}{dt} = \frac{RYM}{235} - \lambda n \qquad (1)$$

Table 1

SOME NATURALLY OCCURRING RADIONUCLIDES

Radionuclide	Natural Abundance in Nature[a] Per cent	Mode of Disintegration[f]	Half-life[e]	Maximum Allowable Conc[g]	
				in Body (μc)	in Water[j] (μc/ml)
Uranium 238	99.28	α	4.5×10^9 y	5×10^{-3}	4×10^{-4}
Thorium 234	b	β	24.5 d	4	2×10^{-4}
Protactinium 234	b	β, IT[h]	6.7 h	-	-
Thorium 230	b	α	8.0×10^4 y	0.05	2×10^{-5}
Radium 226	b	α	1,600 y	0.1	10^{-7}
Radon 222 (gas)	b	α	3.8 d	-	-
Uranium 235	0.71[c]	α	7.1×10^8 y	0.03	3×10^{-4}
Uranium 234	0.005[b]	α	2.5×10^5 y	0.05	3×10^{-4}
Thorium 232	100.00	α	1.4×10^{10} y	0.04	2×10^{-5}
Radium 228	d	β	6.7 y	0.06	3×10^{-7}
Thorium 228	d	α	1.9 y	0.02	7×10^{-5}
Radium 224	d	α	3.6 d	0.06	2×10^{-5}
Radon 220 (gas)	d	α	54.0 s	-	-
Bismuth 212	d	α, β	1.0 h	0.01	4×10^{-3}
Polonium 212	d	α	Very short	-	-
Thallium 208	d	β	3.0 m	-	-

[a] F. L. Culler, Nature of Radioactive Wastes, ORNL-CF-59-1-106 (Jan. 26, 1959). Paper prepared for the record of the JCAE hearings on Waste Disposal, January 1959.

[b] More important decay products of U^{238}

[c] Decay products of U^{235}; not very important due to low concentration of parent.

[d] More important decay products of Th^{232}

[e] s = second; m = minute; h = hour; d = day; y = year.

[f] Hanson Blatz, editor, Radiation Hygiene Handbook, Section 6, McGraw-Hill Book Co., New York (1959).

[g] U.S. Bureau of Standards Handbook 69

[h] Isomeric Transition

[j] For continuous exposure.

Table 2

PRINCIPAL FISSION PRODUCTS OF INTEREST
IN WASTE DISPOSAL OPERATIONS[a]

Radionuclide	Fission Yield,[e,f] Atoms/100 Atoms U^{235} Fissioned	Half-life[b]	Max Allowable Concentration[c]	
			In body (μc)	In water[d] (μc/ml)
Strontium 90	5.9	28.0 y	2	10^{-6}
Cesium 137	5.9	26.6 y	30	2×10^{-4}
Promethium 147	2.6	2.6 y	60	2×10^{-3}
Cerium 144	6.1	290.0 d	5	10^{-4}
Krypton 85 (gas)	0.3	10.3 y	-	-
Iodine 131 (gas)	2.9	8.1 d	0.7	2×10^{-5}
Zirconium 95	6.4	63.0 d	20	6×10^{-4}
Barium 140	6.3	12.8 d	4	3×10^{-4}
Ruthenium 103	2.9	41.0 d	20	8×10^{-4}
Ruthenium 106	0.38	1.0 y	3	10^{-4}
Strontium 89	4.8	54.0 d	4	10^{-4}

[a] F. L. Culler, Nature of Radioactive Wastes, ORNL-CF-59-1-106 (Jan. 26, 1959). Paper prepared for the record of the JCAE hearings on Waste Disposal, January 1959. (Modified).

[b] d = days; y = years.

[c] U.S. Bureau of Standards Handbook 69

[d] For continuous exposure

[e] For thermal fission

[f] Harold Etherington, editor, Nuclear Engineering Handbook, Section 11, McGraw-Hill Book Co., New York (1958).

where

n = mass of isotope

λ = decay constant = $0.693/t_{\frac{1}{2}}$

($t_{\frac{1}{2}}$ = half-life in convenient time units)

Y = fission yield

M = atomic weight of species considered

R = rate of fission of U^{235} in mass/time (time units consistent with those used for λ).

The solution of equation (1) when $n = 0$ at $t = 0$ is:

$$n = \frac{RYM}{235\lambda}\left[1 - e^{-\lambda t}\right] \qquad (2)$$

This states that each fission product will approach a limiting concentration, $RYM/235\lambda$, the approach being the more rapid the shorter is the half-life of the particular species. This equation neglects the destruction of accumulating fission products within the reactor by neutron absorption. For those species with large absorption cross sections (e.g., Xe^{135} and Sm^{149}) a correction must be made for this factor. The correction results in a lower equilibrium value than that given by these equations Furthermore, equation (2) is accurate only for the first significant member of a decay chain or for any member all of whose progenitors have reached their equilibrium values. For intermediate times the effect of decay chains must be taken into account.

Once the fuel has been discharged from the reactor, only decay equations are needed. Generally, the second decay is to a stable or very long-lived product; thus for the chain

$$A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C_{stable}$$

the only equations needed are

$$\frac{dA}{dt} = -\lambda_A A \qquad (3)$$

and

$$\frac{dB}{dt} = \lambda_A A - \lambda_B B \qquad (4)$$

Upon solution of these equations there are obtained

$$A = A_0 e^{-\lambda_A t_C} \tag{5}$$

$$B = \frac{\lambda_A A_0}{\lambda_B - \lambda_A} e^{-\lambda_A t_C} + \left[B_0 - \frac{\lambda_A A_0}{\lambda_B - \lambda_A} \right] e^{-\lambda_B t_C} , \tag{6}$$

where A_0 and B_0 are the amounts of A and B present at discharge, as calculated from equation (2), and t_C is the cooling time, again in consistent units. These equations have been solved for thermal fission of uranium-235[1] and for fast fission of uranium-235 and plutonium-239.[2] Very precise calculations have been done at Oak Ridge by means of the computer ORACLE.[3]

In the operation of a nuclear reactor some of the neutrons do not effect collisions with fissionable materials but rather are absorbed by the coolant, its contained impurities, or by structural components, to form radioactive isotopes of the absorbing material. These are referred to as activation products. A list of some of the principal coolant and impurity activation products is given in Table 3.

Finally, there occur in a reactor a number of side reactions which lead to highly active isotopes of uranium, plutonium and higher transuranic elements. These reactions consist of neutron absorption (n, γ) and neutron absorption and emission (n, 2n) interspersed with α or β-decay schemes. The patterns are often very complex. As burnups are increased and fuel is recycled, products so formed will become more and more important in waste disposal operations. Table 4 shows some of these heavy isotopes, their sources, half-lives, and allowable concentrations in the body and in water.

Radioactive wastes occur as gases, liquids, or solids, or as mixtures of these. Liquids and gases often contain suspended particulate matter. In fact, most of the activity in waste gas streams is usually associated with suspended solids. Solids may be wet with liquids and liquids may frequently give off a gaseous product at some stage in their handling. The treatment and handling of wastes will be considered, however, in terms of the carrier phase.

Radioactive wastes vary in activity content over a wide range. Liquid wastes, for instance, may contain activity from at or near natural background, about 10^{-16} c/ml, up to very high-level wastes which contain more than 1 c/ml. Part of the great difficulty in dealing with these wastes is that the general public does not understand the import of this wide range of concentration and the fact that wastes at the ends of the range can, and must, be treated differently. Low-level wastes are characterized by high volume and low or nonexistent hazard potential.

Table 3

COOLANT AND IMPURITY ACTIVATION PRODUCTS

Radionuclide[a]	Half-life[a,d]	Mechanism of formation[a]	Maximum Allowable Concentration[b]	
			in Body (μc)	in Water[c] (μc/ml)
Water Reactors				
Coolant activity:				
Nitrogen 16	7.0 s	$O^{16}(n,p)N^{16}$	-	-
Nitrogen 17	4.0 s	$O^{17}(n,p)N^{17}$	-	-
Oxygen 19	30.0 s	$O^{18}(n,\gamma)O^{19}$	-	-
Fluorine 18	1.9 h		20	5×10^{-3}
Impurity activity:				
Sodium 24	15.0 h	$Al^{27}(n,\alpha)Na^{24}$	7	2×10^{-3}
Aluminum 28	2.0 m	$Al^{27}(n,\gamma)Al^{28}$	-	-
Argon 41	110.0 m	$A^{40}(n,\gamma)A^{41}$	-	-
Manganese 56	2.6 h	$Fe^{56}(n,p)Mn^{56}$	2	10^{-3}
Cobalt 58	71.0 d	$Ni^{58}(n,p)Co^{58}$	30	10^{-3}
Cobalt 60	5.2 y	$Co^{59}(n,\gamma)Co^{60}$	10	5×10^{-4}
Iron 55	2.9 y	$Fe^{54}(n,\gamma)Fe^{55}$	10^3	8×10^{-3}
Iron 59	45.0 d	$Co^{59}(n,p)Fe^{59}$	20	6×10^{-4}
Chromium 51	27.0 d	$Cr^{50}(n,\gamma)Cr^{51}$	800	0.02
Copper 64	12.8 h	$Cu^{63}(n,\gamma)Cu^{64}$	10	2×10^{-3}
Tantalum 182	111.0 d	$Ta^{181}(n,\gamma)Ta^{182}$	7	4×10^{-4}
Tungsten 187	24.0 h	$W^{186}(n,\gamma)W^{187}$	30	6×10^{-4}
Sodium Reactors				
Sodium 24	15.0 h	$Na^{23}(n,\gamma)Na^{24}$	7	3×10^{-4}
Sodium 22	2.6 y	$Na^{23}(n,2n)Na^{22}$	10	3×10^{-4}
Rubidium 86	19.5 d	$Rb^{85}(n,\gamma)Rb^{86}$	30	7×10^{-4}
Antimony 124	60.0 d	$I^{127}(n,\alpha)Sb^{124}$	10	2×10^{-4}

[a] Summary-Analysis of Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, page 9, August, 1959.

[b] U.S. Bureau of Standards Handbook 69

[c] For continuous exposure

[d] s = seconds; m = minutes; h = hours; d = days; y = years

Table 4

FORMATION OF SOME HEAVY ISOTOPES

Isotope	Type of Decay	Half-life	Method of Production	Maximum Allowable Concentration[a]	
				in body (μc)	in water[b] (μc/ml)
U^{232}	α	74 y	Daughter of Pu^{236}	0.01	3×10^{-4}
U^{233}	α	1.6×10^{5} y	Daughter of Pa^{233}	0.05	3×10^{-4}
U^{236}	α	2.4×10^{7} y	$U^{235}(n,\gamma)$	0.06	3×10^{-4}
U^{237}	β	6.75 d	$U^{238}(n,2n)$ Daughter of Pu^{241}	-	-
U^{239}	β	23.5 m	$U^{238}(n,\gamma)$	-	-
Np^{236}	β	22 h	$Np^{237}(n,2n)$	-	-
Np^{237m}	IT	6.3×10^{-8} s	Daughter of Am^{241}	-	-
Np^{237}	α	2.2×10^{6} y	Daughter of U^{237}	0.06	3×10^{-5}
Np^{238}	β	2.1 d	Daughter of Am^{242} $Np^{237}(n,\gamma)$	-	-
Np^{239}	β	2.3 d	Daughter of U^{239}	30	10^{-3}
Pu^{236}	α	2.7 y	Daughter of Np^{236}	-	-
Pu^{238}	α	90 y	Daughter of Np^{238} Daughter of Cm^{242}	0.04	5×10^{-5}
Pu^{240}	α	6,300 y	$Pu^{239}(n,\gamma)$	0.04	5×10^{-5}
Pu^{241}	β 99+% $\alpha \sim 10^{-3}$%	13 y	$Pu^{240}(n,\gamma)$	0.9	2×10^{-3}
Pu^{242}	α	$\sim 5 \times 10^{5}$ y	$Pu^{241}(n,\gamma)$	0.05	5×10^{-5}
Pu^{243}	β	5 h	$Pu^{242}(n,\gamma)$	-	-
Am^{241}	α	470 y	Daughter of Pu^{241}	0.05	4×10^{-5}
Am^{242m}	β 60%, EC 20% IT 20%	16 h	$Am^{241}(n,\gamma)$	-	-
Am^{242}	$\beta \sim 99$% $\alpha \sim 1$%	100 y	$Am^{241}(n,\gamma)$	-	-
Am^{243}	α	880 y	$Am^{242}(n,\gamma)$ Daughter of Pu^{243}	0.05	4×10^{-5}
Cm^{242}	α	163 d	Daughter of Am^{242}	0.05	2×10^{-4}
Cm^{243}	α	100 y	$Cm^{242}(n,\gamma)$	0.09	5×10^{-5}
Cm^{244}	α	17.9 y	$Cm^{243}(n,\gamma)$	0.1	7×10^{-5}

[a]U.S. Bureau of Standards Handbook 69

[b]For continuous exposure

High-level wastes, on the other hand, are characterized by quite low volumes but a radioactivity content which is not only a serious hazard now, but will continue to be one for many years. It is a simple economic fact that low-level, low-hazard waste must receive a minimal treatment before discharge if the cost of waste treatment is not to "purify us out of business."[4] And it is a simple biological fact that high-level, high-hazard wastes must receive complete treatment, no matter what the cost, if we are not to do serious injury to our environment.

Definitions

There are a number of units and terms used herein which need definition, some because they are often used loosely; others are included for ready reference.[5]

Units

Curie (c) That quantity of any radionuclide in which the number of disintegrations per second is 3.70×10^{10}. An earlier definition was that quantity (grams) of radon in equilibrium with 1 gram of radium.

Millicurie (mc) One-thousandth of a curie.

Microcurie (μc) One-millionth of a curie.

Roentgen (r) That quantity of X or gamma radiation such that the associated corpuscular emission per 0.001293 gram of air produces, in air, ions carrying 1 esu of electricity of either sign.

Milliroentgen (mr) One-thousandth of a roentgen.

Roentgen equivalent, man, or mammal (rem) The dose of any ionizing radiation that will produce the same biological effect as that produced by one roentgen of high-voltage X radiations.

Roentgen equivalent, physical (rep) A unit of absorbed dose of radiation with a magnitude of 93 ergs per gram of absorbing material (usually soft tissue).

Rad One hundred ergs of absorbed energy per gram of absorbing material.

Electron volt (ev) A unit of energy equal to the energy gained by a particle having one electronic charge when it passes in a vacuum through a potential difference of 1 volt. 1 ev = 1.60×10^{-12} erg.

Terms

Background Ever-present effects in physical apparatus above which a phenomenon must manifest itself in order to be measured. "Background" can take various forms, depending on the nature of the measurements. In electrical measurements of radioactivity and nuclear phenomena, the term usually refers to those undesired counts or currents that arise from cosmic rays, local contaminating radioactivity, insulator leakage, amplifier noise, power-line fluctuations, and so on. In nuclear work and photographic emulsions, the term refers to developable grains unrelated to the tracks under investigation.

Background radiation Radiation arising from radioactive material other than the one directly under consideration. Background radiation due to the cosmic rays and natural radioactivity is always present. There may also be background radiation due to the presence of radioactive substances.

Dose or Dosage According to current usage, the radiation delivered to a specified area or volume or to the whole body. Units for dose specification are roentgens for X or gamma rays, reps or equivalent roentgens for beta rays. In radiology the dose may be specified in air, on the skin, or at some depth beneath the surface; no statement of dose is complete without specification of location. In recent years there has been an increasing tendency to regard a dose of radiation as the amount of energy absorbed by tissue at the site of interest per unit mass. (See rad.)

Maximum permissible dose (MPD) Maximum dose of radiation which may be received by persons working with ionizing radiation.

Maximum permissible concentration (MPC) That concentration of radioactivity in air or water which under specified conditions is expected to give rise to a MPD.

Permissible dose The amount of radiation which may be received by an individual within a specified period with expectation of no significantly harmful result to himself.

Tolerance dose Synonym for "permissible dose." The latter is generally considered the preferable term.

Half-life, biological The time required for the body to eliminate one-half of an administered dose of any substance by regular processes of elimination. This time is approximately the same for both stable and radioactive isotopes of a particular element.

Half-life, effective Time required for a radioactive element fixed in the tissue of an animal body to be diminished 50 per cent as a result of the combined action of radioactive decay and biological elimination.

$$\text{Effective half-life} = \frac{\text{biological half-life} \times \text{radioactive half-life}}{\text{biological half-life} + \text{radioactive half-life}}$$

Relative biological effectiveness (of radiation) (RBE) The inverse ratio of the doses of two different radiations necessary to produce the same biological effect.

High-level waste A characterization of the activity level of waste which has been, and will probably continue to be, used loosely. Herein it will mean liquids containing more than $10^{-2}\mu c/ml$; gases containing more than $10^{-4}\mu c/ml$; solids reading more than 50 mr/hr on contact or containing more than 1 mc of alpha activity.

Low-level waste Any waste containing activity at concentrations less than those specified for high-level waste.

Intermediate-level waste An even looser term generally meaning liquids in the range having 10^{-2} to $10^{4}\mu c/ml$. The term will be used herein sparingly, if at all.

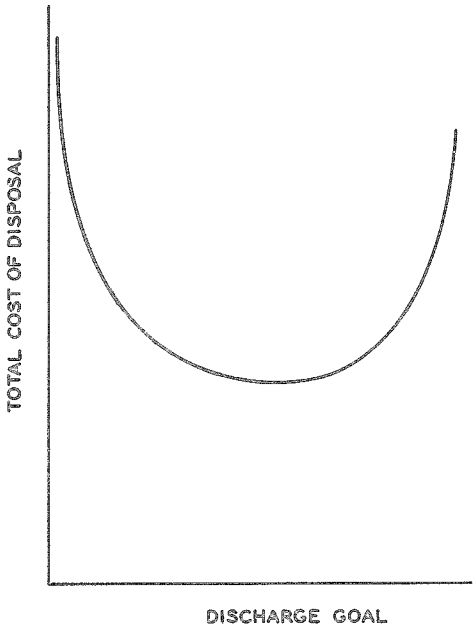
Philosophy

Since there is no practical process whereby the radioactivity of these wastes can be destroyed, there are basically only two things which can be done with them. They may be diluted with nonactive, naturally occurring material until their concentration is so low that they are harmless and may be released to the environment; or they may be concentrated to a point where it is technically and economically feasible to maintain them under control in some manner or other. The first method is called dispersal and the latter containment. There is also a combination of the two called partial containment, which consists of discharging a waste to the environment in such a way that, although control is lost, the rate of return of the activity to that part of the biosphere used by man is so slow that no damage will occur. An example of the first method is the discharge of a waste into the atmosphere or a water way, using the air or water as diluting agents. The second is exemplified by the storing of wastes in tanks for an indefinite period; the third by the practice of discharging a waste into a natural geological formation such that the natural ion exchange properties of the soil plus the calculated time for the liquid to reach ground water and then get back to man is so long that the waste will have decayed by the time it returns.

In devising a waste disposal system it is first necessary to decide which of these three basic methods of approach is going to be used - or what combination of them. From the standpoint of protection of the environment, it would be desirable to discharge no radioactivity. It is impossible, however, to have any waste concentration process so efficient that 100 per cent cleanup can be accomplished. Therefore, while a desirable goal might be to discharge material containing no radioactivity, in practice this is impossible. It is, therefore, necessary to select some realistic disposal level which can be maintained at a reasonable cost. If the total cost of operating a waste disposal system were plotted against the discharge goal, some sort of U-shaped curve similar to that shown in Figure 1 would be obtained. To realize a discharge level approaching zero would be extremely expensive. As the discharge level is relaxed the cost of processing would diminish quite rapidly for a while, the effect dropping off exponentially. As discharge levels were increased, however, the point would be reached where real or imagined injury to the surroundings and its inhabitants would take place. Under these circumstances, damages awarded as the result of law suits would become a highly significant factor and the cost would increase markedly. No scales have been placed upon the axes of Figure 1 for the reason that the actual position of this curve will vary from site to site and is dependent upon many factors. In fact, its position probably cannot be calculated very accurately for any situation. However, some such curve exists and it is desirable to operate any integrated waste disposal system in such a manner that it is located somewhere in the trough of the U.

Figure 1

Relative Cost of Waste Disposal as a Function of Discharge Goal



Finally, still another philosophic decision must be made - that is, just how much segregation of wastes at the source will be practiced. The working scientist prefers to have a single avenue through which all of his waste will flow. If he is permitted to have his way, the waste problem will be one of processing a very large volume of waste which is just a little contaminated. If, on the other hand, the waste processor has his way, wastes will be divided at the source into many classifications by physical state and concentration level. This will result in processing a very much smaller volume of waste at a somewhat higher

level. But a large part of the waste will be discardable directly without treatment. The solutions arrived at usually represent a compromise between these two views.

Concept of Permissible Levels

In discussing dispersal it was indicated that wastes must be discarded at levels which will not be harmful to the environment. This implies that something must be known about the toxicity of the waste. The determination of concentrations which are believed to be harmless is an extremely complex problem. It has involved much work by biologists, physicians, roentgenologists, public health officials, chemists, and radiological physicists. In the U.S., this work has been the responsibility of the National Committee on Radiation Protection under the sponsorship of the National Bureau of Standards. A predecessor of this committee dates back to 1929 and both have always worked closely with the International Committee on Radiological Protection. The findings of the NCRP have been published as Bureau of Standards Handbooks.[6,7] Values published in these Handbooks do not have the force of law, but they are the best numbers available and they are used by essentially everyone. Operations in this country carried out by AEC Licensees are controlled by rules given in Title 10, Code of Federal Regulations, Part 20 - Standards for Protection Against Radiation. These rules, published in the Federal Register, are based on the NCRP values. They do have the force of law for licensees.

Methods of Estimating Permissible Levels.

There are at least five methods which have been used in estimating maximum permissible levels of radiation exposure, maximum permissible amounts of radioisotopes in the body, and the maximum permissible concentrations in air and water.

1. Comparison with X-ray Damage

There has been more than 50 years of experience in the use of X rays. From estimates of dosages received by long-time X-ray workers and observations of their medical histories, a working limit of 100 mr/day was set during the war. Afterward, this was dropped to 0.3 r/week and more recently was set at 5 rem/year.

2. Comparison with Radium Damage

Man's experience with radium fixed in the body goes back more than 25 years. The body burden of an individual can be determined by measuring the total gamma radiation from the body, by measuring the radon exhaled, or by autopsy findings. After correlating as much data as was available, the NCRP set the permissible body burden of Ra^{226} as 0.1μc. Then by estimating the effectiveness of various isotopes as compared to radium, values can be set for each in turn.

3. Comparison with Background Values

The naturally occurring radium content of water varies considerably from one place in the world to another, as illustrated in Table 5. If in some part of the world a large group of people can be found having 10 times the average radium content in their bodies compared to the rest of the people and they have shown no detectable damage from this, the conclusion could be that the higher concentration could be considered safe. Just such a group is being studied at Stateville Penitentiary at Joliet, Illinois (see Table 6).

Table 5

RADIUM CONTENT OF SOME PUBLIC WATER SUPPLIES AND NATURAL WATERS[a]

Location	Water Source	Radium Concentration,[b] μc/ml
MPC[c]		10^{-7}
City Supplies[d]		
Baltimore, Md.	Gunpowder R	0.2×10^{-10}
Bismarck, N.D.	Missouri R	2.4×10^{-10}
Chicago, Ill.	L Michigan	0.24×10^{-10}
Joliet, Ill.	Deep wells	65×10^{-10}
Miami, Fla.	Shallow wells	4.8×10^{-10}
Pittsburgh, Pa.	Allegheny R	37×10^{-10}
Tacoma, Wash.	Green R	0.02×10^{-10}
Natural Waters		
Boulder, Colo.	Curie Spring	2.7×10^{-4}
Shimane, Japan	Hot Spring	7.1×10^{-4}

[a]Hanson Blatz, editor, Radiation Hygiene Handbook, McGraw-Hill Book Co., New York (1959), pp. 4-10, 11.

[b]Reference gives concentrations in g Ra/ml and g Ra/l. For comparison these have been converted to μc/ml, assuming the radium is all Ra^{226} and using 1g = 1 curie.

[c]National Bureau of Standards Handbook 69 for continuous exposure.

[d]Data given are for raw water.

Table 6

MEAN VALUES OF BODY RADIUM FOR GROUPS OF SUBJECTS[a]

Group	Number of Subjects	Mean Age, years	Mean Time at Stateville, years	Mean Body Content of Radium,[b] μc
Maximum Permissible Body Burden[c]				0.1
Chicago Adult	1	29	0	0.4×10^{-4}
Stateville:				
New	11	27	0.3	1×10^{-4}
Intermediate	8	38	7.6	2×10^{-4}
Long Term	11	44	19.7	2.4×10^{-4}
Chicago Boys	7	16.6	0	0.4×10^{-4}
Lockport Boys	8	16.6	0	3.7×10^{-4}

[a]A. F. Stehney and H. F. Lucas, Jr., Studies on the Radium Content of Humans Arising from the Natural Radium of Their Environment, Proceedings of the First International Conference on the Peaceful Uses of Atomic Energy, Geneva (1955) Vol. 11, p. 49.

[b]Reference reported data in units of 10^{-10} g of radium. Converted for comparison assuming all the radium is Ra^{226} and 1 g = 1 curie.

[c]National Bureau of Standards Handbook 69 for continuous exposure.

4. Experiments with Animals

A wide variety of animals are being used to determine the initial retention, concentration in various organs, and biological half-life of specific isotopes. Observations are made on living and sacrificed animals to determine the amount of damage to various organs. Extrapolation is necessary to relate these data to man. More weight is given to the data the more nearly like man is the experimental animal.

5. Experience with Man

This is limited. But data on man will give the only completely reliable data. Accidents must be prevented whenever possible; but when they happen they are examined very carefully to obtain as much data as possible. Cases where Pu^{239} or Sr^{90} have been accidentally inhaled have given some of the best estimates of the biological half-lives of these isotopes in man. There have also been a few cases where volunteers have deliberately taken very small doses of ingested activity to determine initial retention and biological half-life.

Factors Determining Hazard of a Particular Isotope

Factors which help to determine the relative hazard of the various radioisotopes are as follows:

1. Quantity Available

As long as the only radioactivity to which man was exposed was that which occurs naturally, there was a quite limited hazard, particularly since there was little use for uranium. The most celebrated example of a hazard existing before the discovery of fission is the damage which occurred to painters of radium clock dials. With the advent of accelerators, reactors, and bombs, the quantities of radioactivity available to cause trouble has increased tremendously, and the numbers of isotopes have likewise increased. Even so, it is only the more commonly used radioisotopes and the longer-lived components of bomb debris which present major problems because of the quantity available.

2. Initial Body Retention

There are several avenues by which a radioisotope may invade the body. These include inhalation, ingestion, absorption through the skin, or into cuts or wounds. It is necessary to discover the path that the isotope takes once it has entered the body. Some of the isotopes are eliminated rapidly through the gastrointestinal tract. Some get into the blood stream from whence they may be eliminated rapidly, or they may be deposited rather preferentially into a specific organ or organs.

3. Radiosensitivity of Tissue

Some body tissues are more radiosensitive than others. Lymphatic tissue and bone marrow are among the most sensitive. Muscle and nerve tissue are less so. Therefore, plutonium, which concentrates in the most sensitive part of the bone, is more hazardous than uranium, which goes to relatively less sensitive parts of the body. Elements such as plutonium (and strontium) are often referred to as bone-seekers.

4. Size of Critical Organ

For a given quantity of isotope ingested, the hazard will be greater the smaller is the organ which that particular isotope seeks out, since the concentration of the radioisotope will be higher, as will be the dose delivered to the critical organ. This, for example, is part of the reason why iodine is a relatively hazardous element even though most of the iodine isotopes have short half-lives. Iodine concentrates rather completely in the thyroid gland, which weighs about 20 grams.

5. Essentiality of the Critical Organ

Not all body organs are equally important. Some can be removed and medicaments supplied to compensate for their reduced function. Others cannot. When damage is to the latter type (bone marrow, kidneys, eyes), the hazard becomes greater.

6. Biological Half-life

This is a rather loose term applied to the time needed for the body to get rid of one-half of the amount of an isotope which it has ingested. There is an initial period during which the elimination of an ingested radioisotope is quite rapid. This usually takes place before the radioisotope is translocated from the blood to a more permanent area such as the bone. The initial period is usually of the order of a few days to a few weeks. After that, the elimination rate becomes more nearly exponential and the application of the term biological half-life has more meaning. Those elements which deposit in portions of the body where the rate of turnover is very slow are the most dangerous. The hazard of radium, plutonium, and strontium is added to by this factor. On the other hand, there are radioisotopes such as carbon, sodium, and sulphur which have very short biological half-lives, thus reducing their hazards.

7. Radiological Half-life

The specific activity of a radioisotope, number of disintegrations per unit (time) (mass), varies inversely with the radioactive half-life. Therefore, very long-lived activities are not too much of a hazard because their specific activities are low. For instance, the mixture of uranium isotopes which occurs in nature does not present much of a radiation hazard (if the radioactive daughter elements are removed) because with the very long controlling half-life of U^{238} (4.5×10^9 yr) it requires 1.5×10^6 grams of this uranium isotopic mixture to make a curie of alpha activity. The maximum permissible amount of this mixture in the body is 5×10^{-3} μc. This corresponds to about 0.01 gram and it is unlikely that a person would get this much uranium in the body. If he did, it probably would result in a chemical hazard before any detrimental effects of radiation would become evident.

At the other extreme, even though short-lived radioisotopes have high specific activities, they are not much of a hazard because the radioactivities decay very rapidly and the exposures take place over relatively short periods of time. Such radioisotopes become serious hazards only when the exposure is maintained by continuous uptake.

It is the radioisotopes of intermediate half-life, from a few to 50 years, which present the greatest hazards, other factors being equal. Again on this count, strontium 90 with a 28-yr half-life is a particularly hazardous material.

8. Energy of the Radiation

The radiation hazard associated with radioisotopes deposited in the body is proportional to the average energy of disintegration weighted for the biological effectiveness of the radiation. Since the energy of disintegration may vary from many Mev to a few ev, the relative effect of the energy alone is quite large. The relative biological effectiveness of beta and gamma radiation is taken as unity, whereas that of alpha disintegrations is taken as twenty, that is, alpha particles are considered to be twenty times as damaging on an energy-absorption basis as the beta or gamma radiation, because of the high specific ionization.

Beta radiation is absorbed in the immediate vicinity of the atoms from which it is emitted, whereas the attenuation of gamma radiation of the same energy is much slower. For example, a beta emitter with a maximum energy of 2 Mev will be almost completely absorbed in one centimeter of tissue. Alpha radiation is even more localized; almost all the energy of the 6-Mev alpha from At^{211} is absorbed in the thyroid gland in which it localizes.

All of these factors must be considered together in determining permissible levels. Estimates are first made of the permissible body burden for each isotope, and from this and from estimates of the absorption of this isotope from air or water, calculations of the permissible concentrations in air and water are made. It should be recognized that there are great uncertainties in these data - they are not precise determinations. For this reason, they should be considered maxima and ideally the goal is to work as near to background levels as possible.

Use of Permissible Levels

The first compilation of permissible concentrations was put out in National Bureau of Standards Handbook 52.[6] The data were recently (June 1959) brought up to date and refined somewhat in National Bureau of Standards Handbook 69.[7] In the latter, figures are given both for a 40-hour week and for continuous exposure (168-hour week). The data on specific isotopes cover 65 pages in Handbook 69. They are particularly useful when only one, or at worst a few, isotopes are being used. In the case where a wide variety of materials are being used, for example mixed fission products, the analytical problem to determine the concentration of each component is so great as to make the use of individual levels almost out of the question. In this case permissible levels for unidentified radionuclides

must be used. In Handbook 52 this was stated rather simply as shown in Table 7. In Handbook 69 cognizance has been taken of the fact that a partial analysis may be made to show the absence of the more hazardous isotopes and thus a more relaxed level may be used. The current suggestions are given for water in Table 8 and for air in Table 9. Values only $\frac{1}{10}$ of these are recommended for use beyond the site control area on the theory that the general public, not being afforded the careful health monitoring programs used for nuclear workers, should be provided an additional safety factor.

Table 7

PROVISIONAL LEVELS OF PERMISSIBLE CONCENTRATION OF RADIOACTIVE CONTAMINANTS FOR USE BEYOND THE CONTROL AREA (OCT 1951)[a]

Medium in which contained	β or γ emitter (μc/ml)	α emitter (μc/ml)
Air	10^{-9}	5×10^{-12}
Water	10^{-7}	10^{-7}

[a]National Bureau of Standards Handbook 52, Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water, (March 1953).

Exposure of Man to Natural Background Radiation

One of the tests of adequacy of maximum permissible concentrations is the effect their use has on the existing natural radiation background. The natural background is due partly to the naturally occurring radioactive materials and partly to cosmic radiation. Although the background varies considerably from place to place, at sea level it is about 100 mr/year. Cosmic rays account for a little less than half of this and the contribution from this source approximately doubles for each 5000-ft increase in elevation.

Table 8

PROVISIONAL MAXIMUM PERMISSIBLE CONCENTRATION OF UNIDENTIFIED RADIONUCLIDES IN WATER $(MPCU)_w$[c]

Values that are applicable for occupational exposure (168 hr/wk) to any radionuclide or mixture of radionuclides.

Limitations	$\mu c/cc$ of water[b]
If Sr^{90}, I^{129}, Pb^{210}, Po^{210}, At^{211}, Ra^{223}, Ra^{224}, Ra^{226}, Ac^{227}, Ra^{228}, Th^{230}, Pa^{231}, Th^{232}, and natural Th are not present,[a] the continuous exposure level $(MPC)_w$ is	3×10^{-5}
If Sr^{90}, I^{129}, Pb^{210}, Po^{210}, Ra^{223}, Ra^{226}, Ra^{228}, Pa^{231}, and natural Th are not present,[a] the continuous exposure level $(MPC)_w$ is	2×10^{-5}
If Sr^{90}, Pb^{210}, Ra^{226}, and Ra^{228} are not present,[a] the continuous exposure level $(MPC)_w$ is	6×10^{-6}
If Ra^{226} and Ra^{228} are not present,[a] the continuous exposure level $(MPC)_w$ is	10^{-6}
In all cases the continuous occupational level $(MPC)_w$ is	10^{-7}

[a] In this case "not present" implies that the concentration of the radionuclide in water is small compared with the MPC value in Table 1, NBS Handbook 69.

[b] Use $\frac{1}{10}$ of these values for interim application in the neighborhood of an atomic energy plant.

[c] National Bureau of Standards, Handbook 69, Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air and in Water for Occupational Exposure, (June 1959).

Table 9

PROVISIONAL MAXIMUM PERMISSIBLE CONCENTRATION OF UNIDENTIFIED RADIONUCLIDES IN AIR $(MPCU)_a$[c]

Values that are applicable for occupational exposure (168 hr/wk) to any radionuclide or mixture of radionuclides.

Limitations	$\mu c/cc$ of air[b]
If there are no α emitters and if β emitters Sr^{90}, I^{129}, Pb^{210}, Ac^{227}, Ra^{228}, Pa^{230}, Pu^{241}, and Bk^{249} are not present,[a] the continuous exposure level $(MPC)_a$ is	10^{-9}
If there are no α emitters and if β emitters Pb^{210}, Ac^{227}, Ra^{228}, and Pu^{241} are not present,[a] the continuous exposure level $(MPC)_a$ is	10^{-10}
If there are no α emitters and if β emitter Ac^{227} is not present,[a] the continuous exposure level, $(MPC)_a$ is	10^{-11}
If Ac^{227}, Th^{230}, Pa^{231}, Th^{232}, natural Th, Pu^{239}, Pu^{238}, Pu^{240}, Pu^{242}, and Cf^{249} are not present,[a] the continuous exposure level $(MPC)_a$ is	10^{-12}
If Pa^{231}, natural Th, Pu^{239}, Pu^{240}, Pu^{242} and Cf^{249} are not present,[a] the continuous exposure level $(MPC)_a$ is	7×10^{-13}
In all cases the continuous occupational level $(MPC)_a$ is	4×10^{-13}

[a] In this case "not present" implies the concentration of the radionuclide in air is small compared with the MPC value in Table 1, NBS Handbook 69.

[b] Use $\frac{1}{10}$ of these values for interim application in the neighborhood of an atomic energy plant.

[c] National Bureau of Standards, Handbook 69, Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air and in Water for Occupational Exposure, (June 1959).

To date, the operation of the nuclear industry, with the exception of weapons testing, has had a negligible effect upon the natural background. This is indicated by the data presented in Table 10 which were put together for the National Academy of Science.[8]

Table 10

TOTAL DOSE FROM VARIOUS SOURCES TO PERSONS IN U.S. ACCUMULATED IN THIRTY YEARS[a]

Source	Average Dose,[b] rads	% of Total Dose
Cosmic rays	1.1	14
Earth	2.4	31
Internal	0.8	11
Subtotal from Natural sources	4.3	56
Medical examination	3.0	39
Fallout	0.4	5
Luminous dials	0.03	-
Nuclear reactors	-	-
Total	7.7	100

[a]I. Pullman, Physical Properties of Ionizing Radiation Which Affect the Population of the United States, National Academy of Science (April 18, 1956).

[b]gonad dose

PROBLEMS FOR CHAPTER 1

1. Assuming the continuous operation at full power of a 100-Mwt reactor using uranium with low enrichment, plot the buildup of the following nuclides during the first year of operation of the reactor and calculate their equilibrium quantities. Express in grams and in curies.

Group A	Group B
Sr^{90}	Cs^{137}
Pm^{147}	Ru^{106}
Ce^{144}	Zr^{95}
Sr^{89}	Ru^{103}

2. Assume that each of these isotopes is the first significant member of its chain or that all progenitors and the isotope itself are at equilibrium. At the end of a year the reactor of Problem 1 is shut down and the fuel removed. By means of the simplifying assumption that the eight nuclides calculated in Problem 1 are the only ones present, calculate the per cent of total fission product activity due to each of these at

a) time of discharge
b) after 100 days
c) after 3 years
d) after 30 years

3. If a sample of a waste effluent liquid reads

α not detectable
$\beta - \gamma$ 5 dpm/ml,

is it discardable directly?

4. If the sample of Problem 3 is analyzed for Sr^{90}, Pb^{210} and Ra^{228} and they are not detected, is it discardable? Assume that sufficient dilution exists in the immediate environment to take care of the factor of 0.1 for off-site exposure.

5. If a laboratory purchases the following isotopes

Isotope	MPC, μc/ml
Na^{24}	2×10^{-3}
P^{32}	2×10^{-4}
Co^{60}	5×10^{-4}
Ru^{106}	10^{-4}

and has no other sources of radioactivity, and if it is assumed that any activity in the waste comes equally from each of these isotopes, what is the MPC for discharge from this laboratory?

6. If the laboratory of Problem 5 began experimentation with Sr^{90}, using the same assumptions, what does this do to the MPC for the Laboratory? MPC for Sr^{90} is 10^{-6} μc/ml.

CHAPTER 2

ORIGIN AND MANAGEMENT OF WASTES

The nuclear energy industry encompasses a wide spectrum of effort starting with mining of ore and continuing through milling operations to enrich the ore, the preparation of various feed materials, fuel fabrication, reactor operation, and fuel processing. In addition, isotope enrichment may be practiced on part of the stream. Nor is this just a once-through process. At several points material is recycled back to an earlier stage. This complex of operations is often referred to as the nuclear fuel cycle. A schematic representation of the fuel cycle is shown in Figure 2, which gives particular emphasis to the wastes encountered. At each step some wastes are produced but the volumes, types of activity, and activity levels vary widely throughout.

In the mining and feed materials steps, only naturally occurring radioactive elements are encountered, and the activity levels in wastes are very low. During the operation of a reactor, fission and activation products and some transuranic isotopes are formed. Except in the case of an accident, the activation products form the normal reactor waste, and the concentrations, although higher than in mining and milling, are quite moderate. Of the four classes of radioactivity discussed in the first chapter, the fission products represent by far the greatest potential problem. In normal operation they do not enter a waste stream until the spent fuel is processed chemically. Therefore, the most serious problems in this field are associated with fuel reprocessing.

This chapter will deal not only with the origination of wastes at each step of the fuel cycle, but also with systems of waste management. This term means the overall system which is used to handle the total problem at any site. In succeeding chapters some of the specific unit operations and unit processes which are used within the framework of any of these overall systems will be discussed in detail.

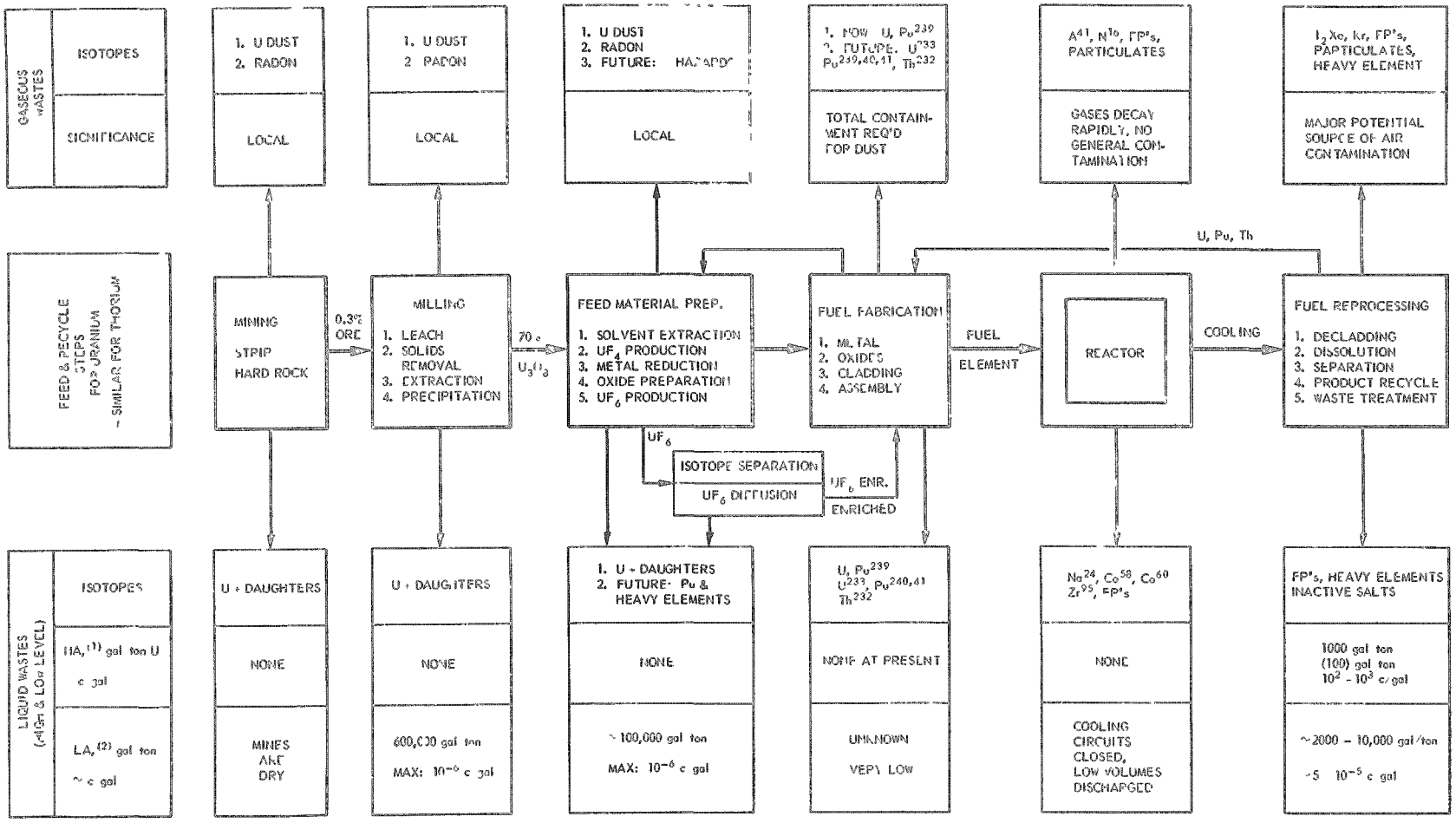
Mining

Uranium ore is taken from many mines, large and small, in the United States, at a current rate of about 18,000 dry short tons (2000 lb) per day and an average concentration of 0.25 per cent U_3O_8. Since 1943 about 125,000 tons of U_3O_8 have been procured from domestic and foreign sources.(10)

In the mines the most important radioactive waste products are radon and its short-lived daughters. Studies made in a number of mines have measured emanation rates for radon ranging from 5×10^{-7} to 2×10^{-5} curie/(min)(1000 cu ft) of mine volume.(11)

Figure 2

Wastes from the Nuclear Fuel Cycle[a]



(1) HIGH ACTIVITY WASTE ACTIVITY IS ORDER OF MAGNITUDE ESTIMATE, CURIES GAL
(2) LOW ACTIVITY WASTE,

[a]F. L. Culler, Nature of Radioactive Wastes, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 72, August, 1959.

As radon diffuses into the mine air, it is free of its daughters, but because the immediate daughters have very short half-lives they rapidly build back up again, thus increasing the hazard by several-fold. To maintain the radon concentration below the maximum permissible concentration for a 40-hr week exposure of 3×10^{-8} μc/ml, a complete change of air is required in the mine about every four minutes, with 500 cfm supplied per worker from an outlet not more than 30 ft from the working face.(12) Ventilation costs range from 15¢ to $1.35 per ton of ore, with an average of about 40¢ per ton. Disposal of these gaseous wastes to the atmosphere is no problem, since the air is exhausted at concentrations below 3×10^{-8} μc/cc and is further diluted with atmospheric air. The total quantity of radon discharged to the atmosphere from uranium mines is very small compared to that which is naturally produced from the surface of the earth.

Ventilation is also required in the mines to take care of diesel-operated equipment as well as dusts including uranium, silica, and arsenic.

Most mines on the Colorado plateau are dry or nearly so and pumping of mine water is negligible. However, in the Ambrosia Lake, New Mexico, area some mines are pumping as high as 600 gal/min. Pumping costs vary from nothing up to 75¢ per ton of ore, with an average of about 25¢ per ton of ore. The mine water contains only the normal amounts of radioactive materials for local ground water and its disposal is no problem.

Thus, the waste products of uranium mines are not of major significance in the overall waste disposal picture. The principal problems are the protection of miners from radon gas and radioactive dust. Adequate ventilation prevents harmful concentrations of these materials. The exhaust air discharged to the atmosphere presents no problem of general atmospheric contamination.

Milling

The ores are shipped from the mines by truck or rail to mills where they are processed by hydrometallurgical methods. The ores are first crushed in jaw or gyratory crushers and ground in rod and ball mills. The coarse crushing produces large amounts of dusts containing all the constituents of the ore. The primary health hazard in these areas is produced by silica dusts rather than by a radioactive material. A survey made on the Colorado plateau in which 124 samples were taken in mill crusher areas gave dust counts from less than 5×10^6 to over 10^8 particles per cu ft of air. Proper ventilation or wet crushing can control this hazard without difficulty.(11)

Ground ores which contain small amounts of lime are treated with strong sulphuric acid to dissolve the uranium (acid leach). Ores which contain large amounts of lime are treated with a solution of soda ash and

sodium bicarbonate (carbonate leach). Frequently, an oxidant is added to the leach solutions. Addition of 10 lb of manganese dioxide per ton of ore is typical. Acid consumption varies from 40 to 350 lb per ton of ore.

The various leach solutions are treated in different ways to recover the uranium. The bulk of the tailings is separated by clarification equipment and the remainder removed by filtration in most plants. Since filtration of the slimes is difficult in many cases, addition of filter aids and coagulants is common. In resin-in-pulp plants, the filtration step is omitted and the slimes are left in the solution.

The uranium is recovered from the solutions by ion exchange, solvent extraction or precipitation. In resin-in-pulp plants, coarse ion exchange resins in buckets are slowly moved up and down in the leach solution and slimes to absorb about 99.7 per cent of the uranium dissolved in the sulphate solution. The uranium is recovered from the ion exchange resins by an acidified nitrate solution containing about 50 grams of nitrate ion per liter at a pH of abour 1.2. Uranium is precipitated from this solution by magnesium oxide, filtered, repulped, dried on a drum drier, and barrelled for shipment. In solvent extraction plants, nitrate ion is added and the uranium removed from the filtered solution as uranyl nitrate hexahydrate (UNH) by an organic solvent, such as tributylphosphate dissolved in kerosene. The uranium is re-extracted from the organic solvent by very dilute nitric acid and recovered by precipitation. In the alkali plants, the leach solution is filtered and the uranium precipitated by addition of strong caustic. The precipitate is filtered, dried, and drummed for shipment.

In the chemical processing sections of these plants, the waste products are mainly gases from chemicals added to the slurries and solutions - such toxic compounds as hydrogen sulphide, arsine, or acid gases. In the final processing area, the primary wastes are uranium compounds which are disseminated into the workroom air as dusts. With proper ventilation neither represents much of a hazard.

Since the ore contains such a low concentration of uranium, practically all the material fed to the mills is discarded as wastes, slimes, and sands. In addition, 13,000,000 gal of water are used each day, most of which is discharged as plant effluent, carrying with it to the tailing ponds most of the activity delivered to the mills. The solid wastes, which are stored in the immediate areas of the concentration plants, usually contain about 1 mg of radium per ton which may be in a fairly soluble form that is leachable by surface water. At a plant using the resin-in-pulp process, 99.8 per cent of the radium remained undissolved and was effectively retained in the sands and slimes in the tailing ponds.[13] The radium content of the dried sands (440 tons/day) was about 1.5×10^{-4} $\mu c/g$ and of the slimes (80 tons/day) was about 2.5×10^{-3} $\mu c/g$. Other surveys have indicated that small amounts of both thorium and radium are dissolved in the leach solution. The alkali mills dissolve about 1 per cent of the radium while the acid mills dissolve up to 5 per cent. Individual mills vary considerably.

The 13,000,000 gal/day of liquid wastes contain, then, small amounts of radium, usually several orders of magnitude above the maximum permissible concentration for unlimited use (10^{-8} μc/ml). They also frequently contain added chemicals in amounts far above tolerance for dispersal in streams that are to be used as drinking water. Manganese and nitrate ions are of particular concern. They represent a considerable disposal problem to some of the Colorado plateau mills.

Table 11 presents measured and calculated tailings pond overflow compositions for several types of uranium mills in comparison with Utah specifications for drinking water. The data presented in this table were obtained on composite samples taken in the fall of 1957 and the spring of 1958.

Table 11

CHEMICAL COMPOSITIONS FROM TAILINGS POND OVERFLOWS FOR VARIOUS TYPES OF URANIUM MILLS[a]
(All values are ppm)

Constituent	State of Utah Specification	Acid RIP[b]	Alkaline Filtration[b]	Mixed Acid RIP	Acid Counter Current Decantation[c]	Acid CCD Solvent Ext EHPA[c]	Acid CCD Amine Solvent Ext
Chloride	250	190	81		615	105	110
Sulfate	250	3035	1760	1490	3195	3850	2910
Nitrate	20	2000	-	1830	0	0	-
Mg	125	290	<10	325	200	55	70
Cu	3	1	-	<0.1	4	<0.6	-
F	1.5	1	-	1.7	-	<5.6	4
B	1	0.15	-	0.2	-	-	-
Fe	0.3	20	<0.1	<0.1	0.1	0.1	220
Mn	0.3	55	-	170	370	395	30
Pb	0.1	0.35	-	0.2	-	-	-
As	0.05	0.1	-	0.06	-	-	-
U_3O_8	10	1.8	13	21	5	17	-
Dissolved solids	1000	6900	10,000	8900	5750	6300	4400
pH	5.9	5.5	10.5	6.9	-	-	2.6

[a]Condensed from Nature of Wastes from the Milling Industry, Statement for the Record, Hearings on Industrial Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 63, August 1959.

[b]Average of two mills

[c]Calculated

RIP Resin-in-pulp

CCD Counter current decantation

EHPA di-2-ethylhexyl phosphoric acid

Results indicate that:

1) manganese is the most serious consistent offender. Five pounds of manganese dioxide per ton will give a soluble manganese

concentration of 200 to 400 ppm, and the iron and manganese specifications will require a 1,000-fold dilution.

2) Nitrate is a serious problem in those mills using nitrate solutions.

3) Sulphate and total solids are offenders in most mills.

4) Magnesium exceeds specifications in acid-leach mills and chloride-elution mills.

5) Copper, fluoride, boron, lead, and arsenic may be problems

Table 12 gives data on the radioactivity contents of effluents from a number of different kinds of mills. In all cases the specification for radium-226 is exceeded in some cases by as much as a factor of 1000.(14) A U.S. Public Health Service survey conducted in 1958 and 1959 showed that the water in the Animas River in Colorado and New Mexico exceeded maximum permissible levels by from 40 to 160 per cent.(15) A number of the Colorado plateau mills have been ordered to eliminate the excess radioactivity in their effluents as a condition of continuance of their licenses.

Table 12

RADIUM CONCENTRATION IN URANIUM MILL TAILINGS POND EFFLUENTS[a]

Type of Plant	pH	U_3O_8, g/l	Radium 226, μc/ml
Specification[b]		0.002	4×10^{-9}
Acid Leach - Solvent Ext	1 5	0.01	4×10^{-6}
Acid Leach - RIP	3.3	0.0034	5.4×10^{-6}
Acid Leach - RIP	7.7	0.0003	3.3×10^{-7}
Acid Leach - RIP plus Alkaline Leach - Prec	6.9	0.02	8.1×10^{-8}
Alkaline Leach	9.9	0.009	2.0×10^{-6}
Acid Leach - CCD - Solvent Ext	2.0	0.002	2.7×10^{-6}

[a]Condensed from Nature of Wastes from the Milling Industry, Statement for the Record, Hearings on Industrial Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 69, August, 1959.

[b]Title 10, Code of Federal Regulations, Part 20.

Radium can be removed from acid tailings by two means. Simple neutralization removes radium by coprecipitation and adsorption on the precipitated material. Contact of the radium-bearing solution with natural barium sulfate (barite) removes radium by adsorption and possibly by inclusion in the barite. The neutralization-barite treatment is also effective in decontaminating alkaline tailings.(16)

Feed Materials

Ore concentrates from the mills contain about 70 per cent U_3O_8. Feed materials plants convert this and uranium which has been recylced from spent fuel reprocessing plants into pure uranium metal for the production reactors, uranium hexafluoride for the isotope separations plants, and uranium oxide for power reactors.

There are several feed materials plants in operation in the United States, some government and some privately owned. Large integrated government plants are located at Fernald, Ohio, and Weldon Springs, Missouri. Each is capable of handling thousands of tons of each of the different uranium compounds per year. Several private plants carry out one or more of the steps conducted in the Fernald and Weldon Springs plants. Of particular interest is the General Chemical plant which converts ore concentrates to uranium hexafluoride in fluidized beds. All the other plants purify the raw materials by solvent extraction.

At Weldon Springs and Fernald the ore concentrates are dissolved in nitric acid and the uranium purified by solvent extraction with ether or tributylphosphate dissolved in kerosene. The first solvent extraction column waste contains practically all the radium and other radioactive materials in the ore concentrate other than the uranium. Since the amount of radioactive materials is small and most of it is attached to solid particles, the wastes are run to settling tanks, a coagulant added, and they are filtered. The solid residues are combined with other solid wastes for burial, and the filtered liquid wastes are run to large settling basins and allowed to overflow into neighboring streams.

The purified product from solvent extraction, a solution of uranyl nitrate, is evaporated to liquid UNH and heated to 450 C to decompose the nitrate and form uranium trioxide. This oxide is reduced with hydrogen to uranium dioxide at 800 C. The dioxide, in turn, is converted to uranium tetrafluoride by treatment with hydrogen fluoride at 600 C. Part of the tetrafluoride is converted to metal by reduction with powdered magnesium in refractory-lined bombs. The remainder is made into uranium hexafluoride for the isotope separations plants.

The radiochemical nature of wastes from these operations is similar to that of the mill wastes - containing natural uranium and its decay daughters.

Gaseous wastes are insignificant. Very slight amounts of radon and solid particulate material are present. The ventilation air, particularly from the uranium reduction areas, contains considerable amounts of non-radioactive dusts which are removed by filters.

The solid wastes include the small amount of filter residues containing radium and large amounts of magnesium fluoride slags. The slags contain less uranium than the original ore. Some cleanup materials (rags, rejected equipment, etc.) contain sufficient radioactive material that these wastes, along with the slags, crucibles, and other debris, are placed in clay-lined pits or buried. Combustible wastes are buried or burned. Contaminated equipment containing uranium is frequently washed with nitric acid and the washings added to the feeds to the solvent extraction process for recovery.

For each ton of uranium processed, approximately 1,000 gal of liquid waste are produced. Gases from the reduction and hydrofluorination operations are scrubbed to remove chemical fumes and entrained uranium. Waste materials from uranium metal production are sometimes leached to remove contained uranium. The leach liquors are treated by the solvent extraction purification cycle or by ion exchange.

Isotope Concentration Plants

Some of the product of feed materials plants is diverted to the gaseous diffusion plants to obtain enriched uranium-235. The diffusion plants also use as a source of feed recycled uranium from production reactors such as those at Hanford, Savannah River, or the Idaho Chemical Processing Plant. The latter products are stored long enough to allow decay of uranium-237. They have a plutonium content of less than 10 parts per billion parts of uranium. Fission products are removed almost completely.

The diffusion plants start with pure uranium tetrafluoride, which is converted to uranium hexafluoride by treatment with elemental fluorine. By means of a series of gaseous diffusion stages an enriched product containing about $93\frac{1}{2}$ per cent uranium-235 is produced. For power reactor fuels, side streams containing lesser concentrations of uranium-235 are also produced. Depleted uranium tailings, containing about 0.2 per cent uranium-235, are stored. Some work is being done to devise uses for this material.

Very small amounts of wastes are produced and every effort is made to recover the maximum amount of uranium, particularly in the enriched section of the plant. Gaseous, liquid and solid wastes contain small amounts of uranium-235 and uranium-238 but negligible amounts of daughter products. The radioactive waste gases are principally uranium hexafluoride and dusts and mists from machinery and chemical operations.

The uranium hexafluoride feed, after conversion from uranium tetrafluoride, is passed through cold traps and condensed. Very small amounts of oxygen and nitrogen are present in this gas stream and these pass through the condensers carrying traces of the hexafluoride with them. These gases are exhausted at intervals to the atmosphere. About 0 7 mc/day is lost at Oak Ridge and 1.75 mc/day at Paducah.[16a] The building ventilation air also picks up very small amounts of uranium hexafluoride, but the concentration is so low that the exhausted air creates no hazard The highest value of radioactivity measured at the edges of the control area of the Paducah plant has been 1×10^{-13} μc/cc, which may be compared with the maximum allowable concentration for insoluble uranium (continuous exposure outside the controlled area) of 2×10^{-12} μc/cc.

Liquid wastes produced are almost entirely as a result of decontamination and recovery procedures. Nitric acid is used to remove uranium hexafluoride from surfaces of equipment The resulting solutions are stripped of uranium by solvent extraction. The wastes consist of the aqueous column raffinate, spent acid, and rinse water. These wastes are passed through settling basins and discharged to local streams.

The solid wastes include contaminated paper, floor sweeping compounds, lubricants, containers, activated alumina, etc. These are generally disposed of to land fills.

Fuel Element Manufacture

Before they can be used as fuel in a reactor, the products of the feed materials plants must be made up into fuel elements of varying degrees of complexity. Fuel elements are made from uranium, its alloys, and uranium oxide. The uranium may be natural or enriched. Operations involved include melting, casting, rolling, machining, and metal cleaning. Varying quantities of uranium-bearing wastes are produced in the form of alloy scrap, liquids, contaminated pieces of metal, paper, rags, and a certain amount of airborne dust.

In handling natural or slightly enriched uranium, gaseous wastes are almost negligible. The regular building ventilation is adequate throughout most of the plant It is necessary to check the air around melting and heat-treating furnaces, hot rolling mills and extrusion processes but nothing more serious than the use of face masks is required under normal conditions. When highly enriched uranium is used, good ventilation, use of face masks, and the usual health-physics surveys are required. The exhaust air is filtered through high efficiency AEC filters The dusts so collected usually do not warrant recovery

Liquid wastes formed in surface cleaning operations are usually very dilute A typical waste after neutralization contains 0 005 gram of uranium per liter, with calcium, iron and sodium ions present in large quantities. These wastes may be processed by ion exchange or by precipitation of uranium followed by filtration, or they may be monitored, diluted and discharged to the river The volume of such waste at one plant was 5,000 gal per year.(17)

Solid wastes may be divided into four kinds of material

1) Turnings, cuttings, trimmings, powder recovered from grinding operations, and rejected materials which contain a high percentage of fuel.
2) Crucibles and molds used in casting operations
3) Contaminated equipment
4) Contaminated rags, cleaning materials, clothing, etc

Each of these four categories is handled separately.

The turnings, cuttings, trimmings, etc., may contain 10 to 50 per cent of the total fuel and cannot, therefore, be treated as wastes even when natural uranium is being processed. They must be recycled to a feed materials plant, where they are dissolved and recovered by solvent extraction.

Crucibles and molds used for natural or slightly enriched uranium are added to the general solid wastes and shipped to one of the permanent burial grounds. Those used for enriched fuel must be crushed and the uranium leached out with acid and recovered.

Some contaminated equipment is decontaminated. Some is simply buried. Used steel and copper extrusion cladding is etched with nitric acid for uranium recovery before burial. At one plant, 7,000 lb per year of slightly contaminated copper and steel were produced by hot-working operations.

Cleaning materials which contain very small amounts of natural uranium may be buried or combined with solid scrap for shipment to a permanent burial ground Materials containing even small amounts of enriched fuel are sent to a recovery plant

As burnup of fuels becomes higher and as fuels are recycled through reactors several times, and as the use of plutonium for power reactor fuel becomes more widespread, the concentration of higher isotopes can be expected to build up to a point where the contact metallurgical operations now practiced may become impractical Under these circumstances, incomplete decontamination processes (such as the pyrometallurgical processes now

being developed for the EBR-II) may become practical. If this is the case, the entire gamut of metallurgical operations will have to be done remotely, and the wastes produced by this portion of the fuel cycle will become radically different - more nearly akin to those now produced by chemical processing. At the present time, however, essentially all fuel element fabrication is done as described and the waste problems are relatively minor.

Reactor Operation

Radioactive wastes stemming from reactor operations are produced by two general processes: (1) fission and (2) neutron activation of the coolant, its impurities, and structural components to form activation products, or of the fuel itself to form transuranic elements. A reactor which has operated for any length of time represents a most considerable inventory of all of these products. In case of an accident, it is conceivable that hazardous quantities of radioactivity could be released to the surrounding environment. Such a possibility is a proper consideration in the hazards evaluation of any reactor but not in the operation of its waste disposal system, which is set up for handling only those small quantities of activity which may be expected to escape from the system during normal operation.

Reactors are classified into two broad categories: heterogeneous and homogeneous. Heterogeneous reactors are typified by a core lattice of solid fuel elements clad with a structural material (aluminum, zirconium, stainless steel) to protect the fuel from the coolant (water, liquid metal, organic, air) and to contain fission products. So long as the integrity of the cladding is maintained, fission products are not released into the coolant. However, in all operating heterogeneous reactors occasional failure of fuel elements can be expected as a normal occurrence. The failed fuel elements then permit the escape of small amounts of fission products into the coolant stream, where they are added to the coolant activation products, increasing somewhat the concentration of radioactivity. These reactors feature elaborate electronic devices designed to detect clad ruptures so that the failed element may be removed before significant amounts of fission products are released into the coolant stream Bypass coolant purification systems provided for the removal of corrosion and impurity activities are also capable of retaining the fission products.

Homogeneous reactors have fuel dispersed in the coolant-moderator which is cycled through an integrated primary and secondary heat-removal system. The entire coolant system, then, is the primary container of both induced activities and fission products. In this case, the induced activities are insignificant in comparison with the fission products. The complete system is enclosed in a secondary container to prevent the release of these highly radioactive materials in the event of the system failure. The waste

problems connected with a homogeneous reactor are more analogous to those of a fuel-processing plant than to those of a heterogeneous reactor. Homogeneous reactors have not yet become important in the commercial aspects of the nuclear power industry.

Reactor waste problems vary somewhat with reactor type. Therefore, a number of examples will be described. Operating heterogeneous reactors are at present predominantly water cooled. During operation the major source of radioactive wastes is the short-lived isotopes produced by nuclear reactions in the bulk coolant (see Table 3, page 15). After a short shutdown period, however, these decay away and the impurity activation products are left as the major contributor of activity. To these induced activities may be added fission-product activities introduced into the coolant by fuel-element failure. These fission products seldom markedly increase the difficulty of the waste problems. The activity level of coolant is, in any case, low.

Hanford Production Reactors[18]

In the Hanford reactors the heat of the fission process is removed by passing Columbia River water through the reactor tubes. The reactor locations are dictated by the combined needs of access to river water and relative isolation from populous areas. The latter feature was required for protection only in the extremely improbable event of a major operational accident. It should be remembered in this connection that the site selection was made before anyone had successfully operated a reactor. At the present time reactor sites are routinely considered for much more populous areas.

The Hanford production reactors are designed for low-pressure, single-pass water cooling. Since the amount of activation products which get into the effluent is controlled by the concentration of stable isotopes in the cooling water, close control of water quality is maintained. The raw river water is treated by coagulation with alum, settling, filtration through beds of coal and sand, chlorination, addition of sodium bichromate for corrosion control, and pH adjustment.

During their passage through the reactor, traces of minerals in the coolant water are made radioactive as a result of neutron bombardment. Small quantities of fission products arise from natural uranium in the river water and from occasional ruptures of fuel elements. The effluent leaving the reactor thus contains a complex mixture of many different radioisotopes. The effluent flows by gravity to retention tanks where it can be held up from one to three hours - long enough for radioactive decay to reduce the gross activity by 50 to 70 per cent. Under normal circumstances the discharge is then put into the river The effluent enters the river

through large pipes buried in the river bottom and is immediately mixed with the large volume of water flowing in or near the main channel. There is, however, a tendency for higher concentrations of the effluent to remain in midstream for some distance.

At the time of discharge, approximately 90 per cent of the gross activity consists of Mn^{56}, Cu^{64}, Na^{24}, Cr^{51}, Np^{239}, As^{76}, and Si^{31}. It takes 24 hours for the effluent to reach Pasco, Washington, the first point of substantial use. By this time radioactive decay has further reduced the gross activity to less than 10 per cent of the level at which it entered the basin, and the isotopes of greatest abundance are Cu^{64}, Na^{24}, Cr^{51}, Np^{239}, and As^{76}.

The retention basins may also be used to intercept effluent which has an unusually high radioactive content. Such situations arise from occasional ruptures of fuel elements and from "purges" of corrosion product film from the tubes. In some cases abnormally contaminated effluent water is discharged to trenches along the river bank where it seeps into the ground and is naturally filtered before it gets into the river.

Monitoring of the effluent is an important aspect in the management of the reactor wastes and is accomplished at several different points. Of the total number of isotopes detected in the effluent, 24 are of sufficient interest that their concentrations are measured on a routine basis. The first indication of the radioactive composition of the effluent is obtained from samples which originate from various parts of each reactor. The signals from instruments which scan the samples usually provide the basis for shutting down the reactor when there is a fuel-element rupture. This system works quite well and it is estimated that ruptures which occurred during 1958 contributed only about 5 per cent of the radiation exposure received by people in the Pasco-Kennewick area from the drinking of Columbia River water.

The effluent is next monitored at the time it leaves the retention basin. As an added precaution to assure that reactor operations do not affect fish, a continuous sample of the effluent being discharged to the river is pumped to an aquatic biology laboratory and its toxicity tested with live fish.

Gases and aerosols form a minor source of radioactive waste from reactor operation. Ventilation air picks up some contaminated dust from portions of the building which are normally unoccupied and some is added by gas which leaks from the reactor shielding. Release of these materials through high stacks to the atmosphere has produced no distinguishable contamination levels in the air or on the ground outside of the restricted area.

Solid wastes, such as contaminated paper, boards, tools, construction items, and aluminum spacers used to center the fuel charge are

buried in shallow trenches in the reactor area. The bottoms of the trenches are well above the water table and the rainfall is low enough so that percolation through the deposits is practically nonexistent.

Materials Testing Reactor (MTR)[19]

The MTR at Idaho uses a recirculating water system The feed water is pretreated by ion exchange to reduce impurities to less than one part per million. The total flow through the core and reflector is about 22,000 gallons per minute. Sources of liquid waste are (1) a continuous bleedoff of demineralized primary coolant, (2) canal overflows, (3) waste water from test loops, (4) waste streams from various laboratories, and (5) primary coolant purges at the time of shutdown.

To prevent the gradual buildup of activity in the primary cooling water, not only is a side stream put through a cation resin bed, but a portion of the coolant is bled off and replaced with fresh demineralized water. The water removed from the system is fed to one of two 350,000-gallon concrete retention basins. The average holdup time in these basins is 150 hours. The overflow from the basins is returned to the ground by means of a leaching pond.

During the first 2 years of operation of this reactor, very minor fission-product activities were released to the cooling system. The activity near the piping was about 50 to 100 mr per hour during operation and 10 to 20 mr per hour during shutdown. Corrosion product and other activities identified included aluminum-28, sodium-24, cobalt-60, and trace amounts of nine other isotopes. In 1954 a series of fuel-element ruptures occurred, which gradually increased the activity levels to as high as 3 r per hour. It was at this time that the bypass cation resin bed was installed and operated at a rate of 1,000 gal per minute on the bulk coolant. The activity decreased slowly to its original value. This experience indicates that while fuel ruptures can produce an increase in coolant activity, normal levels can be restored with the aid of relatively simple treatment procedures.

The major source of gaseous activity from the MTR is the cooling air which removes the heat from the thermal shield and graphite. This air flows through the reactor at the rate of 24,000 cfm and in doing so argon-41 is formed. This stream is monitored and discharged through a 250-foot stack

Solids requiring disposal are:

1) fuel element and control rod end boxes which are removed prior to shipment to the chemical processing plant.
2) parts of experimental equipment, and
3) spent ion exchange resin.

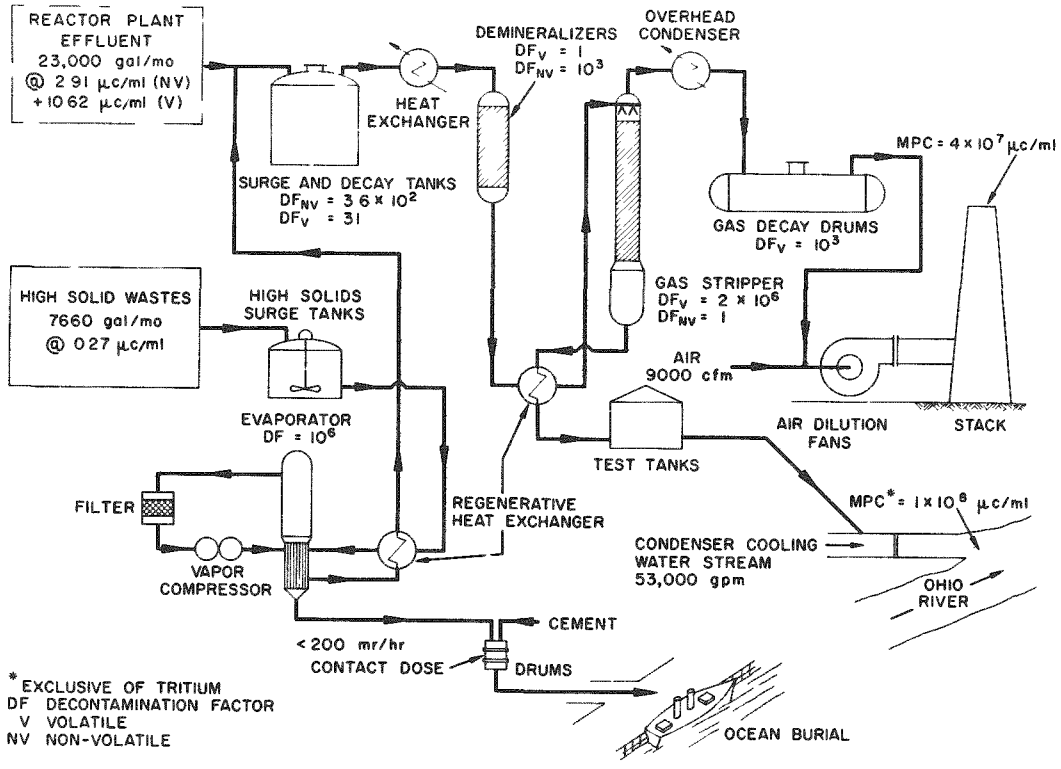
The first two of these categories are stored for a while in the reactor canal to permit some decay. They are then removed from the canal in large garbage cans and shipped to the burial ground in a lightly shielded two-wheel trailer. When the resin is exhausted, it is sluiced into steel tanks, sealed, and buried with the other solid wastes.

Shippingport (PWR)[20]

The reactor at Shippingport is the United States' first full-scale nuclear power station. Demonstration of its integrated waste disposal facilities (see Figure 3) has therefore been an important part of its operation. The station is in a populated area and on the heavily used Ohio River. Unlike the Hanford reactors, which use a once-through water system, the PWR uses recirculated pressurized water as primary reactor coolant.

Figure 3

Radioactive Waste Disposal Facilities,
Shippingport Atomic Power Station[a]



[a]Bethel, A. L. et al., Presentation on Shippingport Atomic Power Station (PWR) Waste Disposal Facilities, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 656, August, 1959.

Activity builds up in this coolant due to

1) activation of corrosion products,

2) formation of tritium from lithium hydroxide used to raise the pH of the water for corrosion control, and

3) from fission products introduced by fuel ruptures.

To limit the buildup of these contaminants, the coolant is continuously purified by circulating a portion through a bypass demineralizer. When saturated, the resin is not regenerated but is transported to buried storage tanks on the site. The volume of reactor waste effluents is about 23,000 gal/month and it contains about 3 μc/ml of nonvolatile activity and about 10 μc/ml of gaseous activity. The reactor plant effluents are first transported to type 347 stainless steel holdup tanks known as "surge and decay tanks." They are designed to provide storage volume sufficient for 45 days if necessary. From here the liquid is cooled in a heat exchanger and then passed through a series of four ion exchangers. Upon leaving the ion exchangers, the waste will have had its nonvolatile activity reduced sufficiently for discharge, but it may still contain a higher level of volatile activity than is desired. Volatiles are stripped from the waste using steam and the liquid then goes to one of two 5,000-gal test tanks. When satisfactory analyses have been obtained, the waste is discharged to the river. The volatile radioactivity which was removed in the gas stripper is sent to a circulating flush gas system. At infrequent intervals gas is discharged from this system into one of four decay drums where it is stored at 50 psig. The gas is stored for a sufficient period of time (up to 60 days) for the radioactivity levels to decrease to a point where subsequent controlled discharge and dilution with 9,000 cfm of air will permit discharge of the gases at less than $\frac{1}{10}$ MPC. This treatment process reduces the activity of the liquid to about 5×10^{-5} μc/ml for the nonvolatile and 1×10^{-8} μc/ml for the volatile activity.

Combustible solid wastes are burned in an incinerator, the flue gases of which are scrubbed and filtered before discharge. Noncombustible wastes consisting of resins, residue ash from the incinerator, solids from strainers in the pipe lines, and contaminated equipment are stored on site or embedded in concrete and disposed of at sea.

Naval Reactors[21]

All nuclear-propelled U.S. Naval ships now planned, in construction, or in operation are powered by pressurized-water reactors. The reactor coolant passes through heat exchangers which transfer the heat to a steam system, which is used as a source of power for the propulsion plant as well as for auxiliary machinery. Since these are mobile reactors, waste problems, although similar to those of stationary pressurized-water reactors, have some special considerations. The principal source of radioactive waste is the reactor-coolant water, which contains small quantities

of activated impurities. Some of the reactor-coolant water is discharged during plant startup due to expansion as the reactor plant is brought up to operating temperature. This normally happens a few times a month on each ship, and the quantity of cooling water discharged on each averages about 500 gallons per month. The radioactive species in the coolant water and the dumping tolerances established are shown in Table 13.

Table 13

MEASURED ACTIVITIES OF COOLANT FROM U.S. NAVAL NUCLEAR-POWERED SHIPS[a]

Nuclide	Half-life	Measured Activities of Coolant		Dumping Tolerance, μc/ml
		Maximum, μc/ml	Average, μc/ml	
Mn^{56}	2.5 h	9.3×10^{-2}	2.2×10^{-2}	15
Co^{60}	5.2 y	2.5×10^{-2}	5.7×10^{-6}	2
Fe^{59}	45 d	2.8×10^{-3}	1.5×10^{-4}	1×10^{-2}
Ni^{65}	2.56 h	1.3×10^{-3}	1.6×10^{-4}	1.9
Cr^{51}	27 d	5.5×10^{-3}	1.0×10^{-5}	50
Na^{24}	15 h	2.0×10^{-2}	8.0×10^{-5}	3×10^{-1}
Cu^{64}	12.8 h	9.1×10^{-3}	1.5×10^{-5}	8
Ta^{182}	112 d	5.6×10^{-2}	7.3×10^{-3}	10
F^{18}	1.87 h	6.8×10^{-2}	1.2×10^{-2}	90
W^{187}	24 h	9.0×10^{-3}	3.3×10^{-4}	9×10^{-2}
Gross activity measured 15 min after sampling		1.5×10^{-1}	5.0×10^{-2}	3
Gross activity measured 120 h after sampling		3.6×10^{-2}	3.1×10^{-3}	1×10^{-1}

[a]T. J. Iltis and M. E. Miles, Radioactive Waste Disposal from U.S. Naval Nuclear Powered Ships, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal. JCAE, 86th Congress of the United States, Vol 1, page 920, August, 1959

The basic criterion adopted for disposal of coolant is that disposal should not increase the average concentration of radionuclides in the surrounding environment by more than one-tenth of the maximum permissible concentrations for continuous exposure as listed in National Bureau of Standards Handbook 52.[6] On this basis the Navy has adopted the instructions that for discharge in port, gross activity of reactor coolant must be less than 3 $\mu c/ml$ and the fission product iodine-131 must be below 10^{-3} $\mu c/ml$. In determining these dumping tolerances it was assumed that discharged waste will almost immediately be diluted in the harbor by a factor of at least 1,000. This requires mixing with a volume of water approximately equal to the displacement of the ship. Actual measurements indicate that the immediate dilution factor is about 100,000.

There are other sources of radioactive wastes derived from the operation of naval nuclear-powered plants which require only infrequent waste disposal considerations. These include the disposal of

1) the ion exchange resin that is used to purify the coolant water of the reactor plant,
2) reactor feed water,
3) solid wastes from maintenance operations, and
4) special wastes from laundry or decontamination operations.

The ion exchange resin becomes exhausted and must be replaced approximately every six months. Table 14 shows the radioactivity associated with the spent resin. If resin replacement is necessary in port, the resin is dumped to a disposable catch tank which is subsequently sealed and buried by land or sea. Resin discharge at sea can take account of the great dilution available in the ocean. When dumped overboard, the resin will sink and as it sinks the radioactive ions in the resin are rapidly replaced by ions of the sea water. Thus, within a few minutes the radioactivity is transferred from the resin to the sea water in the wake of the ship, where it will readily be dispersed. Assuming, conservatively, that the wake is no larger than the path of the ship itself, the distributed activity from the resin results in a gross concentration in the ship's wake of less than 10^{-3} $\mu c/cc$. Subsequent action of wind, wave and current will rapidly decrease these concentrations. Therefore, Navy instructions allow resin disposal in the open ocean. However, in order to avoid any possibility of having such discharges increase the radioactivity to which people are exposed, there are the following restrictions:

1. the ship must be more than 12 miles from shore;
2. the ship must be underway;
3. no other ships shall be within 3 miles; and
4. the ship must not be in a known fishing area.

Table 14

RADIOACTIVITY OF SPENT ION EXCHANGE RESIN[a]

Nuclide	Half-life	Maximum Activity,[b] curies
Co^{60}	5.2 y	10
Co^{58}	71 d	0.5
Fe^{59}	45 d	0.5
Cr^{51}	27 d	0.3
Mn^{54}	300 d	0.2
Hf^{175}	70 d	1
Total		12.5

[a]T. J. Iltis and M. E. Miles, Radioactive Waste Disposal from U.S. Naval Nuclear Powered Ships, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 940, August, 1959.

[b]Maximum radioactivity expected based on measurements from operating plants.

Some attenuation of the radiation emanating from the reactor core is accomplished by using water in a shield tank around the reactor. This shield water will seldom, if ever, be dumped during the life of a ship. Potassium chromate (0.2 per cent) is used in this water as a corrosion inhibitor. Neutron activation of potassium chromate and impurities in the shield water produces small concentrations of radionuclides. These concentrations are well below dumping tolerance and there are no waste disposal restrictions necessary on disposing of this shield tank water.

Solid wastes from nuclear ships result primarily from maintenance operations. Such materials include metal scrap, pieces of insulation, rags, sheet plastic, and paper. These solid wastes are given by the ships to shore or tender facilities for subsequent packaging and burial.

Two other operations associated with reactor plants require disposal of radioactive liquids: decontamination of radioactive tools and equipment, and laundering of radioactive clothing which may be performed on some ships. Laundering wastes are held up for monitoring and treatment by ion exchange if necessary, but they ordinarily may be dumped under the same restrictions as the reactor coolant water.

Boiling Water Reactors[22]

Direct-cycle boiling water reactors, such as EBWR, and the Dresden and Vallecitos reactors, generate the same kinds of wastes as

pressurized water reactors with some added importance to gaseous wastes. Predominant gaseous activities from the EBWR, for instance are xenon-138 and krypton-88. The normal daily discharge rates to the atmosphere at 20-megawatt operation of EBWR are 0.2 to 0.4 curie of xenon-138 and 0.005 to 0.01 curie of krypton-88. During a test in which an oxide fuel element was deliberately defected, the discharge rates of these isotopes rose to 1 and 0.08 curie per day, respectively.

Each time the EBWR reactor vessel is opened, the system is filled with water and given a hydrostatic test at the conclusion of which the excess water must be bled off. These operations have produced about 30,000 gallons per year of liquid waste with a specific activity of about 5×10^{-5} μc/ml. There is also produced about 100 cu ft per year of solid waste containing about 100 millicuries of activity per cu ft. Ten to twenty per cent of this is represented by spent ion exchange resin beds which read about 10 r/hr on contact.

Heavy Water Reactors[23]

Recirculated heavy water is used as a moderator and/or coolant in such reactors as CP-5 at Argonne, NRX at Chalk River, Ontario, and the Savannah River reactors. The economics of such systems dictates indirect heat exchange, and thus problems related to the disposal of radioactivity are alleviated, since the cooling water is not exposed to the neutron flux in the reactors and, therefore, does not build up induced radioactivity. The fuel is jacketed with aluminum or other corrosion-resistant metal to reduce the possibility of spreading fission products, not only in the reactor, but also during handling after discharge.

At Savannah the moderator-coolant is cooled by river water in a shell-and-tube heat exchanger. The river water is monitored to detect moderator leakage and, also, to ensure that radioactive materials accompanying a leak are not discharged directly to the river.

In addition to nitrogen-16 and-17 and oxygen-19 formed from the oxygen, tritium is formed in the coolant by irradiation of deuterium $H^2(n,p)H^3$. The presence of the first three of these requires the use of shielding for the circulation equipment outside the reactor during operation, but during a shutdown they decay out very rapidly. Tritium, with a half-life of about 12 years, continues to build up as the reactor continues operation and eventually additional precautions may be necessary. The heavy water also picks up activity from the activation of corrosion products. Suspended solids and dissolved metals are removed by a filter-ion exchange system. Spent filters and resins are removed and transported to the burial ground. Any leakage of the heavy water will release tritium into the atmosphere. The reactor buildings were designed with adequate ventilation to dilute such activity and discharge it through a 200-foot stack. To date, there has been no occasion where the tritium concentration either on or off site has approached the permissible limit for continued exposure.

The irradiated fuel discharged from the reactor is removed to a storage basin where the short-lived isotopes are permitted to decay to reduce the problems encountered in further processing. Following decay cooling, extraneous housing tubes, etc., are removed, baled and sent to the burial ground. The clad fuel is shipped in shielded containers to the separations areas.

Liquid-metal-cooled Reactors

Liquid metals, such as sodium and NaK, are employed as coolants in reactors such as EBR-I, EBR-II, the land-based prototype of the Submarine Intermediate Reactor (SIG), and the sodium-cooled, graphite-moderated reactor. The characteristics of the principal activities found in liquid metal coolants were given in Table 3 of Chapter 1. The radiation from activated impurities in commercial sodium is usually negligible compared with that from the bulk sodium. The radioactivity associated with the sodium decays by a factor of ten every two days during a shutdown.

Disposal of waste sodium or NaK is complicated not so much by the radioactivity content as by the vigorous chemical action of the metals with water, oxygen, and carbon dioxide. The several possible methods of disposal all aim at forming water-soluble compounds of the metals. Water, steam, alcohols, and liquid ammonia are used.

Air-cooled Reactors

Two reasonably large air-cooled research reactors are in operation, one at Oak Ridge and one at Brookhaven. The principal radiation associated with air cooling of reactors is argon-41 formed from argon-40 in the atmosphere. At the Brookhaven reactor, for instance, some 4,000 curies of argon-41 are discharged through the stack each day.[22] This quantity, while troublesome insofar as instrumentation is concerned, causes no problem in regard to the public because of local meteorological conditions and the low radiotoxicity of the argon.

Organic-moderated Reactors

The Organic Moderated Reactor, OMRE, uses polyphenyls as the moderator and coolant. The wastes which accumulate during normal operation are primarily decomposition products resulting from the irradiation of the organic material. These products contain very little activity and they are readily disposed of by burning.

Homogeneous Reactors

Maintenance of the coolants of homogeneous reactors requires the separation of fission products from the fuel-coolant mixture external to the reactor. The waste problems will be equivalent to those experienced

in the operation of a chemical processing plant since the entire system complex becomes contaminated with deposited activities to a degree many orders of magnitude greater than in heterogeneous reactors. In an aqueous homogeneous reactor, krypton, xenon, and iodine are continuously removed from the coolant along with hydrogen, oxygen, and steam. Following recombination of the hydrogen and oxygen, the fission products are physically adsorbed on activated charcoal. The effluent gases are discharged through a stack to the atmosphere.

Fuel Processing

Irradiated reactor fuels are taken from the reactor, cooled for about 90 days to allow decay of short-lived nuclides, and then chemically reprocessed to reclaim the unburned nuclear fuel and recover bred fissionable materials.

During processing, the fuel jackets are first dissolved, after which the irradiated fuel elements are dissolved in acid. Plutonium and uranium are then separated from the highly radioactive fission products and purified by solvent extraction. Finally, the plutonium and uranium are processed separately to their required final forms.

The magnitude of waste problems associated with chemical processing plants far outweighs those of all other parts of the fuel cycle. During the dissolving and feed-preparation steps, radioxenon and radioiodine are discharged. The aqueous waste from the first solvent extraction cycle contains more than 99.9 per cent of the fission products, and this stream represents the most difficult part of the problem. Approximately 1,000 gallons of this waste are formed per ton of uranium processed. Typical chemical compositions of these wastes are shown in Table 15, and radiochemical compositions for such wastes concentrated ten times before storage are shown in Table 16.

These wastes are evaporated to smaller volumes and are frequently neutralized before being stored in underground steel and concrete tanks. After 100 days of cooling, high-level wastes may contain as much as 1,000 curies per gallon and emit heat at a rate as high as 20 Btu/(hr)(gal). The total activity level decreases, and the contribution of specific fission species changes, with time.

High-level wastes now in storage at the major processing plants amount to approximately 65 million gallons. There is a total tank capacity of about 110 million gallons. In addition to the high-level wastes from chemical processing, there is also produced a much larger volume of low-level liquid wastes at fuel-processing plants. These derive from second- and third-cycle solvent extraction operations, fuel dejacketing, laboratory and laundry operations, and cell decontamination

Table 15

TYPICAL COMPOSITIONS OF WASTES FROM THE REPROCESSING OF NUCLEAR FUELS[a]

Type of Waste:	High Level			Decontamination	Decladding
Process:	Redox	Purex	$BiPO_4$	$BiPO_4$	Caustic
Composition, molarity					
Na^+	6.4	2.4	4.1	4.1	4.9
Bi^{+++}			trace	trace	-
Fe^{+++}	trace	trace	-	trace	-
U	trace	trace	-	trace	-
AlO_2^-	1.4	-	-	trace	1.2
OH^-	0.8	0.1	-	0.2	1.0
NO_3^-	4.2	2.1	0.6	2.5	0.6
NO_2^-	-	-	-	-	0.9
$SO_4^=$	trace	trace	0.25	0.1	-
$CO_3^=$	-	0.1	1.0	-	-
$CrO_3^=$	trace	trace	-	-	-
SiO_3	-	-	-	-	trace
$HPO_4^=$	-	-	0.27	0.5	-
Specific Gravity					
	1.32	1.10	1.25	1.15	1.18
Fission Products, approximate per cent contained					
	100	100	90	10	0.1

[a]R. E. Tomlinson, Storage of High-level Fission Product Wastes, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 282, August, 1959.

At the present time all major chemical processing sites are located in remote or semiremote areas. It is not yet clear whether this will always have to remain the case. The major processing sites are Hanford in the State of Washington, Savannah River in South Carolina, and the Idaho Chemical Processing Plant in Arco, Idaho. Because of the importance of this phase of waste handling, the operations at each of these major sites will be briefly described.

Table 16

CONCENTRATION OF SIGNIFICANT RADIOISOTOPES IN THE LIQUID WASTE FROM IRRADIATED URANIUM[c]

Nuclide	Half-life, Years	Concentration,[a] $\mu c/cc$	MPC,[b] $\mu c/cc$ of H_2O	Number of Times Above MPC
Sr^{89}	0.148	1.6×10^{4}	7×10^{-5}	2×10^{8}
Sr^{90}	28	2.3×10^{4}	8×10^{-7}	3×10^{10}
Y^{91}	0.159	2.3×10^{5}	3×10^{-4}	8×10^{8}
Zr^{95}	0.178	2.7×10^{5}	6×10^{-4}	4×10^{8}
Ru^{103}	0.110	3.5×10^{4}	9×10^{-4}	4×10^{7}
Ru^{106}	1.0	7.2×10^{4}	1×10^{-4}	7×10^{8}
Te^{129}	0.090	3.1×10^{3}	2×10^{-4}	2×10^{7}
Cs^{137}	30	2×10^{4}	2×10^{-3}	1×10^{7}
Ba^{140}	0.035	5.2×10^{3}	3×10^{-4}	2×10^{7}
Ce^{141}	0.088	9×10^{4}	4×10^{-1}	2×10^{5}
Pr^{143}	0.038	5.5×10^{3}	5×10^{-4}	1×10^{7}
Ce^{144}	0.78	6.9×10^{5}	1×10^{-4}	7×10^{9}
Nd^{147}	0.032	1.6×10^{3}	6×10^{-4}	3×10^{6}
Pm^{147}	2.6	9×10^{4}	2×10^{-3}	4×10^{7}
Sm^{151}	80	5.6×10^{2}	8×10^{-3}	7×10^{4}

[a]Assumes the fission products from one ton of irradiated uranium (2500 MWD/T at 5 MW/T) are segregated in 100 gallons of water ninety days after reactor discharge.

[b]The maximum permissible concentration (MPC) for each nuclide in potable water is given for each parent in equilibrium with its radioactive daughters. These values were taken from HW-25457, Rev. 1, Manual of Radiation Protection Standards, Hanford Atomic Products Operation (May 1, 1957).

[c]R. E. Tomlinson, Storage of High-level Fission Product Wastes, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 288, August, 1959.

Hanford[18]

The gaseous effluents from the separation plants are exhausted to the atmosphere through 200-ft concrete stacks having stainless steel liners. Shortly after startup of the two original bismuth phosphate plants in 1945, radioactive iodine-131 was identified on samples of vegetation collected within the project. Iodine-131 has a radioactive half-life of 8 days and the immediate answer to the problem of release of this fission product was to increase the cooling period between reactor discharge and fuel element dissolution, even though this action temporarily delayed plutonium production. In addition, controls were established such that dissolving could be performed only during weather conditions favorable for iodine dispersion. In a short time, silver reactors (see Chapter 3) were designed and installed to remove more than 99.5 per cent of the radioiodine from dissolver off-gas before it was vented to the atmosphere.

Hanford has carried on over the years a program of continual sampling and analysis of exhaust air, the environmental air, and the vegetation in the environs. Based on these studies, Hanford has established a working limit of 10 curies/week total emission of iodine-131. Actual measured dispersal of iodine for the years 1953 through 1958 are shown in Table 17.

Table 17

ATMOSPHERIC DISPOSAL OF RADIOACTIVE IODINE FROM THE SEPARATIONS PLANTS STACKS[a]

Year	Curies of Iodine-131 per Day
1953	2.0
1954	1.5
1955	3.2
1956	1.0
1957	1.0
1958	1.2

[a]R. E. Tomlinson, Release of Gases, Vapors, and Particles to the Atmosphere, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 275, August, 1959.

The necessity of installing filters for the large-volume ventilation exhaust air was first recognized in 1947, when radioactive particles were found on the ground around the stack. Large filter beds of graded

layers of sand were constructed through which the building exhaust air was routed before being released to the stack. In the more recently constructed Purex Separation Plant, the filter bed is made of graded mats of glass fiber (see Chapter 3). Either type retains more than 99.5 per cent of the particulate radioactive material entering the unit; the glass fiber unit is somewhat more efficient.

Another radioactive fission product which required specific study is ruthenium. Beginning in 1952, ruthenium-103 and -106 were found to be escaping to the environs and were detected on the ground and vegetation. Process changes were made to reduce ruthenium loss and, in addition, the process off-gas was routed with the ventilation air exhaust through the sand filters. The sand filter not only removes ruthenium particles but also acts as an absorber for any ruthenium tetroxide vapor remaining in the air. Since these changes, ruthenium emissions have been insignificant.

The bulk of the fission products present in the irradiated fuel elements are retained in the aqueous waste solution from the first stage of the separations process. Typical waste compositions arising from past and current operations are presented in Table 15. The radioisotopes produced by the fission of uranium are of such character that they must be positively contained for many hundreds of years. The concentrations of significant isotopes are presented in Table 16 on the assumptions that the uranium contained an initial enrichment of 1.2% uranium-235, was irradiated at 5 MW/T to a total exposure of 2500 MWD/T, and all of the fission products were segregated in 100 gallons of waste for storage. Each concentration is then compared with the respective maximum permissible concentration (MPC) for that isotope in drinking water. Any unrecovered plutonium and other transuranic elements are also present and must be positively contained with the fission products.

These high-level wastes are stored as alkaline slurries in underground tanks. The tanks are constructed of reinforced concrete lined with steel plate. Each has a capacity of 500,000 to 1,000,000 gallons. The use of such tanks is considered storage rather than disposal, and no environmental hazard exists as long as the tanks maintain their integrity. The life of the tanks is not yet known since none has ever failed, but it is estimated to be at least several decades. Laboratory data indicate that carbon steel exposed to simulated neutralized Purex waste media at 220 F corrodes at a rate between 10^{-5} and 10^{-3} inch per month. Based on this, the $\frac{3}{8}$-in. steel side liner of the Purex waste tank could be expected to last at least several decades. The bottom plate could be exposed to higher temperatures and might corrode somewhat more rapidly. Even so, a life expectancy measured in decades appears to be conservative.

The underground waste storage tanks were constructed in groups of six to 18, called farms. Surrounding each farm are 2 or 3 wells which penetrate to the ground water (200 or more feet) and 5 to 10 dry wells 100 to

150 ft deep. Table 18 presents data concerning the size, costs, and time of construction of the tank farms at Hanford. Approximately 52 million gallons of radioactive wastes are currently being stored at Hanford.

Table 18

WASTE-STORAGE TANKS AT HANFORD[a]

Farm	Tanks per Farm	Capacity per tank (gal)	Capacity per farm (gal)	Year Constructed	Cost per farm[b]	Cost per gal, $
T	16					
	4	54,500	6,617,000	1943-44	$ 3,087,000	0.467
	12	530,000				
U	16					
	4	54,500	6,578,000	1943-44	2,969,000	0.451
	12	530,000				
B	16					
	4	54,500	6,590,000	1943-44	3,019,000	0.458
	12	530,000				
C	16					
	4	54,500	6,578,000	1943-44	2,938,000	0.447
	12	530,000				
BX	12	530,000	6,360,000	1946-47	2,208,000	0.347
TX	18	758,000	13,644,000	1947-48	5,859,000	0.429
BY	12	758,000	9,096,000	1948-49	2,651,000	0.291
S	12	758,000	9,096,000	1950-51	3,961,000	0.435
TY	6	758,000	4,548,000	1951-52	1,846,000	0.406
SX	15	1,000,000	15,000,000	1953-54	3,983,000	0.266
A	6	1,000,000	6,000,000	1954-55	4,989,000	0.831
Total	145	-	90,107,000	-	$37,510,000	0.416[c]

[a]R. E. Tomlinson, Radioactive Waste Management Operations at the Hanford Works, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 293, August, 1959.

[b]Includes original tank farm cost plus improvements, including instrumentation, agitation systems, and transfer lines from separation plants. A farm costs include the estimated cost of remotely maintained stainless-steel condensers and auxiliaries ($800,000) currently being installed. Operating and maintenance costs of approximately $200,000 per year for the entire complex are not included.

[c]Average

Liquids which have intermediate concentrations of radioactive materials include waste streams from the later decontamination steps, spent solutions used to absorb or scrub gases, condensed vapors from self-boiling tanks, various other condensates from processing equipment, and aged solutions from which the long-lived isotopes have been scavenged. This type of waste is put into the ground by seepage through structures known as cribs. Because of the favorable geological and hydrological conditions in the separations area, it is possible to retain the vast majority of the radioactive materials in a thick layer of sediment. Thus the wastes are essentially "stored" in the ground and the water percolating to the water table is substantially decontaminated. Swamps and trenches are also used in addition to cribs for the disposal of essentially uncontaminated but suspect waters. Through 1958 there were about 35 billion gallons of water disposed to swamps. There were over 3.7 billion gallons of water with about 1.9 million curies of gross beta emitters disposed to the ground through a total of 71 crib structures, and 28 million gallons containing 647,000 curies of gross beta emitters disposed to 18 trench sites.

Savannah River[23]

The process used at Savannah River is essentially the same as that used at Hanford, and the methods of handling the highly radioactive wastes are also similar. Dissolver off-gases are passed through a reactor containing silver nitrate to remove iodine. Particulate matter in the off-gas is removed by a filter. After processing, the dissolver off-gases along with exhaust air from the processing building are discharged to a 200-ft stack.

As at Hanford, the bulk of the fission products appear in a first-cycle aqueous waste stream. To reduce the volume of material to be stored, all waste streams are evaporated to the maximum solids concentration possible without the formation of precipitates. All liquid wastes are neutralized and stored in underground carbon-steel tanks.

Cooling water from various vessels and steam condensate from evaporator coils are normally not contaminated and are discharged to surface streams. To avoid release of activity, however, the water is monitored and flows through a delaying basin which has sufficient capacity to permit shutting down the operation before activity actually reaches the streams.

High-level wastes are stored in tanks. The primary tank is constructed of carbon steel. The tank rests in a steel saucer designed to retain leakage from a faulty primary tank, at least for a period. The tank and saucer are enclosed in a reinforced-concrete structure with an annular space to permit inspection of the tank proper. Tank vents are provided with condensers and filters. Cooling coils are provided to avoid the possibility that the radioactive decay heat will lead to uncontrolled boiling.

It has been found that the high-level wastes generate a precipitate during storage, which carries an estimated 90 per cent of the fission products to the bottom of the tank with the result that the bottoms of the tank have been heated to about 300 F although the supernate was cool. The costs of the Savannah River tanks are shown in Table 19.

Table 19

HIGH-LEVEL WASTE STORAGE TANKS
AT SAVANNAH RIVER PLANT[a]

	Number of Tanks	Total Gallons	Cost
Original Plant	12	9,000,000	$14,200,000
Added 1956	4	4,100,000	4,700,000
Under Construction	4	5,200,000	2,300,000
Total	20	18,300,000	$21,200,000

[a]R. J. Christl, Waste Management, Savannah River Plant, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 40, August, 1959.

Open seepage basins are used for disposal of very low-level wastes since it would be economically unfeasible to evaporate the hundreds of thousands of gallons involved. Flow to these basins averages about 80,000 gallons per day, and the total activity to the basin to date has been 2.5 curies of alpha emitters, 240 curies of nonvolatile beta emitters, and 2,300 curies of iodine-131.

All solid wastes are buried in a centrally located fenced area. Most burial is in slit trenches which are backfilled as waste accumulates.

Idaho Chemical Processing Plant[24,25]

The Idaho Chemical Processing Plant is devoted principally to the recovery of enriched uranium from spent reactor fuel elements. This recovery process also involves dissolution of the fuel elements in acid, followed by solvent extraction. The wastes produced are similar to those at Hanford and Savannah River. The high-level wastes are stored in underground stainless steel tanks of 300,000-gallon capacity. These tanks are water cooled to eliminate boiling and to reduce corrosion of tank materials. The wastes from later cycles are stored together in uncooled tanks. The volume stored varies from 50 to 150 gallons per pound of uranium recovered.

The total investment in storage tanks is $7,700,000, made up as follows: nine 300,000-gallon permanent storage tanks for aluminum wastes, of which six are cooled. The average cost is $789,000, which is equivalent to a storage cost of $2.63 per gallon, including a portion of piping. Four 30,000-gallon tanks are provided for storage of zirconium wastes. The total cost is $580,000, which is equivalent to a per gallon cost of $4.90.

Low-level wastes, such as cell floor drains, laboratory drains, and equipment decontamination solutions, are collected in small underground tanks, sampled for radioactivity, and thenfed to an evaporator where most of the water is taken overhead at an activity level low enough to permit disposal after dilution directly to the area water table by means of a disposal well. The concentrate is added to the storage tanks.

All solid radioactive wastes generated at the National Reactor Testing Station are disposed of in a common burial pit, either by dumping into trenches or stacking in large pits. The burial ground consists of an area of approximately 80 acres enclosed by a barbed wire fence. It is located in the southwestern portion of the station and not directly up the ground water stream from existing or potential plant sites. It is at least 60 miles from the nearest downstream populated area where water may be used. Trenches average 900 ft in length, 10 ft in depth, and from 5 to 10 ft in width. Large open pits, 6 to 10 ft in depth, 40 to 50 ft in width, and 900 ft in length, are also used for disposal of low-level radioactive material.

The bulk of the solid wastes is low-level material consisting mainly of contaminated items such as rags, papers, sample bottles, lumber and metal scraps. These wastes are transported to burial sites in cardboard boxes by means of closed dump trucks.

High-level wastes (defined at Idaho as those reading greater than 5 roentgens per hour at contact) require special handling. Remote-handling equipment, shielded containers, and remote truck dumping control are utilized for the disposal of these wastes. This type of material involves metal pieces which have been exposed to high levels of neutron radiation and constitutes a personnel exposure hazard. Such waste is deposited in the lower level of the trenches and is covered with earth to achieve shielding.

Low-level wastes from other AEC installations which do not have suitable disposal facilities are shipped to NRTS for disposal. This waste contributes largely to the total volume of solid wastes which have been deposited in the NRTS burial grounds. Table 20 shows the amount of material which has been disposed of to date.

Table 20

AMOUNT OF SOLID RADIOACTIVE WASTE DISPOSED OF AT THE NATIONAL REACTOR TESTING STATION[a]

Year	Activity, curies	Volume, cubic yards
1952	70	2,000
1953	800	
1954	1,500	
1955	1,500	2,500
1956	10,000	5,000
1957	15,000	6,500
1958	10,000	9,000
Total	40,000	25,000

[a]J. R. Horan, Radioactive Waste Disposal Management at the National Reactor Testing Station, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 593, August, 1959.

Isotope Distribution

In 1946 the AEC initiated a program for distributing reactor-produced radioisotopes. Since then nearly 1 million curies of activity have been shipped to over 4,000 institutions throughout the United States. About 98 per cent of the isotopes shipped have half-lives greater than 30 days. Almost all of this longer-lived material is contained in sealed sources.

The amount of radioactive wastes generated through the use of radioisotopes is exceedingly small when compared to the Nation's overall nuclear energy program. At the present rate of use it is estimated that about 200 curies of wastes with a half-life greater than 30 days and 400 to 500 curies with a half-life less than 30 days are being generated each year.

Research Operations

There are a number of AEC sites whose business is primarily research. The amount of high-level wastes handled at such sites is quite small, and the major problem is to arrive at a satisfactory working solution for large-volume, lightly contaminated liquids, solids and gases. The problem is complicated in most cases by the fact that the research sites

tend to be more closely associated with urban populations than do the production sites. The basic operating philosophy at essentially all the research sites is to segregate at the source the very small volumes of relatively high-level material handled so that it does not contaminate the large bulk of material, thus permitting the latter to be returned to the environment with as little treatment as possible. Gases are, in general, filtered through AEC (CWS) filters at the point of production. This treatment is sufficient in almost all cases to permit discharge through short stacks. Solids are collected in convenient containers, accumulated in larger shipping containers and either shipped to a more remote site for burial (usually Oak Ridge National Laboratory, sometimes Idaho) or, in the case of installations located on the coast, dumped at sea.

The problems of liquid disposal and the systems used to handle them are the most complex, and they tend to vary more from site to site. In general, they consist of segregation into two or more activity levels, monitoring and discarding without treatment as much of the waste as possible, and collecting and routinely treating by evaporation, ion exchange, or coagulation only the low-volume, high-activity-level fractions.

Flowsheets for the complete system used at Brookhaven (Figure 4) and liquid system at Argonne (Figure 5) are given. As an example of the type of integrated disposal program developed for use at a research site, the system at Argonne National Laboratory will be described in some detail.(26)

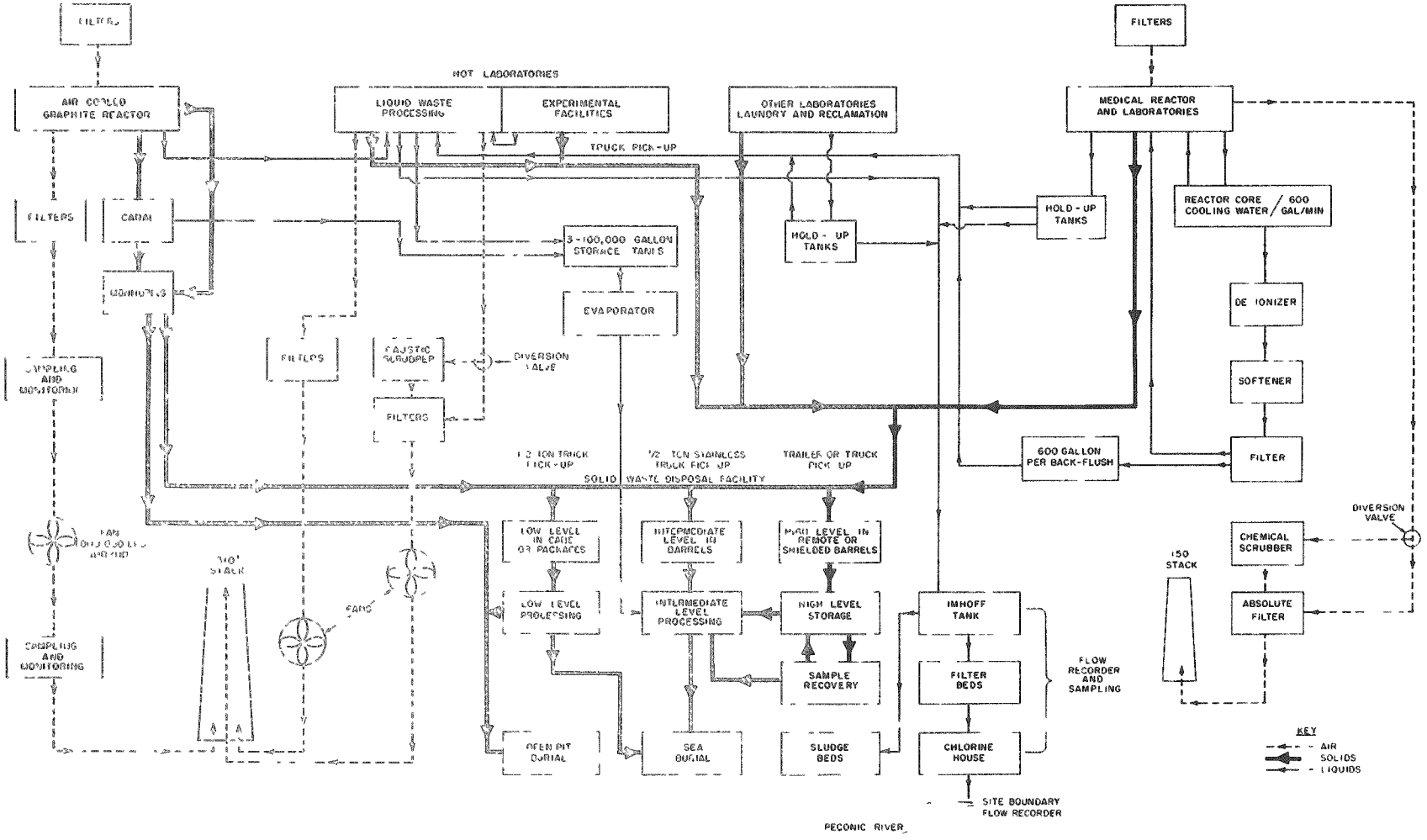
Argonne National Laboratory, a research and development laboratory operated by the University of Chicago under a contract for the Atomic Energy Commission, is located on a 3700-acre plot 25 miles southwest of Chicago, Illinois. The Laboratory has about 3700 employees, of which about 900 to 1000 are staff members. This location is in a highly populated area, the ground water of which receives heavy use. In setting up the site, the Laboratory was obliged to develop complete waste disposal facilities of all types.

The system originally planned for the Laboratory was described in 1951 by Rodger and Fineman.(27) There have been a number of significant changes but the overall pattern now used is very similar to that originally proposed.

The basic philosophy upon which the Argonne waste disposal system was based is that: (1) no fluids will be released from control which exceed the standards (then) established,(6) (2) no wastes will be stored permanently on the site, and (3) all fluid wastes will be reduced to solids for shipment off-site.

Figure 4

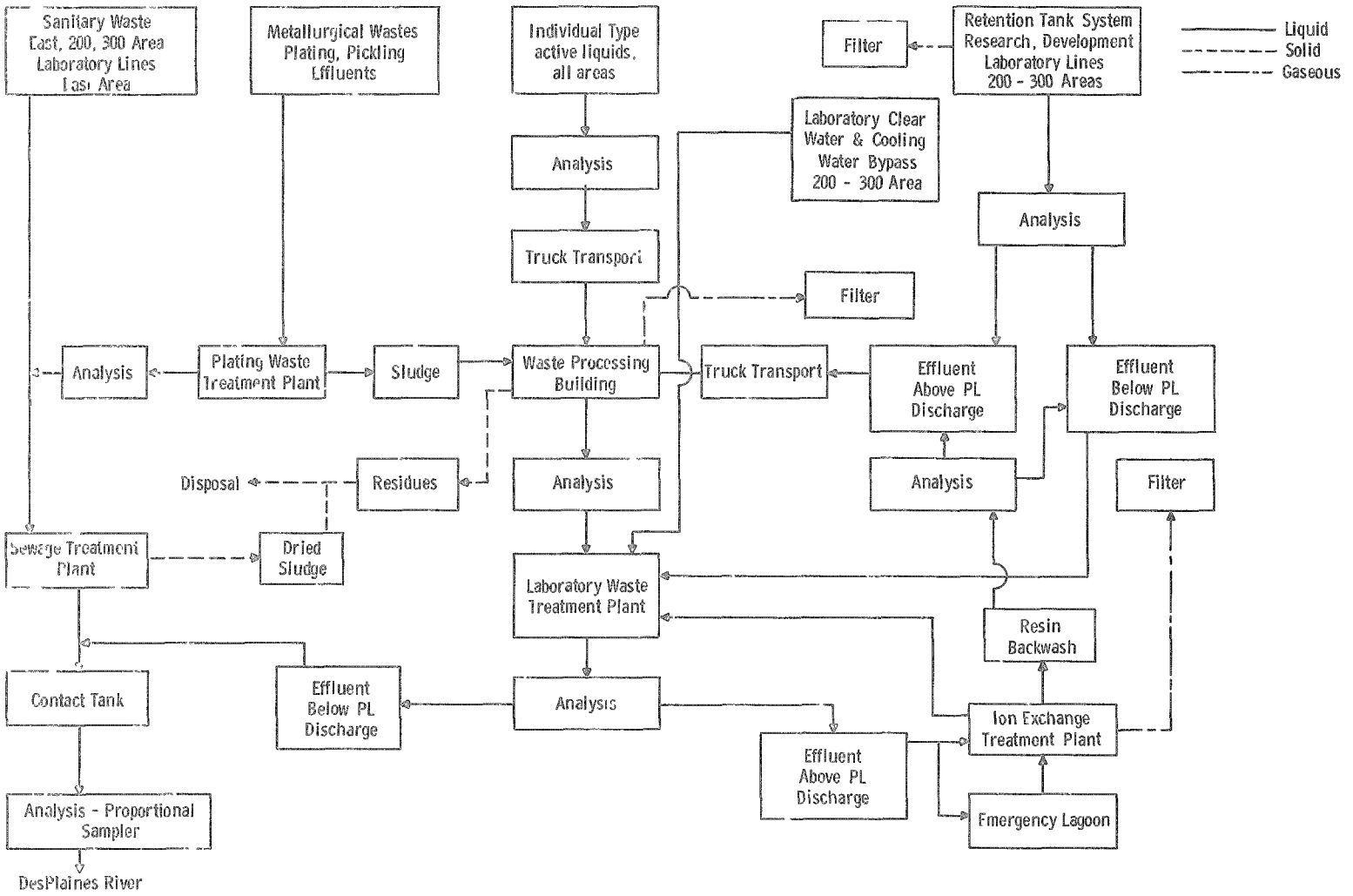
Simplified BNL Radioactive Waste Disposal Flowsheet[a]



[a] F. F. Cowan and L. Gommell, Waste Management Operations at Brookhaven National Laboratory, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 745, August, 1959.

Figure 5

Liquid Waste Disposal System at Argonne National Laboratory[a]



[a]Management of Radioactive Wastes at Argonne National Laboratory, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 723, August, 1959.

Solid Wastes

Two divisions of solid radioactive waste material exist. The first is material which will not exceed restrictions for off-site shipment. This specification is defined by the Interstate Commerce Commission regulations for transportation of radioactive material. The second is material which does exceed this level. At present, the latter is stored under controlled conditions to allow for natural radioactive decay.

For the major portion of the solid waste produced, a stainless steel container with a one-cubic foot fibre drum insert is used as the standard waste receptacle. These containers, supplied as needed by the Waste Control Group, are designed to operate by stepping on a treadle to operate a sliding cover. There are some 470 of these receptacles in approximately 200 locations within the Laboratory. In those areas where considerable quantities of waste are produced or the physical dimensions of the waste are larger than can be accommodated in the standard container, a $4\frac{1}{2}$-cubic foot cardboard container is supplied. For that waste which exceeds a 50-milliroentgen-per-hour limit, shielded containers are available (see Figure 6).

Figure 6

Containers Used for Most of the Solid Waste Collected - Argonne National Laboratory

When a container is full or approaches the radiation limit, it is surveyed by Radiation Safety personnel and the pertinent information recorded on a Hazardous Disposal Form. Pickup from some 27 facilities in the Laboratory is maintained on a scheduled basis. It consists of removing the inner fibre drum insert, sealing the lid of the drum if required, and removal from the area. Those wastes requiring shielding are handled essentially the same with the exception that they are transferred to additional shielded units. The daily collection is transported to the Waste Storage Area in a specially designed vehicle.

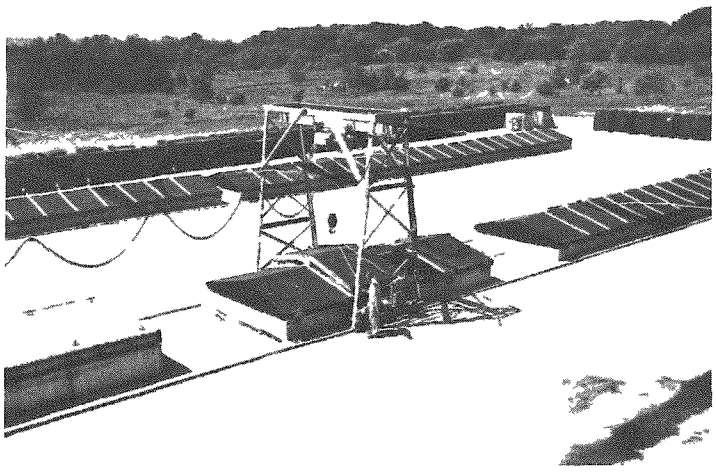
The Waste Storage Area comprises 10 acres in which the various control, concentrating and storage units are located. In this area all wastes are segregated into that which can be shipped off-site and that which requires storage. Material that does not exceed 200 mr/hr and/or does not contain any alpha-active material other than natural uranium is baled. Concentration of about half of the solid waste is accomplished by

compression in a commercial-type hydraulic baling press. The baled material and other nonbalable material is packaged into a final container for off-site disposal. This container, fabricated of 12-gauge steel, reinforced with angle iron, is 4 feet wide by 5 feet long by 6 feet deep. A steel lid with a built-in gasket when bolted to the bin results in a shipping unit of sufficient integrity to meet the requirements for interstate transportation on public carriers. Larger components of shippable waste, such as 55-gallon drums of solidified liquid waste and major components of pilot operations, transported directly to the Waste Storage Area, are handled similarly to baled material.

Five underground storage vaults, some of which are shown in Figure 7, provide the temporary storage requirements for the Laboratory. Three of the vaults, constructed of reinforced concrete, are in the shape of an open box with a drainage control on the bottom. These are about 95 feet long by 13 feet wide by 11 feet deep. They extend some 8 feet below grade and will accommodate a total of 38 bins each. After the bins are placed in the vault, a 1-foot-thick concrete lid is placed above the bins. Protection from the weather is provided by peaked wooden roof sections covered with tar paper. Storage space for approximately five years exists in these vaults. The fourth vault, of reinforced concrete, constructed in the open box shape, consists of six adjacent cells, each cell 6 feet by 10 feet in area by 21 feet deep. The vault, some 18 feet below grade, is utilized for storage of bulky and highly active items of waste. The fifth storage vault is an assemblage of vertical pipe casings, 4-inch to 10-inch diameter, cast as part of a concrete slab. Its dimensions are 12 feet by 30 feet in area by 21 feet deep, with 19 feet below grade. The pipe casings serve as receptacles for aluminum map cases and similarly sized containers. Six-inch-thick lead plugs are used to cap the exposed pipe casings. Storage space for some 9 to 10 years is available in the last two described vaults. A 5-ton-capacity gantry crane provides the means for the handling of vault lids, roofs and miscellaneous equipment required for storage operations. It directly services the 6-cell storage vault. This allows for the remote control

Figure 7

Some of the Temporary Storage Facilities for Solid Waste - Argonne National Laboratory



handling of the crane in connection with the storage of high-level waste. Various types of remote control equipment, such as drum tongs, various size chutes, guide rods and accessory slings, are utilized in these operations.

At present, one shipment of packaged waste is anticipated per year. In April 1957 a 240-bin, 12-car shipment was made to the Oak Ridge National Laboratory. Prior to shipment each bin was numbered, the average weight determined, and a radiation survey made of the five exposed bin surfaces. The 240 bins were loaded at the Waste Collection Area onto a low-boy trailer by a mobile crane and transported to the railroad siding on the site. Here they were loaded by another crane into the railroad cars. Each car was shored to insure minimum movement of material during transit. A final radiation survey was made of each car to insure compliance with ICC regulations. The combined operation was completed in about 4 days, utilizing personnel of the Plant Services, Materials Handling and Industrial Hygiene and Safety Divisions.

A tabulation of total solid waste representing the accumulation for the Fiscal Year 1957 is shown in Table 21. It is separated into the various operations and level of activities that are encountered in the waste disposal program at Argonne National Laboratory. Approximately 97 per cent of all solid wastes were considered low level and prepared for off-site disposal.

Table 21

ACCUMULATION OF SOLID WASTE FY 1957[a]

Total Volume Low-level Waste Collected	23,900 cu ft
Volume Low-level Waste Concentrated	10,200 cu ft[b]
Total Low-level Waste Packaged for Shipment	16,100 cu ft
Collection and Storage, High Level	850 cu ft

[a]Management of Radioactive Wastes at Argonne National Laboratory, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 708, August, 1959.

[b]Compressed volume - 2400 cu ft

Liquid Wastes

The approximate liquid waste volumes of various types which may be expected from a research institution are indicated in Table 22. The varied properties of the different liquid wastes present problems which cannot be handled in a single system. Consequently there are four major systems in use at the Argonne National Laboratory. A general schematic of these systems is shown in Figure 5. The following resume covers the physical operations of the disposal system for the Fiscal Year 1956. Current figures have increased by about 20 per cent.

Table 22

EXPECTED WASTE VOLUMES FROM A RESEARCH LABORATORY[a]

Type of Waste	Volume, gallons/(day) (employee)[b]
Sanitary Sewage	100
Monitored Laboratory Drain Wastes	6.5
Laboratory Cooling Water (Unmonitored)	60
Processed Radioactive Wastes	0.1

[a]Data from Argonne National Laboratory, Fiscal Year 1959.

[b]Based on total employment - 3700.

The first system is the sanitary system which flows to the sewage treatment plant, the outfall of which goes to the Des Plaines River. No radioactivity is knowingly permitted in this system. The second, a small one, is a plating waste treatment plant which handles plating and pickling wastes from the Metallurgy Division. The effluent from this plant discharges into the sanitary sewage system In the handling of radioactivity two systems are used. In the first place, the working scientist is expected to put active liquid waste into supplied containers. These may be shielded or unshielded as required These containers are picked up on request and trucked to the Waste Processing Building. The producing scientist has the responsibility of providing information concerning the content of the waste These wastes are then processed by various means, as will be explained later (Chapter 5), and the effluent discarded via the Laboratory Waste Treatment Plant. The final system is the so-called retention tank system

All sink drains in areas in which radioactivity is used are conducted to retention tanks. These are 1500-gallon glass-lined tanks operated in pairs. When one is full, flow is diverted to the other, a sample taken and checked for radioactivity. If it is above the maximum permissible discharge level, it is pumped out into a tank truck and trucked to the Waste Processing Building for treatment. If it is below this level, it is discarded directly to the Laboratory Waste Treatment Plant. Each of these systems will be discussed in more detail and operating figures for the fiscal or the calendar year 1956 given.

Sanitary System

The sanitary system and sewage treatment plant consist of the following equipment: comminuter, Dorr Clarigester, trickling filter, final clarifier, sand sewage filters, chlorine-contact tank and sludge drying beds. The plant was designed for an average 24-hour flow of 217,000 gallons/day, a peak rate of 570,000 gallons/day, and a maximum storm rate of 1,300,000 gallons/day. The design flowrate has been continuously exceeded. Nevertheless, the quality of the effluent has continued very high, due chiefly to final intermittent sand filtration. The average BOD reduction of 94.1 per cent and the average effluent BOD of 3.6 ppm are well within operating goals. The actual discharge to the Des Plaines River is a combination of sewage treatment plant effluent and Laboratory Waste Treatment Plant effluent discharged at an average ratio of 2.1 to 1. The effluent BOD from Laboratory Waste Treatment Plant varies between 0.6 and 45 ppm and averages 10 ppm. It follows that the BOD of the combined discharge averages 5.7 ppm. This discharge is within operating goals and meets the state criteria for sewer effluent discharges to the Des Plaines River.

Composite samples of the sewage plant effluent and the digested sludge are regularly checked for activity. No activity above MPL has been detected in the sewage plant effluent. Activity above MPL has been found in the digested sludge, requiring its disposal as solid active waste.

Plating Waste Treatment Plant

A small plating waste treatment plant handles some special metallurgical wastes. The plant contains three batch retention tanks which are used for plating, cyanide and pickling wastes. The principal equipment consists of recirculation pumps, agitators, a chlorine feeder, a sulfur dioxide feeder, a caustic feeder, filters and pH indicating and control equipment.

Removal of cyanide is accomplished by chlorination with basic pH control. Chromium is removed by treating with sulfur dioxide and caustic; acid wastes are merely neutralized. Copper and heavy metals are removed by precipitation with sodium hydroxide. The effluent from this

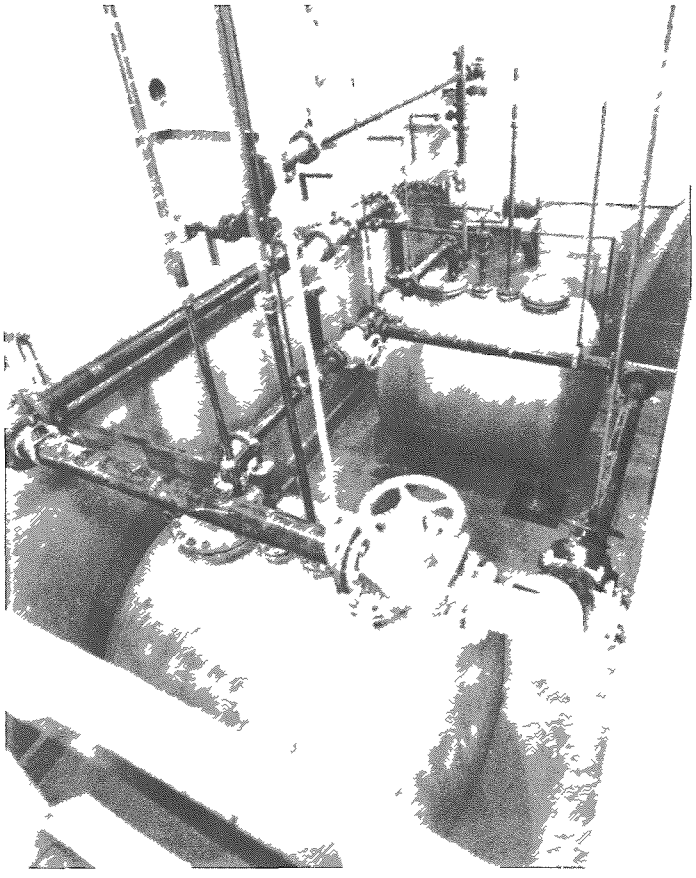
plant is discharged to the sewage treatment plant. The chief goals of this plant are to prevent the discharge of copper, chromium and heavy metals in quantities that would affect the biological operation of the sewage plant, to reduce cyanide to nontoxic constituents, and to adjust the pH. About 400,000 gallons per year are treated.

Treatment of Active Liquid Wastes

The operating philosophy of the active waste treatment system at Argonne is based upon a high degree of segregation at the point of production of the waste. The operating scientist is asked to put any active wastes into containers, which are supplied to him upon request, and removed whenever he desires. He is requested to provide information concerning the chemical and radiochemical constituents of the wastes that he has produced. These are then trucked to the Waste Processing Building. The total volume thus collected each month seldom exceeds a thousand gallons.

Figure 8

A Typical Retention Tank Installation for Monitoring Laboratory Drains



No radioactivity is knowingly permitted in the Laboratory drains. However, to provide complete control of Laboratory effluents, the drains in active working areas are connected to retention tanks. These are glass-lined tanks of 1500-gallon capacity (see Figure 8). They operate in pairs. When one is full, flow is diverted to the other, a sample taken and its radioactivity content determined. The maximum permissible level (MPL) for building retention tanks is as follows:

1. Gross count (alpha plus beta) $\geq$1000 dpm/ml: Process.
2. Gross count <1000 dpm/ml but $\geq$100 dpm/ml: Analyze for Sr^{90} and count for total α.
   a. Sr^{90} level $\geq$10 dpm/ml and/or α level $\geq$5 dpm/ml: Process.
   b. Sr^{90} level <10 dpm/ml and α level <5 dpm/ml: Discharge to Waste Treatment Plant.
3. Gross count <100 dpm/ml: Discharge to Laboratory Waste Treatment Plant.

If the radioactivity found is below the MPL, the waste is pumped directly to the Laboratory Waste Treatment Plant. If above MPL, the water is pumped into a portable tank and trucked to the waste processing area.

It is expected that most of the time activity found in the retention tanks will be less than MPL. Operating experience has been very good in this regard. During the fiscal year 1956, 98 per cent of the retention tanks were below MPL and discardable directly. The operating experience is indicated in Table 23.

Table 23

ARGONNE NATIONAL LABORATORY
OPERATION OF LABORATORY DRAIN WASTE SYSTEM[a]

July 1, 1955 through June 30, 1956

	Number	Volume, gallons
Retention tanks samples and analyzed	4,736	7,191,750
Retention tanks below maximum permissible level for discharge to laboratory waste treatment plant	4,645	7,097,725
Retention tanks above maximum permissible level for discharge to laboratory waste treatment plant	91[b]	94,025

[a] Management of Radioactive Wastes of Argonne National Laboratory, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 708, August, 1959.

[b] Retention tanks above gross permissible discharge level 59
Retention tanks above alpha permissible discharge level 24
Retention tanks above strontium-90 permissible discharge level 3

Active waste collected in various pots and containers and the above-tolerance retention tanks are treated at a central processing facility (Building 310) wherein there is equipment for evaporation, filtration, flocculation, concrete solidification, vermiculite absorption, solvent washing and ion exchange. These will be discussed in Chapters 5 and 6. The collected wastes are treated in whatever manner will give the needed decontamination at the least cost. Operating experience for the calendar year 1956 is shown in Table 24.

Table 24

ARGONNE NATIONAL LABORATORY
OPERATION OF RADIOACTIVE LIQUID WASTE
TREATMENT BUILDING[a]

January 1, 1956 through December 31, 1956

Waste Treated by	Volume, gallons
Evaporation	159,330
Filtration	3,850
Ion Exchange	0
Flocculation	23,450
Concrete Solidification	30
Vermiculite Absorption	980
Solvent Washing	280
Total	187,920

[a]Management of Radioactive Wastes of Argonne National Laboratory, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 708, August, 1959.

These treated wastes are also discharged to the Laboratory Waste Treatment Plant along with the retention tank wastes which were determined to be below MPL and with cooling water which bypasses the retention tanks

The Laboratory Waste Treatment Plant, part of which is shown in Figure 9, consists of the following principal units:

a) manually raked bar screen for screening coarse suspended matter,

b) pH recording and control equipment, dry chemical feed machines, mixing tanks for automatic pH correction by means of caustic addition;

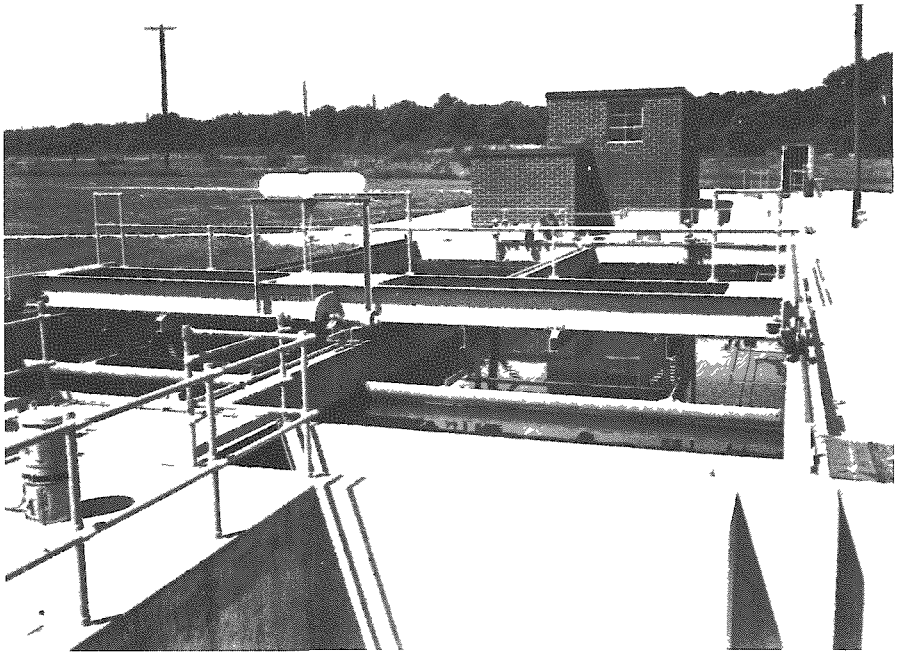
c) four 70,000-gallon tanks containing recirculation facilities and sludge scraper mechanism for equalizing the pH and collecting the settled sludge;

d) one rate controller to control the discharge flow; and

e) one 385,000-gallon capacity earth lagoon for emergencies.

Figure 9

Portion of Laboratory Waste
Treatment Plant at
Argonne National Laboratory



The plant was designed to neutralize acid wastes by means of caustic addition. It was designed to treat a total flow of 137,000 gallons per day at a peak rate of 420,000 gallons per day. Since this time the original tank capacity of this plant has been doubled from two to four 70,000-gallon holding tanks. The plant is so arranged that it can be operated by the batch method or by continuous flow.

The MPL for final effluent discharge were established in 1954 at the rates shown in Table 25.

Table 25

YEARLY DISCHARGE GOALS FOR VARIOUS TYPES OF ACTIVITY

Type of Activity	Yearly Discharge, curies	Equivalent Concentration, $\mu c/ml$
Alpha Emitters	0.15	2.5×10^{-7}
Beta Emitters (other than Strontium-90)	10	1.67×10^{-5}
Strontium-90	0.45	7.5×10^{-7}

Reducing these values to operating levels for discharge of the 70,000-gallon batches of waste from the Laboratory Waste Treatment Plant results in the following rules:

1. Gross count (alpha plus beta) $\geq$125 dpm/ml: Hold for process.
2. Gross count $<$125 dpm/ml, but $\geq$5 dpm/ml and/or α count $\geq$1 dpm/ml: Analyze.
   a. Sr^{90} level $\geq$2 dpm/ml and/or α level $\geq$1 dpm/ml: Obtain ruling from Industrial Hygiene & Safety Division as to disposition.
   b. Sr^{90} level $<$ 2 dpm/ml and α level $<$1 dpm/ml: Discharge to outfall.
3. Gross count $<$5 dpm/ml and α count $<$1 dpm/ml: Discharge to outfall.

It should be noted from U.S. Department of Commerce Handbook 69,[10] that the only maximum permissible concentrations which could be exceeded using these MPL values would be I^{129} for a 40-hour week and I^{129} and Ra^{226} for a 168-hour week. The additional dilution contributed by the Des Plaines River has not been included in the calculations.

Operating results for the fiscal year of 1956 are shown in Table 26.

Gaseous Wastes

The bulk of the gaseous wastes comes from hood and cave exhausts. Chemical, metallurgical or biological operations involving radioactive materials, as well as operations using hazardous although nonradioactive materials, are done in hoods. The exhaust from such a hood may run from one thousand to several thousand cubic feet per minute. In the Chemistry Research laboratory building alone there are 175 of these hoods

Table 26

ARGONNE NATIONAL LABORATORY
LABORATORY WASTE TREATMENT PLANT OPERATION

July 1, 1955 through June 30, 1956

Total number of Tanks Analyzed	1,111
Total Gallons Discharged	63,986,360
Number of Tanks Containing Activity above Provisional MPL	2
Total Gallons in above Tanks	123,770

Disposition: Both tanks were discharged to the Emergency Lagoon. The Emergency Lagoon contents were eventually discharged through the outfall sewer, the effluent of which was below provisional MPL as a result of (1) decay, (2) dilution due to rainfall and laboratory waste overflows into the lagoon.

Radioactivity Discharged to the Laboratory Outfall

Type of Activity	Yearly MPL	Activity Discharged July 1, 1955 through June 30, 1956
Alpha Emitters	150 millicuries	13.7 millicuries
Beta Emitters (Other than Strontium 90)	10 curies	0.135 curie
Strontium 90	450 millicuries	0.42 millicurie[b]

Minimum "pH" of Raw Waste	0.7
Maximum "pH" of Raw Waste	9.7
Minimum "pH" of Treated Waste	5.2
Maximum "pH" of Treated Waste	9.7
Minimum BOD Treated Waste[a]	0.6 ppm
Maximum BOD Treated Waste[a]	45 ppm
Average BOD Treated Waste[a]	10.2 ppm

[a]Results obtained from occasional checks of tank contents

[b]Result of specific analyses. Total volume discarded if just at detection limit would be equivalent to 300 mc.

Easily accessible at the back of the hood are four prefilters that even out the air flow throughout the hood volume and serve as prefilters for both dust and radioactivity, thus lessening the load on the rest of the system. The exhaust gases are conducted through a final filter before they reach the blower that discharges them through 3-foot stacks on the roof.

Radiation Safety personnel periodically survey filters to insure that personnel working in close proximity are not exposed to radiation above the permissible level. Personnel of the Reclamation Department are requested to install new filters when particulate matter retained on the filter restricts the passage of air below the limit prescribed by design requirements or when the level of radioactivity rises above the permissible level. The removal and installation of filters includes the delivery of the spent filters to the Waste Storage Area where they are disposed of as solid waste.

Tools used to machine uranium are each individually hooded. Metal and oxide dusts make up the bulk of the contaminants in the 1,200 cu ft/min discharged from each of these machines. These dusts are removed in a Rotoclone system. The Rotoclone consists of two sections: the first, a water impingement tank, the second, an electrostatic precipitator. The liquid scrubber waste is eventually trucked to the Waste Processing Building for further processing.

Costs

The total capital investment in the Argonne waste disposal system is $2,740,000. The annual operating charges are about $340,000 or roughly one per cent of the total Laboratory budget. These data are shown in Table 27.

Table 27

CAPITAL AND OPERATING CHARGES FOR THE ARGONNE NATIONAL LABORATORY WASTE DISPOSAL SYSTEM

	Capital Cost of Facilities		
	Capital Investment	Equipment	Total
Solid Waste System	$ 180,000	$ 23,000	$ 203,000
Liquid Waste System	1,865,000	215,000	2,080,000
Gaseous Waste System	695,000	-	695,000
Total	$2,740,000	$238,000	$2,978,000

	Annual Operating Cost [a,c]
Solid Waste System	$ 90,000
Liquid Waste System	190,000
Gaseous Waste System	60,000
	$340,000[b]

[a] Includes all operations, maintenance, amortization and overhead.

[b] Amortization and depreciation totals $88,500

[c] For Fiscal 1957

PROBLEMS FOR CHAPTER 2

1. How many curies of activity are mined each day by the uranium mining industry at the current production rate ?

2. Radon diffusing out of rock in a mine is free of any daughter products. Taking the major decay products as

$$Rn^{222} \xrightarrow[\alpha]{3.825\ da} Po^{218} \xrightarrow[\alpha]{3.05\ min} Pb^{214} \xrightarrow[\beta]{26.8\ min}$$

$$Bi^{214} \xrightarrow[\beta]{19.7\ min} Po^{214} \xrightarrow[\alpha]{0.00016\ sec} Pb^{210} \xrightarrow[\beta]{19.4\ y} ,$$

calculate how long it takes for Po^{218} and Pb^{214} to reach their maxima.

3. Estimate the quantity of radium-226 and thorium-230 (ionium) which is stockpiled at various places in this country. Where will most of it be found ?

4. The uranium lost into the gaseous waste streams at the Oak Ridge and Paducah gaseous diffusion plants is largely natural uranium. Taking its value as \$40/kg, what is the total yearly dollar loss from the two plants ?

5. Convert the data of Table 16 into curies/gallon of initial high-level waste, assuming that 1000 gallons of waste are produced per ton of uranium processed. Compare this result (at 90 days) to the result you obtained in Problem 2, Chapter 1, for a total of eight isotopes assuming that this fuel is burned to 2500 Mwd/ton.

6. Assuming that the average energy of disintegration is 1 Mev, convert the answers of Problem 5 to watts/gallon and Btu/(hour)(gallon).

7. Coppinger and Tomlinson give data from which an "effective h" for transfer of heat from a buried tank to soil can be deduced to be approximately 0.2 Btu/(hour)(sq ft)(F). Making the very simplifying assumptions that this is for average soil and that the heat removal will be proportional to the surface area of the tank and to Δt, estimate the temperature in spherical tanks of 10,000-, 100,000-, 500,000- and 1,000,000-gallon capacity holding waste generating 20 Btu/(hour)(gallon). If the temperature reaches the boiling point estimate the boilup rate.

8. Using your solution to Problem 2 of Chapter 1, estimate how long the 500,000-gallon tank of Problem 7 will boil.

9. A 100-Mw (t) reactor is cooled by once-through river water which is permitted to increase 10 F upon going through the reactor. Assume that 1 gallon of the high-level waste described in Table 16 were mixed with this cooling water effluent. How long would the reactor have to operate to produce sufficient cooling water to dilute this high-level waste just to MPC?

CHAPTER 3

HANDLING OF GASEOUS WASTES

In the second chapter, consideration was given to the overall systems of waste management in use at various sites. In this chapter the details of processes which are available for handling gaseous wastes will be covered. In subsequent chapters methods for solids and liquids will be detailed. These processes have been studied rather extensively and there is a wealth of information available on many of them. They are available for use within the framework of any waste management procedure.

Gaseous wastes may truly be gases but more likely the activity associated with a gas stream will be contained in various forms of suspended solid or liquid particles. Definitions of some of these forms are:

Aerosol - a dispersion of solid or liquid particles of microscopic size in gaseous media.

Dust - a term loosely applied to particles of solid capable of being suspended in a gas. Size of particles can vary from microscopic to visible.

Fume - solid particles formed by condensation from the gaseous state.

Smoke - small particles of liquid or solid, usually formed by incomplete combustion and consisting primarily of carbon or other combustible material suspended in a gas stream.

Mist - a term loosely applied to dispersions of liquid particles in a gas.

Fog - a visible mist formed by the condensation of water.

Gas Mixture - a state of dispersion in which the dispersed material is in units of molecular size.

In Table 28 there are listed ten gaseous radioisotopes and their MPC values for continuous exposure. Note that most of them have MPC values of the order of 10^{-6} or 10^{-7} μc/cc. Comparing this with Table 9 (Chapter 1) it may be seen that the MPC for a completey unidentified source is 4×10^{-13} μc/cc and that no beta emitter requires a limit lower than 10^{-11} μc/cc. Thus not only is the activity of a gas stream more likely to be associated with aerosols, but these are considerably more hazardous than the true gases. This is a rather fortunate circumstance since there are more ways to get at the removal of aerosols than of gases. The methods to be considered herein are ventilation control, filtration, electrostatic precipitation, scrubbing, and chemical adsorption.

Table 28

MAXIMUM PERMISSIBLE CONCENTRATIONS OF SOME RADIOGASES IN AIR (For 168-hour Week)

Isotope	Half-life	MPC, μc/cc of air[a]
H^3	12 years	5×10^{-6}
C^{14} (CO_2)	5570 years	10^{-6}
Cl^{36}	4×10^5 years	10^{-7}
A^{41}	1.82 hours	4×10^{-7}
Kr^{85}	9.4 years	10^{-6}
Ru^{106}	1.0 year	3×10^{-8}
I^{129}	1.7×10^7 years	6×10^{-10}
I^{131}	8.14 days	3×10^{-9}
Xe^{133}	5.27 days	3×10^{-6}
Xe^{135}	9.2 hours	10^{-6}

[a]National Bureau of Standards Handbook 69, June 1959.

Sampling[29]

In handling any radioactive waste a reasonably accurate sample is desirable. In some cases, particularly for solids, this is practically impossible. Gases may be samples with a variety of devices, but obtaining a meaningful sample may be quite difficult. The various types of dispersions defined require somewhat different treatment.

No matter what form gas contamination takes, the condition is likely to be transitory and localized. Therefore, samples taken over a short period of time can be misleading. Selection of the sample point is particularly important; a sample taken a few feet from the actual breathing zone may be quite unrepresentative of the actual exposure. Ideally, samples should be obtained continuously from a variety of points.

The most common air-sampling devices employ filtration. Such a unit would contain a source of suction, a flow-measuring device, and a holder containing the filter medium. Various crystals, glass fibers, minerals, plastics, sand, and other materials may be used as filter media, but those most generally used are papers, some of which are listed in Table 29. The all-cellulose filter papers (Whatman) are cheap, can be counted directly with low loss, and can be dissolved in reagents for analysis. They have low resistance to heat and moisture. Cellulose and asbestos papers (CWS, HV-70) are more expensive, more resistant to

Table 29

FILTER PAPERS USEFUL IN AIR SAMPLING[a]

Designation	Manufacturer	Reference	Usage
HV-70	Hollingsworth and Voss	b,d,e,f	General, cannot be dissolved
CWS-6	Chemical Corps Cambridge Filter Corp. Flanders Mill Mine Safety Appl. Co.	b,c,d,e	Same
MSA type S	MSA	b,c	Low resistance, must be ashed before counting
Glass paper	MSA 1106-B	d,e,f	High strength and chemical resistance
Glass paper	Hurlburt X935-B	b,f	Can be leached but not dissolved
Whatman No. 1	Balston, Ltd.	b,f	Cheap, available, can be dissolved
Whatman No. 4	Balston, Ltd.	b	Cheap, available, can be dissolved
Whatman No. 40	Balston, Ltd.	b,d,e	Cheap, available, can be dissolved
Whatman No. 41	Balston, Ltd.	b,c,d,e,f	Variable quality, can be dissolved, widely used
Whatman No. 41-H	Balston, Ltd.	b	Higher resistance, stronger
Whatman No. 42	Balston, Ltd.	b,f	Cheap, available, can be dissolved
Whatman No. 44	Balston, Ltd.	b	High efficiency
Whatman No. 50	Balston, Ltd.	b	Cheap, available
Whatman No. 52	Balston, Ltd.	f	Cheap, available
Millipore HA	Lovell Chemical Co.	b,g	Dissolves in solvents, high resistance, low strength
Millipore AA	Lovell Chemical Co.	b,g	Lower resistance than HA, fragile

[a]Hanson Blatz, Editor, Radiation Hygiene Handbook, McGraw-Hill Book Co, New York (1959) Section 20.

[b]Smith, W. J. and N. F. Surprenant, Properties of Various Filtering Media for Atmospheric Dust Sampling, presented at meeting ASTM, Philadelphia, Pa. July 1, 1953.

[c]Adley, F. E. et al., A Study of Efficiencies and Pressure-drop Characteristics of Air-filtering Media, Document HW-28065, Hanford (August 10, 1953).

[d]Fitzgerald, J. J. and C. G. Detwiler, Collection Efficiency of Air-cleaning and Air-sampling Filter Media, Am. Ind. Hyg. Assoc. Quart., 16 122-130 (June 1955).

[e]Fitzgerald, J. J. and C. G. Detwiler, Collection Efficiency of Air-cleaning and Air-sampling Filter Media in the Particle Size Range of 0.005 to 0.1 Micron, KAPL-1463 (Dec. 9, 1955).

[f]L. Silverman and P. LaTurre, Collecting Efficiencies of Filter Papers for Sampling Lead Fume, A.M.A. Arch. Ind. Health, 11 243 (1955).

[g]M. W. First and L. Silverman, Air Sampling with Membrane Filters, A.M.A. Arch. Ind. Hyg. and Occup. Med., 7, 1-11 (January 1953).

heat and moisture, and can be counted directly with a somewhat higher loss; they cannot be dissolved. The glass papers are more expensive and quite variable in quality. They have high strength even when wet and are highly resistant to most chemicals and to heat. They can be counted directly with low loss but cannot be dissolved. Millipore filters are sieves made from plastic membranes perforated with small conical holes. They are expensive, fragile, and require special holders, but they are highly efficient. They are soluble in many organic solvents and they can be rendered transparent with microscope immersion oil.

Electrostatic precipitators have been used for fume and dust sampling. They have not been widely used for radioactive samples except in cases where the sample is to be subjected to subsequent chemical manipulation.

Impingement and impaction devices put the gas to be sampled through a jet to increase its velocity and collect the particles in a liquid (impingement) or on the surface of a plate (impaction). Neither is efficient for fine dust and hence their usefulness in radioactive gas sampling is limited. For hot wet atmospheres they sometimes have to be used. Incinerator stack gases are often sampled in this way.

Determination of truly gaseous activity is difficult. The technique most generally used is to introduce a sample of the gas into an ion chamber and measure the ion current produced.

Samples of particulates obtained by filtration or other means may be taken from the sampler and counted in a separate instrument or the counter may be made a part of the sampling device. If filter papers are used, corrections must be made for self-absorption in the paper, particularly in counting alpha emitters. This correction may be determined by counting the sample on the paper, then ashing it, and plating out the remaining activity on a metal disk and recounting. Self-absorption in Whatman 41 paper has been measured to be about 30 per cent. With millipore filters, electrostatic precipitator tubes, and impactor slides there is essentially no self-adsorption.

Air samples must also be corrected for counts obtained from the disintegration products of radon and thoron. Since the concentration of these varies considerably, no standard correction factor is possible. If no appreciable thoron-decay products are present, it is only necessary to allow the sample to decay for a few hours, since the radon daughter products are short lived and will decay out quickly. But the time necessary to permit decay of the thoron products is too long to be practical. In this case the sample is generally counted twice, once in about 4 hours and one after 24 hours. The true count of long lived alpha emitters is then given by

$$C = \frac{C_2 - C_1 e^{-\lambda \Delta t}}{1 - e^{-\lambda \Delta t}} ,$$

where

C_2 = total count after 24 hours (t_2)

C_1 = total count after 4 hours (t_1)

$\Delta t = t_2 - t_1$

λ = decay constant for thorium B

$= 0.693/10.6 = 0.0653 \text{ hr}^{-1}$

If the measurements are made exactly at 4 and 24 hours, $\Delta t = 20$ and $e^{-\lambda \Delta t} = 0.271$. In this case

$$C = (C_2 - 0.271\ C_1)/0.729$$

It is apparent that this procedure causes a delay in determining air-concentration data. No hardship is ordinarily thus imposed because it is only in the case of an emergency that immediate answers are needed, and in an emergency the counts will be so high as to make the correction superfluous anyway.

When gas samples are taken from ducts, stacks, or any flowing stream, additional precautions are necessary. The inserted sampling lead should be as small as possible so as to disturb the flow pattern as little as possible. It should point upstream so that it is not necessary to bend the path of the particle. Suction to the sampler should be adjusted so that the flow rate in the sample tube matches that in the stream being sampled. This latter condition is known as isokinetic sampling. It is more important the larger the particles being sampled. For particles smaller than 5 microns it is not necessary.

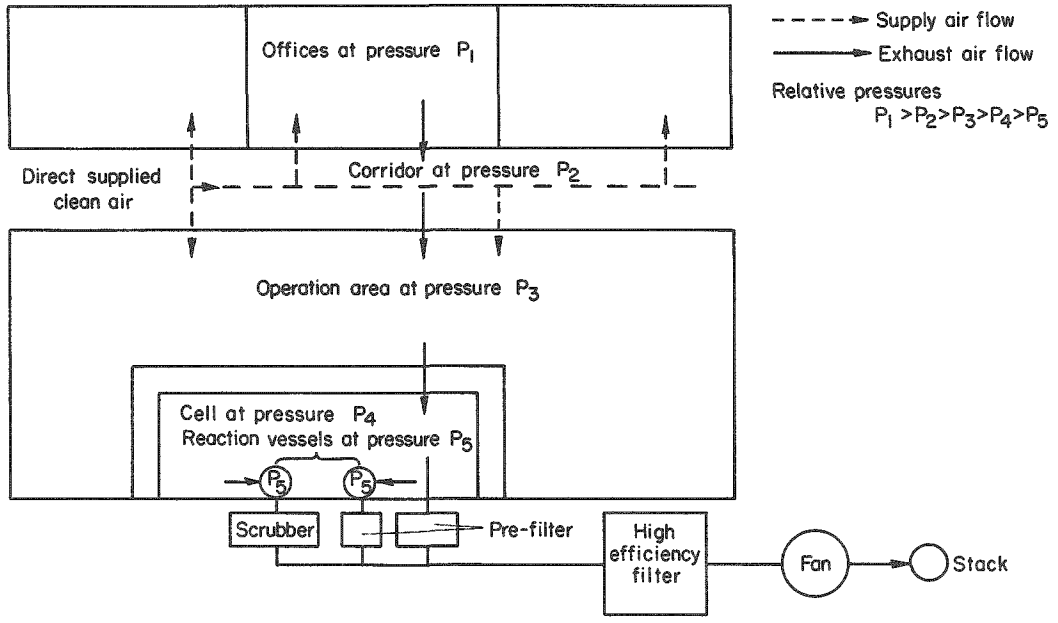
Ventilation

A primary step in the handling of gaseous wastes, one which applies to both gases and aerosols, is the proper selection and operation of ventilation flow patterns. Areas containing radioactive material should always be maintained at a pressure negative to the areas in which operating personnel are to be permitted. A negative pressure equivalent to about 0.025 in. of water is desirable. Sufficient air flow should be provided so that a linear velocity of at least 135 ft/min will be maintained at all times through any opening to the working areas, and such openings should be kept to a minimum. It is desirable to arrange air flow patterns so that clean air moves progressively through more and more contaminated areas and exits through

ducts from the most contaminated area. This procedure has the advantage of air conservation, which is particularly important in air-conditioned areas and reduces the size of treating facilities. Figure 10 shows an example of a schematic layout for good ventilating flow pattern.

Figure 10

Desirable Ventilation Flow Pattern[a]



[a]H. Etherington, editor, Nuclear Engineering Handbook, McGraw-Hill Book Co., New York (1958) p. 11-134

Air leaving the working areas will generally receive some form of cleanup treatment, after which it is safe to discharge it to the atmosphere. Atmospheric discharge is made in such a way that maximum permissible concentrations are not exceeded. If cleanup has been quite thorough and true gaseous activities are known to be absent, discharge may be through quite short stacks. Particularly at production sites where krypton, xenon, and iodine contribute the bulk of the remaining radioactivity, discharge is made through tall (about 200-ft) stacks.

Much study has been made of meteorological conditions in the immediate environs of most sites and of the dispersion of gases through stacks. Under favorable weather conditions, the plume from an industrial smokestack will rise gradually as it flows down-wind, and gases will be dispersed until only a negligible concentration prevails in the atmosphere. There are, however, several adverse conditions which arise occasionally

to disturb this orderly dispersion of the stack gases, such as (1) aerodynamic influences, (2) unfavorable terrain, (3) meteorological and micrometeorological influences, and (4) the settling of particulate matter.

The patterns of flow which may be exhibited by a chimney plume are as varied as the weather and the topography of the world. The technique of estimating and accounting for the dispersion of gases in the atmosphere is based on the separate theories of Sutton and Bosanquet. These theories have been subjected to approximate experimental verification by Thomas et al.,[30](among others) who studied the dispersal of sulphur dioxide in the environs of four widely separated smelters in western United States (see also page 139).

The process stacks at Hanford stand 200 feet above grade and are constructed of reinforced concrete with a free-standing, stainless-steel liner, of 3 ft 9 in. inside diameter. The liner is capped at the top to cover the annulus between the stack and the liner. A dished head anchored to the base of the stack is welded to the base of the liner. The stainless steel inlet breaching is welded to the stack liner and enters at a 45-degree angle. Sampling points are located at the top and bottom of the stack. Spray rings are installed at three levels for washing down the inside of the liner. Condensate accumulation is drained from the stack liner to a drain tank. It is estimated that approximately 200 curies of krypton-85 per day are diluted by such a stack to the maximum permissible concentration. Partial dilution is achieved when 150 cfm of dissolver gases are mixed with 35,000 cfm of ventilation air. Another 500-fold dilution factor is necessary and this is provided by the stack.

Some sites have found it necessary at one time or another to tie certain of their operations to meteorological control, that is, the meteorologists have dictated whether or not a particular operation could be carried on at a given time. It has sometimes been necessary to wait several days for satisfactory weather conditions.

Filtration

Just as filtration is the most versatile method of gas sampling available, it is also the most widely used process for handling gaseous waste streams in bulk. Paper, sand, and glass fiber filters have been used extensively.

Paper

For almost all research purposes the so-called AEC filter is used. This filter was developed during World War II for the Office of Scientific Research and Development and the Chemical Warfare Service. The filters are made of asbestos-bearing cellulose paper by pleating approximately

250 sq ft of filter medium in 11-in. pleats and fastening these into a 24-in. square plywood frame using a high-softening-point cement. A continuous strip of filter paper is folded back and forth over pleated separator slats. Top and bottom are carefully sealed to prevent leakage. Specifications and characteristics of the AEC filter are given in Table 30.

Table 30

SPECIFICATIONS AND CHARACTERISTICS OF AEC FILTER[a,b]

Sizes (in inches)	Capacities @ 1-in. water pressure drop (cfm)
8 x 8 x $5\frac{7}{8}$ deep	50
20 x 20 x $8\frac{7}{8}$ deep	450
24 x 24 x $8\frac{7}{8}$ deep	500
24 x 24 x 12 deep	800

Filter paper made from specially treated chemical wood pulp and blue Bolivian Asbestos (~15%).

Thickness : 0.035 to 0.045 in.

Ream Weight: 130 pounds

Resistance : 4.3 inches water @ linear air velocity of 28 ft/min

Percentage penetration on smoke tester generating 0.3-micron-diameter particles with air at 28 linear ft/min 0.1% max

[a]Etherington, H. (Ed.), Nuclear Engineering Handbook, McGraw-Hill Book Co., New York (1958), 11-143.

[b]Some manufacturers of this filter are: Cambridge Filter Corporation, Syracuse, New York; Flanders Mill, Riverhead, New York; and Mine Safety Appliance Co., 201 N. Broddock Avenue, Pittsburgh, Pennsylvania.

A high-temperature filter has also been developed, comprising an asbestos-bearing glass-paper medium pleated in the same manner. This paper is inserted in a metal frame with aluminum foil separators. Its performance is comparable to the paper filters even at temperatures

exceeding 250 C. Its use is indicated not only for gas streams at an elevated temperature but also for any installation handling high concentrations of plutonium or other similarly toxic materials where a fire could be catastrophic.

The operating cost of a paper filter installation depends upon the life of the filter, which in turn varies according to conditions in the area in which the filtering system operates. Dusty conditions in an area will shorten the life span of any filter. The average life span of these filters has been shown to be about 18 months. Annual operating costs for a 10,000-cfm system run approximately $65/1000 cfm installed capacity.

Fiberglas

Fiberglas filters are made up of small-diameter (1 to 30-micron) glass fibers packed to a low density (1 to 6 lb/cu ft). They are used in cardboard or metal frames as roughing prefilters, as vessel vents, and in deep packed beds for filtering large volumes of process air before discharge to a stack.

At Hanford considerable study has been made of collection efficiency and flow resistance of glass fibers, using gases containing particles with a geometric-mean particle diameter of 0.2 to 0.7 micron and a dust loading of 0.2 to 0.4 grain per thousand cubic feet.[31] The collection efficiency can be expressed as

$$dF = -\log (1 - \alpha) = CL^a \rho^b V^c$$

and the flow resistance by

$$\Delta P = KL^x \rho^y V^z \quad ,$$

where

dF = logarithmic decontamination factor

ΔP = flow resistance in inches of water

L = bed depth in inches

ρ = packing density, lb/cu ft

V = superficial velocity, ft/min

α = collection efficiency

C,K = proportionality constants

a,b,c,x,y,z = empirical exponents.

The constants were determined in a series of experiments and are given for the more promising glass fibers in Table 31. Throughout the velocity range studied (0 to 75 ft/min) the efficiency for any particular fiber decreased with increasing velocity. This indicates that the predominant mechanism for removal is diffusion. The fact that the exponent on the velocity term in the pressure-drop equation is unity indicates that laminar flow exists throughout the flow range studied (5 to 100 ft/min in this case).

Table 31

EFFICIENCY AND PRESSURE DROP PARAMETERS FOR SOME GLASS FIBERS[a,b]

Fiber Designation	AA	B	55	115K	450
Fiber Diam, microns	1.3	2.5	15	30	115
C	4.6	-	0.085	0.054	-
a	0.8	-	0.9	0.9	-
b	1.0	-	1.1	0.9	-
c	-0.2	-0.25	-0.4	-0.4	-0.5
K	0.082	-	0.00043	0.00020	
x	1.0	-	1.0	1.0	
y	1.5	-	1.6	1.5	
z	1.0	1.0	1.0	1.0	

[a]A. G. Blasewitz and W. C. Schmidt, Treatment of Radioactive Waste Gases, Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva (1958) Vol. 18, 187.

[b]Constants for use with equation on page 87.

Using these data correlations, several dozen glass fiber filters have been designed and put into operation at Hanford. Flow rate requirements have varied from 15 to 125,000 cfm. One such filter, a small one, has been used for vessel vents. The formulation for this unit is shown in Table 32. This filter measured 2.5 ft by 5.5 ft, with an effective cross-sectional area of 12 sq ft. It handled 250 cfm at a superficial vapor velocity of 20 ft/min. The data in Table 32 for efficiency and pressure drop are calculated values. The efficiency of the filter has been measured at >99.9 per cent and is probably close to the calculated value.

Another application is that of decontamination of the ventilation air from chemical processing canyons prior to sending it to the stacks. The original filters installed were sand (see next section) but the newer ones have been made of glass fibers. A filter capable of handling 125,000 cfm with a collection efficiency of 99.9 per cent and a pressure drop of 4 inches of water has been built.

Table 32

GLASS FIBER FILTER[a,b]

Layer	Type Fiber-glas[d]	Packing Density (lb/cu ft)	Bed Depth (in.)	Initial Efficiency[c] (%)	Initial Pressure Drop (in. of water)
Bottom	115K	1.5	12	39	0.10
Second	115K	3.0	10	53	0.24
Third	115K	6.0	20	93	1.34
Clean-up	AA	1.2	1	99.9	2.20
Total	-	-	43	99.99	4.0

[a]Blasewitz, A. G. and Judson, B. F., Chem. Eng. Prog., 51, 1, 6J (January 1955).

[b]A. G. Blasewitz and W. C. Schmidt, Treatment of Radioactive Waste Gases, Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva (1958) Vol. 18, 187.

[c]In equation on page 87 is $\alpha \times 100$

[d]Owens-Corning designations.

It is constructed underground and consists of two parts, a forefilter and a high-efficiency cleanup section. The forefilter, whose purpose is to provide an adequately useful lifetime for the filter, consists of 84 inches of 115K Fiberglas with sufficient area to give a superficial velocity of 50 ft/min. It is packed so that there is a density gradient of from 1.5 to 3.3 lb/cu ft from the top to the bottom of the bed. The high-efficiency section consists of 0.5 inch of B Fiberglas at a density of 1.4 lb/cu ft and 0.5 inch of AA Fiberglas at 1.2 lb/cu ft It has a superficial velocity of 20 ft/min. Air flow is downward through the forefilter (from the lesser to the greater packing densities) and then through the high-efficiency section.

Operating experience with these filters has been good. Two precautions are necessary. The filters must be carefully assembled to prevent bypassing of the gas around the ends of the high-efficiency sections. And the filters will not stand repeated soaking and drying without loss in efficiency.

Small glass fiber prefiltering units are installed in the backs of hoods, where they serve the dual function of evening out the air flow in the hood and of keeping the duct work between the hood and fan clean. They also add materially to the life of the final filter. These units are not particularly efficient for the removal of radioactivity, however, as they are of low density and do not remove the submicron particles.

The use of glass fiber filters for decontaminating evaporator overheads was studied some time ago at Brookhaven.[32] Based on some earlier work by Langmuir[33] and others, three mechanisms of particle removal from the vapor were considered: direct interception, inertial effects, and diffusion. If the particulates are in a size and density range where they may be removed by direct interception or inertial effects, it would be expected that there would be an increase in filtering efficiency with an increase in vapor velocity. On the other hand, a decrease in filtering efficiency with an increase in vapor velocity would indicate that diffusion is the primary mechanism of particulate removal. The optimum characteristics of deep-bed vapor filters for removing submicron particulates are: maximum height, lowest vapor velocity, smallest fiber diameter, and maximum packing density. For the conditions studied,[32] an adequate height and vapor velocity were found to be 3 ft and 1 ft/sec, respectively. The packing density used was 5 lb/cu ft.

Since erosion of the glass fibers by steam is a factor in this use of glass filters, a fiber diameter of not less than 10 μ should be used. It was also noted that unbonded fibers should be used, since the phenolic binder on some fibers could be stripped off by the steam, drained into the still pot, and cause excessive foaming.

Sand

The necessity of installing filters for the large volumes of chemical process canyon exhaust air was first recognized at Hanford in 1947 when radioactive particles were found on the ground around the stack. Large filter beds of graded layers of sand were constructed through which the building exhaust air was routed before release via the stack. Detailed specifications for such a sand filter are given in Table 33.

In the more recently constructed Purex Separation Plant, the filter bed is made of graded mats of glass fibers as described in the previous section. A sand filter designed to handle about 40,000 cfm of air costs about $575,000. The glass fiber filter handles 125,000 cfm and costs about $600,000. Either type of filter retains more than 99.5 per cent of the particulate radioactive material entering the unit, with the glass fiber unit being somewhat more efficient. Either type performs satisfactorily so long as the humidity of the ventilation air is kept well below saturation.

Table 33

SAND FILTER SPECIFICATIONS[a]

Sand Type	No. of Layers from Top	Layers Thickness (in.)	Size[b]	
			Maximum	Minimum
E	1	6	95% through #4 sieve	95% on #8 sieve
G	2	24	Max 2% pass #50 Min 98% pass #20 sieve	30-50% retained on #30 sieve
F	3	12	95% pass #8 sieve	95% retained on #18 or 20 sieve
E	4	6	95% pass #4 sieve	95% retained on #8 sieve
C	5	12	100% pass $\frac{7}{8}$-in. mesh	100% retained on #8 sieve
B	6	12	100% pass $1\frac{3}{4}$-in. mesh	100% retained on $\frac{5}{8}$-in. mesh
A	7	12	100% pass 3-in. mesh	96% retained on 1-in. mesh

[a]Total pressure drop upon installation is about 7.2 in. H_2O.

[b]All sieve sizes refer to U.S. Standard Test Screen. All material should consist of durable grains highly resistant to nitric acid and should be free of silt, organic matter, shale, coal, and acid soluble coatings.

Electrostatic Precipitation

Electrostatic precipitators have been used successfully where gases are heavily dust laden. Their chief value lies in the increased life span they give to filters, since their efficiency for removing low dust concentrations (less than 0.1 grain per 1,000 cu ft and less than 1.0 micron mass median diameter) is seldom more than 95 per cent even under optimum conditions of uniform distribution and low velocity through the unit.[34]

A model used at Oak Ridge for the removal of uranium tetrafluoride dusts from a mixture of gases consisted of two coaxial cylindrical insulated electrodes with a potential difference of 9 kv.[35] The outer cylinder was grounded and the inner negatively charged with respect to the outer. The dust-laden gas flowed between the cylinders after passing through a region of corona discharge. Dust particles in the stream acquire a negative charge

when passing through the corona and are collected on the inner walls of the outer cylinder. The efficiency of the unit under operating conditions was approximately 95 per cent at 230 C, 2.5 psi pressure and a flow of about 1.5 scfm.

Scrubbing

In many process plants or pilot plants it has been found desirable to subject process gases first to a liquid chemical scrubbing operation. Often the primary purpose of this is to remove a chemical constituent which is either itself a hazard or it will complicate further treatment by another method. For instance, in the nitric acid dissolution of uranium, copious amounts of oxides of nitrogen are given off. These are removed by scrubbing the off-gases with a weak caustic solution in a packed tower. This treatment will also remove much of the iodine and ruthenium which may be contained in the gas.

Plants and pilot plants using halogens and interhalogen compounds usually employ scrubbers, not only on process vessels, but on the entire cell ventilation air. The cell ventilation air from the Volatility Pilot Plant at ANL, for instance, is put through a horizontal, cocurrent spray tower in which 6000 cfm of air is scrubbed with 10 per cent potassium hydroxide.(36) Removals of halogens of about 95 per cent are obtained with no pressure drop. The scrubbed gases are dried and filtered through AEC filters before discharge.

Adsorption(31)

As operating experience has been gained, it has been found desirable to add specific processes to separation plant ventilation systems to remove specific isotopes which have been shown to be troublesome. Iodine is a particular case in point. Shortly after the startup of the original separations plant at Hanford in 1945, radioactive iodine-131 was identified on samples of vegetation. The first immediate step taken to reduce this contamination was to increase the cooling period between reactor discharge and fuel-element dissolution and to tighten up meteorological control of operations. A little later, equipment was designed and installed to remove more than 99.5 per cent of the radioiodine from the dissolver off-gas. This equipment consists of a heater to raise the gas temperature to about 200 C and a column containing a porous packing material impregnated with silver nitrate. The heated iodine combines chemically with the silver and is retained on the column. The column is sized to give a superficial velocity of about 1 foot per second at a length-to-diameter ratio of 4. The most suitable packing is unglazed ceramic Berl saddles which have been immersed in 18 to 20 M silver nitrate solution for one minute, drained and then baked at 105 C for 4 hours.

The chemistry of this process is complex. Entering gases contain NO_2, O_2, H_2O, N_2, and $ICl + I_2$ (or $ICl + Cl_2$ with some NOCl depending on whether iodine or chlorine is in excess). The following reactions probably take place:

$$AgNO_3 + \tfrac{1}{2} I_2 \rightleftharpoons AgI + \tfrac{1}{2} O_2 + NO_2$$

$$AgNO_3 + \tfrac{1}{2} I_2 + O_2 \rightleftharpoons AgIO_3 + NO_2$$

$$AgIO_3 \rightleftharpoons AgI + \tfrac{3}{2} O_2 \quad .$$

Under operating conditions the stable product has been shown to be AgI. If, as sometimes happens, the off gases contain NH_3 and H_2, the reactions become even more complicated.

The equilibrium pressure of iodine increases with the square of the NO_2 pressure and directly with the O_2 pressure. High collection efficiencies are favored by high temperatures, but the temperature must be kept below 200 C lest the silver nitrate melt and run off the packing. At temperatures below the condensation temperature for nitric acid vapors (110 C) essentially no iodine removal occurs.

Such silver reactors have operated for about a year with iodine removals of 99.99 per cent. The more generally expected figure is about 99.5 per cent.

When the efficiency of a unit begins to fall, it can be regenerated. This is done by cooling the reactor to 65 C and spraying the packing with 0.04 cu ft of 5M silver nitrate per cu ft of packing. The bed is then heated to 105 C and baked for four hours. The unit is again cooled, sprayed, and baked for 6 hours at 110 C. A reactor may be regenerated about ten times. Regeneration produces a liquid waste which is sent to the underground storage tanks. Satisfactory operation of this equipment has permitted shortening cooling times and relaxing meteorological control. The capital cost of a heater-silver reactor unit runs about $20,000 to $50,000.

PROBLEMS FOR CHAPTER 3

1. Derive the formula given on page 83:

$$C = \frac{C_2 - C_1 e^{-\lambda \Delta t}}{1 - e^{-\lambda \Delta t}}$$

2. A chemical plant processes 1 ton/day of a fuel which has had a burnup of 5000 MWD/ton. Assuming that the irradiation time is 135 days, that 99.5 per cent of the iodine is retained in the plant, and that the allowable iodine discharge is 1 curie per day, what is the minimum cooling time necessary to meet this limit for iodine emission? What volume of air will be required to dilute this quantity of iodine (131) to MPC? Fission yield for I^{131} = 0.029.

CHAPTER 4

HANDLING OF SOLID WASTES

At any site at which radioactivity is handled, a truly remarkable variety of solid material becomes contaminated to a greater or lesser degree. These articles range from ordinary paper, rubber gloves, laboratory glassware and equipment to large pieces of contaminated process equipment. In some cases fairly large buildings have been dismantled and buried. The problem in handling solid wastes has four parts:

(1) collection
(2) volume reduction (if desired)
(3) shipment, and
(4) storage.

Collection Methods

The first problem in handling solid wastes is to develop satisfactory handling methods. The usual practice consists of accumulating all hazardous wastes in suitable containers for shipment, further treatment, or ultimate disposal. In handling these wastes, provision must be made to safeguard personnel from radioactive hazards and to prevent the spread of contamination. Protective clothing is usually required, masks are used when inhalation hazards exist, and radiation surveys are made prior to and during handling. Segregation of the wastes into combustible or noncombustible types as well as by activity level may be practiced.

Low-level wastes are handled directly and generally require no particular precaution. Collection practices for this type of wastes are quite uniform throughout the United States and consist of distributing suitable containers throughout the work areas to receive discarded contaminated material. These containers are plainly marked with brightly colored paint and radiation symbols to distinguish them from ordinary uncontaminated trash cans. They range from cardboard cartons and kitchen-style garbage cans to 55-gal steel drums These containers are picked up routinely on schedule by an assigned crew and buried directly if possible. If not, they are held until sufficient quantity is available to warrant shipping to an alternate site.

At Argonne, the major portion of the solid wastes produced throughout the Laboratory is disposed into a stainless steel container with a slide-operated top containing a removable 1-cu ft fiber drum insert which is used as the standard waste receptacle. In areas where considerable quantities of wastes are produced and the physical dimensions of the wastes are larger than can be accommodated in the standard containers, a $4\frac{1}{2}$-cu ft cardboard container is supplied. For that waste which exceeds a limit of 50 mr/hr, shielded containers are available.

When a container is full or approaches the radiation limit, it is surveyed by Radiation Safety personnel and pertinent information recorded on a Hazardous Disposal Form. In the event that special precautions are to be observed, the container is tagged and the drum insert is marked. The normal collection of these wastes is accomplished by two men from the waste control group. Pickup from some 27 facilities in the Laboratory is maintained on a scheduled basis. It consists of removing the inner fiber drum insert, sealing the lid of the drum if required, and removal from the area. Those wastes requiring shielding are handled essentially in the same way with the exception that they are transferred to shielded units.

For the wastes of higher level, additional types of shielded containers are available. An aluminum map case is utilized for some reactor wastes and most of the remote control cave wastes. A horizontal pot with 8 in. equivalent of lead shielding is utilized for the transfer and disposal of this waste. Thirty- and fifty-gallon drums of wastes can be accommodated in a specially designed, 10-ton coffin.

Volume Reduction

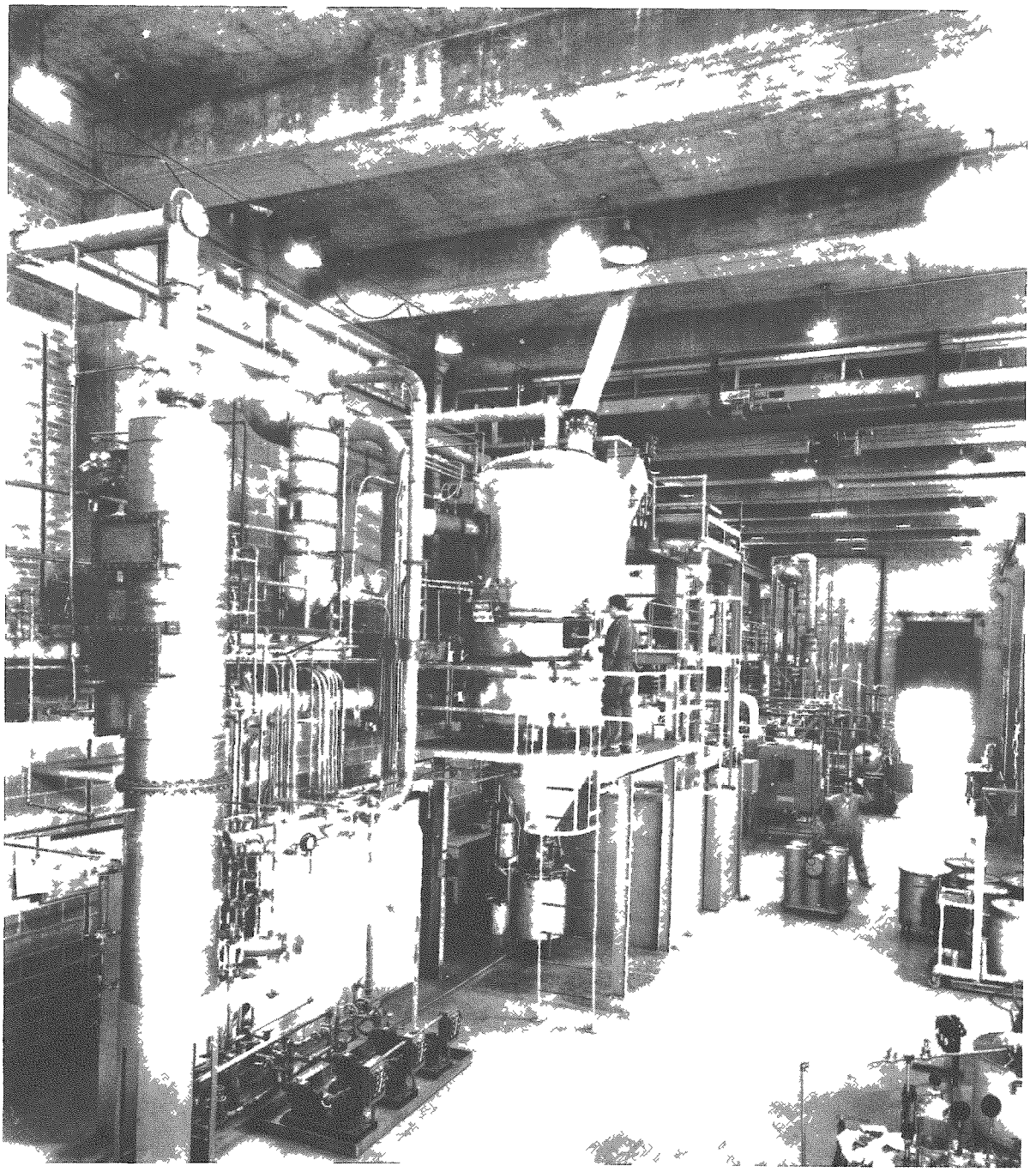
Some sites at which storage space is at a premium have experimented with the segregation and incineration of combustible wastes. Incineration results in a volume reduction of approximately 95 per cent. An incinerator with a burning capacity of 100 cu ft/day was constructed and operated for something over a year at ANL (see Figure 11).[37] The combustion chamber (330 stainless steel) was supplied with air above and below the grates through tuyeres, and the charge was ignited with gas jets. During most of the burning period the combustion was self-supporting. The gas jets were used again during final ashing. Combustion gases were separated from their radioactivity before discharge by a gas-cleanup train consisting of three wet scrubbers in series which removed most of the solid particles greater than 2 microns in diameter. An AEC filter was used for the final cleanup. The following results were obtained from burning 16,000 cu ft of waste:

Volume Reduction	95%
Weight Reduction	70%
Filter Life	Approx 80 hours
Measured Decontamination Factor in Specific Test	10^7.

A careful cost estimate of this operation indicated that even at a site as large as Argonne (3000 total employees at the time) the total volume of combustible waste was not sufficient to permit continuous 24-hour-a-day operation of the incinerator. Under these circumstances the cost of operating the incinerator plus shipment and burial of the resulting ash was a little greater than the cost of shipping the untreated waste.

Figure 11

Active Waste Incinerator[a]



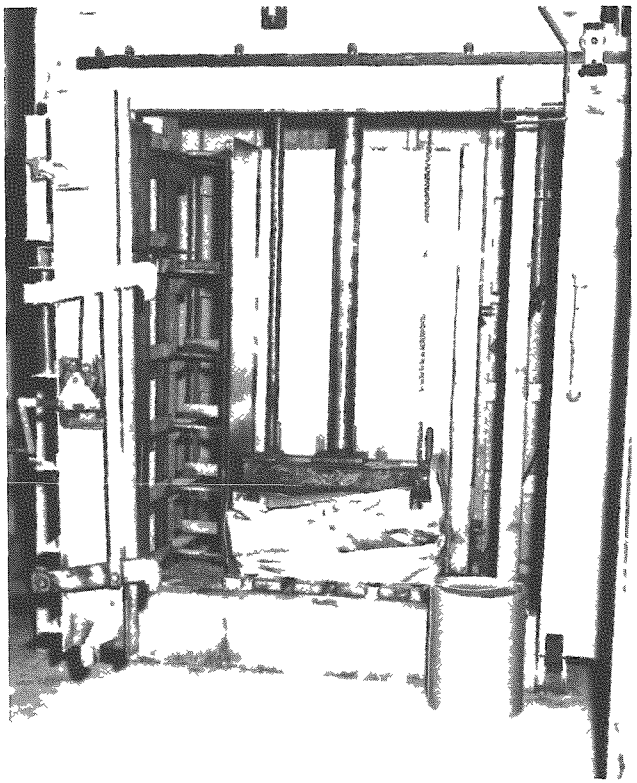
[a] D. C. Hampson, E. H. Hykan, and W. A. Rodger, Basic Operational Report of the Argonne Waste Incinerator, ANL-5067 (February 1953)

Since no system of segregation of wastes is perfect, occasional shutdown of the incinerator was required to remove noncombustible trash. It must also be admitted that an incinerator represents a potential hazard in case highly inflammable material is inadvertently inserted into the collected wastes. Consequently the use of this incinerator was abandoned some years ago. This experience has been corroborated at other sites, although some sites still practice incineration, usually with units smaller and somewhat simpler than this one.

Another technique which may be used to reduce the volume of waste to be stored is baling. Simple paper balers with the addition of a hooded enclosure have been used (see Figure 12). Material that does not exceed 200 mr/hr and/or does not contain any alpha-active material other than natural uranium is baled. Concentration of about one-half of the collected solid waste is accomplished by compression in a commercial hydraulic baling unit. Operating pressures of up to 2100 psi produce an average volume reduction of 4. The baler is ventilated with a 400-cfm exhaust system which includes a two-bank high-efficiency filter unit. At Argonne the baling operation costs about $0.35/cu ft.

Figure 12

Paper Baler for Low-level Waste



Some figures obtained some years ago at KAPL, which indicate the volume reduction of solid waste by baling and incineration, are shown in Table 34. These figures support the thesis that the volume reduction obtained by incineration is not sufficiently great to allow for the economic operation of an incinerator.

Table 34

VOLUME REDUCTION OF SOLID WASTES[a]

	Volume as Collected (cu ft)	Volume after Baling (cu ft)	Volume after Incineration (cu ft)
Paper, Clothes, etc	12,700	1,800	250
Filters	2,200	2,200	1,030
Evaporator Bottoms	1,400	1,400	1,400
Miscellaneous	6,600	6,600	6,600
Total	22,900	12,000	9,280

[a] Larson, R. C. and Simon, R. H., Solid Waste Disposal at KAPL, KAPL-936 (June 1953)

Shipment

Interstate shipment of radioactive material by land or water in the United States is subject to regulation of the Interstate Commerce Commission. The regulations applicable to radioactive materials are published as Title 49, Parts 71-78 of the Code of Federal Regulations. Between revisions, annual supplements are issued and amendments may be published in the daily issues of the Federal Register. These regulations may be obtained from the Supt. of Documents, U.S. Government Printing Office, Washington 25, D.C. The ICC regulations are also published by the Bureau of Explosives(38) and by the American Trucking Association.(39)

Transportation of radioactive material by air (relatively unlikely in the case of wastes) is regulated by the Civil Air Regulations.(40) Some airlines add their own additional restrictions.(41) Transportation of radioactive materials by water is subject to regulation of the U.S. Coast Guard,(42,43) and regulations for mailing such materials are given in the U.S. Postal Guide (44)

Under ICC regulations all radioactive materials are classed as Poison, Class D, and must be labeled for shipment as "Radioactive Materials." There must be no significant radioactive surface contamination on any part

of any shipment. The outside of shipping containers must be at least equivalent to a heavy wooden box or fiberboard box. Emitted radiation must be less than 200 mr/hour at any point on the surface of a package and 10 mr/hour at one meter from the surface.

The present ICC regulations were drafted in 1947 by an ad hoc committee of the National Research Council.(45) At that time isotope shipments were just getting underway. Only two National Bureau of Standards handbooks on radiation protection had appeared - one on medical X rays and the other on radium; both were prewar. Restrictions on the shipment of radioactivity (principally radium) were severe. This came about due to the fact that in 1936 it was postulated that some sensitive X-ray film had been fogged by radium in a mail shipment. Radium was rare and expensive - film was common and widely used. Consequently the movement of radium by common carrier was severely inhibited.

In the drafting of the ICC regulations experience with radium seems to have been extrapolated to all radioisotopes. The maximum amount of any radioisotope which could be shipped without special permission of the Bureau of Explosives was set at 2.7 curies - closely paralleling the existing restrictions on radium. The regulations were later revised to permit shipping up to 300 curies of three industrially used isotopes: cesium-137, cobalt-60, and iridium-192, as sealed sources. The problems (at the time) of shipping large amounts of radioactivity were largely circumvented by a provision which stated that: "Shipments of radioactive materials, made by the Atomic Energy Commission, or under its direction and supervision, which are escorted by personnel specially designated by the Atomic Energy Commission, are exempt from these regulations."(46) It may prove desirable to review these regulations from the standpoint of personnel, rather than film protection.

In an effort to predict something about the problems of shipping radioactivity, the USAEC has had for some time an Ad Hoc Committee on Transportation of Highly Radioactive Materials. Their report has not yet been issued. Some of their preliminary work may be summarized, however. Surveys of current practice showed the existence of over 400 shipping containers for fluid and nonfluid sources of radioactivity (isotope shipping containers not included). The containers differ only slightly in concepts of design and construction. Most of them have an outer and an inner steel cylinder, the annulus being filled with lead for shielding. The inner cylinder, often made of stainless steel, forms the hold for carrying radioactive material. Entrance to the hold is usually gained through one end. Some of the containers have drain lines between the hold and the exterior to determine the existence of leaks and to facilitate decontamination should it be necessary. Containers are provided with trunnions or eye-bolt lifting arrangements.

The cost of the units varies in proportion to the amount of lead and with the intricacy of design. Costs are not available in many cases, but individual containers cost hundreds and even thousands of dollars. The average cost for the larger containers is approximately $1000/ton. A summary of radioactive shipments during 1956 lists 869 container-trips covering over 1,800,000 miles. These represent all or portions of approximately 100 shipments totaling nearly seven million ton-miles. Possibly twice that number were made during 1956 (small shipments of radioisotopes are not included). It is extremely difficult to get at the cost of these shippings since the methods of reporting and accounting from various sites are not comparable. It appears that 15 to 30 cents per ton-mile is the indicated range of transportation costs.

As a particular example, low-level wastes are shipped from the Argonne National Laboratory to Oak Ridge National Laboratory in approximately 10-railroad-car lots at a cost of about $1.50/cu ft. Nearly $1.00 of this cost is attributable to the disposable collection-storage-shipping container. The cost of transporting highly radioactive materials consists mostly of the cost of shipping and handling the shielding containers.

In several cases the weight of radioactive materials being transported was reported. These indicate that the container is some 15 to 20 times heavier than the material contained. The unit cost of shipping in terms of the radioactive material is thus 15 to 20 times more than that indicated. For spent fuel elements, especially those which have been cooled only a short time (e.g., MTR elements shipped to Argonne for use in the Gamma Irradiation Facility after 20 to 30 days cooling), the shield will weigh 500 to 600 times the payload. For such shipments provision must be made for removal of heat during transit.

In testimony before the Joint Committee on Atomic Energy, Bruce[47] estimated the magnitude of the waste shipping problem to be anticipated in the years 1980 and 2000. These results are shown in Table 35. In the study it was assumed that wastes were shipped a distance of 500 miles after 2,000 days of storage, a decay time which is estimated to minimize the combined storage and shipping costs. Under these conditions, the total cost for storage and shipping was about $2 per gal, or 0.05 mill per kilowatt-hour of electricity.

The contemplation of shipping this quantity of radioactive material raises, of course, the question of safety from penetrating radiation and from internal hazard. Penetrating radiation may be taken care of with adequate shielding. Radioactive shipments can cause internal hazards only when they get out of control. Several steps can be taken to reduce and/or minimize internal hazards; these are:

(1) Construct the best possible containers.
(2) Select the physical state of the radioactive material.
(3) Select the chemical form of the radioactive material.
(4) Provide protective measures.

Table 35

ESTIMATED WASTE SHIPPING IN UNITED STATES IN 1980 and 2000[a]

	1980	2000
Number of carriers in transit	505	3,270
Probable carrier weights, tons	6	6
Volume of waste in transit, gallons	227,000	1,470,000
Amount of fission products in transit, curies	1.5×10^8	9.8×10^8

[a] Bruce, F. R., Statement for the Hearings, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 3, page 2353, August, 1959.

The containers are the first line of defense against internal hazards. An ideal container is one which will not rupture or divulge its contents no matter how severe an accident it is involved in. Resistance to shocks, fires or other container-wrecking forces certainly can be designed and built into the packages. The same may be said for the vehicle carrying the container(s).

The physical state of the radioactive material determines its mobility in the event a container loses its integrity. If the radioactive material is solidified as in concrete or incorporated in the structure of a glass or clay, its chances of reaching humans are greatly hindered. Dust may be an ancillary problem, however. If liquids filled the pores of an immobile sponge-like medium, they certainly would be less likely to escape the container to the environment than if they were not absorbed. Altering the physical state may add to the cost of processing; a balance must be struck between cost and protection of health and safety.

The chemical form of the radioactive material determines the rapidity with which it may be absorbed or assimilated by a body. If the material were in a nonassimilable and/or a nonsoluble form and hence relatively nonretainable by a body, the internal hazard would be reduced proportionately. Once again, cost must be balanced against health and safety.

Protective Measures are the last lines of defense against internal hazards. Persons engaged in handling and transporting radioactive material or persons involved in cleanup after accidents should be well trained and provided with adequate health physics coverage and protective equipment.

Storage

The usual method for disposing of low-level solid wastes is by land fill. This consists merely of digging a ditch of sufficient dimensions, dumping the wastes, and backfilling to reduce spread of activity by wind and predatory animals. It is a method which cannot be beaten cost-wise if it can be used. It is most safely practiced in remote areas with favorable geological conditions. At the present time, the AEC operates rather extensive land fills at Oak Ridge National Laboratory, Hanford, and the National Reactor Testing Station in Idaho.

The AEC has announced that it is presently in the process of selecting a site for a national burial ground in the northeast quadrant of the United States. This will be the first attempt to establish a burial ground in one of the more populated areas of the country, although even in this case it seems certain that the site chosen will be somewhat remote.

A considerable quantity of the low-level solid wastes is disposed of into the sea. There are a number of commercial firms which pick up wastes and barge them to designated disposal sites off either coast. Although the amounts of activity which have been placed in the sea off the coast of this country are extremely small, this operation is one which has run into considerable political objection - local, state and international

Technically, there would seem to be no objection to either method, land fill or sea burial, for low-level wastes. The problem of "ultimately" storing or disposing of high-level wastes has not, however, been solved. By one or more of the methods to be covered in Chapter 6, it seems clear that high-level liquid wastes can be reduced to solids if desired. It must be admitted, though, that at the present time it is not clear precisely how the wastes, either liquid or solid, will be finally stored.

In Chapters 7 and 8 various methods which are being considered for "ultimate" disposal of liquids or solids will be described. This discussion will include the discharge of low-level wastes to surface waterways, oceans, or shallow ground formations; the storage of high-level wastes as liquids in tanks for interim periods; the disposal of high-level wastes as liquids to deep wells or to cavities in rock salt; and the storage of high-level solid wastes in geological formations, probably salt.

The problem of removing heat from concentrated high-level wastes during the early years of their storage has been mentioned previously.[28,48] The general conclusion is that solid one-year-old wastes would have to be packaged so that one dimension of the container is less than a yard.[49] Otherwise the interior temperature will be much too high. This implies a multiplicity of containers and thus high packaging cost. It seems logical to assume, therefore, that wastes which are originally produced as liquids will be stored as such for a number of years to facilitate the heat removal problem before they are processed for final disposal.

PROBLEM FOR CHAPTER 4

1. Using the data given in Table 34 and assuming that the cost of shipping to a burial site is $1.50/cu ft, calculate the cost to ship and bury this waste. Compare this to the total cost if that portion which is balable is baled at a cost of $0.10, $0.30, and $0.50 for that operation. Also compare it to the total cost if the conbustible waste is incinerated at a cost of $0.50, $1.00, and $1.50 for that operation.

CHAPTER 5

HANDLING OF LIQUID WASTES

Liquid wastes present by far the most complex of the problems confronting the waste processor. For this reason more research and development work has been done on liquid waste disposal than on any other facet of the problem. Specific processes which have proved valuable in processing liquid wastes include evaporation, acid destruction, precipitation and flocculation, ion exchange, and biological treatment. Each of these, plus separation of individual fission products, will be covered in this chapter. The conversion of liquid wastes to solids will be discussed in Chapter 6.

Evaporation

Evaporation has proved to be exceedingly useful in the processing of radiochemical wastes at both production and research sites. A wide variety of wastes can be evaporated and many types of evaporators have been used. Decontamination factors as high as 10^5 have been achieved in a single effect. Evaporation has been used for preliminary concentration of waste solutions to reduce storage requirements and for complete removal of free liquid to produce an immobile concentrated slurry.

During the early 1950's, an informal cooperative program was carried out at five sites to compare the following types of evaporators:

1. forced circulation with external horizontal heating surface;
2. natural circulation with external vertical heating surface;
3. coil or pot type;
4. vapor compression; and
5. double effect.

Characteristics and operating results of these evaporators are shown in Table 36, and they are described in more detail in subsequent sections.

To reduce entrainment of radioactive contaminants in the condensate, various methods and pieces of equipment have been employed:

1. centrifugal entrainment separators, internal and external types;
2. filtration through a bed of Fiberglas; and
3. reflux.

Table 36

COMPARISON OF VARIOUS PILOT EVAPORATORS[a]

	Knolls	Oak Ridge	Brookhaven	Argonne	Mound
Capacity of equipment	400 gal/hr	285 gal/hr	600 gal/hr	150 gal/hr	100 gal/hr
Type of equipment	forced-feed flash	pot-type removable heating coils	vapor compression	vertical tube circulation	1st effect-vertical tube forced-feed
De-entrainment device	baffled separating column	cyclone-type separator	vapor dome, Fiberglas bed	centrifugal separator, Centrifix scrubber, (reflux if necessary)	2nd effect-vertical tube natural circulation
Typical feed (average activity)	3×10^{-2} μc/cu cm	(β) 2×10^{-2} μc/cu cm	1×10^{-2} μc/cu cm	($\alpha + \beta$) 10^{-3} μc/ml	(β) 10^{-4} μc/cu cm
Typical feed (average solids)	0.3%	8.0%	0.5%	0.2%	3.0%
Overall decontamination factor	10^4-10^5	10^3+	10^6-10^7	10^4-10^5	~10^8
Volume reduction factor	400	15	110	100	32
Slurry (average solids)	70%	70%	65%	20%	60%
Steam efficiency	85%	70%	100%	85%	92.5%

[a]H. Etherington, editor, Nuclear Engineering Handbook, McGraw-Hill Book Co., New York (1958) page 11-105.

Where foam has been a problem, this has been satisfactorily solved by means of:

1. baffles to break foam mechanically;
2. chemicals, e.g., silicones, sulfonated castor oil;
3. pH control; and
4. liquid-level control.

The costs of evaporation are high. The cost figures developed in the aforementioned program varied somewhat with accounting procedures and assumptions made, but approximated $0.10 per gallon. Evaporation is the most expensive of the available processes but also the most reliable. It will handle the widest variety of waste types. It gives the highest degree of decontamination (oftentimes much more than is required for the handling of low-level wastes).

Forced Circulation - External Heating

At Knolls Atomic Power Laboratory, two forced-circulation evaporators were set up.[50] Each had a capacity of 400 gal/hr and consisted of a flash column, circulation pump, heat exchanger, separating column, condensers and receiving tank. This system is shown schematically in Figure 13.[51] The flash columns were 6 ft diameter and 12 ft high. Liquid was circulated by a centrifugal pump at a rate of approximately 650 gal/min. The heat exchanger, which had an overall heat transfer coefficient of 600 Btu/(hr)(sq ft)(F), operated at a pressure slightly greater than atmospheric. The flash column was maintained at 26 in. mercury vacuum. This effectively prevented evaporation and consequent scaling on the heat transfer surfaces. About one per cent of the recirculation stream flashed upon entering the column. The resulting vaporization produced a spray that tended to knock down foam. The overhead from the flash column was passed through baffles to the separating column (6 ft in diameter and 20 ft high). The vapor entered this column tangentially at the bottom, passed through a 12-ft de-entrainment section, and then through 4 bubble-cap trays. The vapor was condensed and received in one of two 5,000-gal tanks.

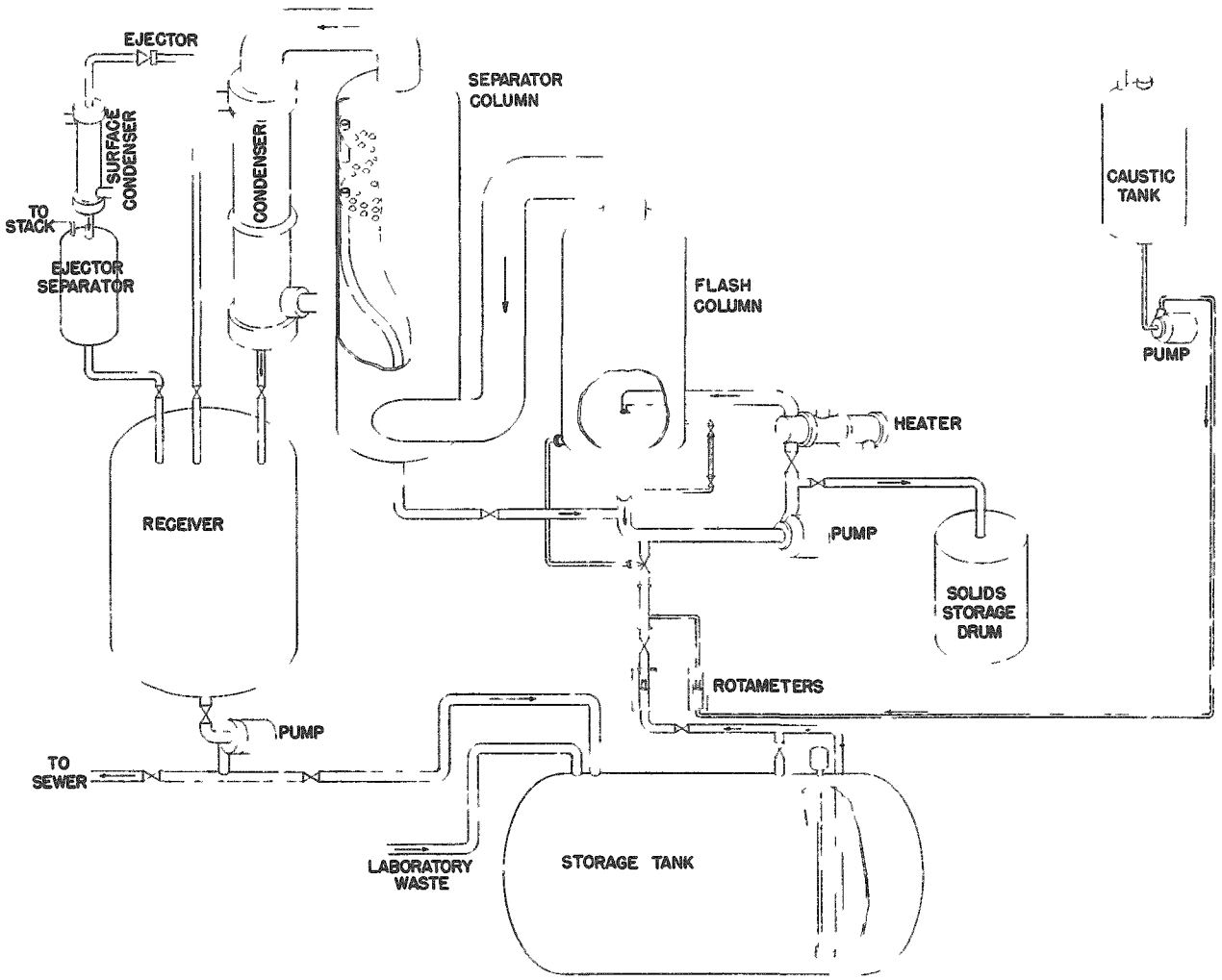
Natural Circulation - External Heating

A natural-circulation evaporator (Struthers-Wells Corp) was set up at Argonne and is still in operation. This type 316 stainless steel unit has a capacity of 150 gal/hr. It consists of a steam chest, separator chamber, overhead condenser, and feed tank. The steam chest is 18 in. in diameter and contains fifty-eight $1\frac{1}{2}$-in. BWG #16 tubes, 5 ft long, that provide 106 sq ft of heat transfer surface. The vapor and entrained liquid leave the steam chest via a centrifugal separating section and enter the

separator, which is 20 in. in diameter and 13 ft high. The entrained liquid is caught in this chamber from which natural circulation returns it to the steam chest. The vapor passes through the separator vapor space and is more completely de-entrained by a Centrifix scrubber and reflux if desired. The vapor is then condensed in a triple-pass surface condenser and returned to a 3000-gal storage tank for monitoring.

Figure 13

Knolls Atomic Power Laboratory Evaporator System[a]



[a]KAPL-594 (August 28, 1951).

Coil Type

Oak Ridge National Laboratory used a pot-type unit heated by 125-psig steam in six removable coils, each coil consisting of two concentric opposite hand helices fabricated from standard pipe. It had a

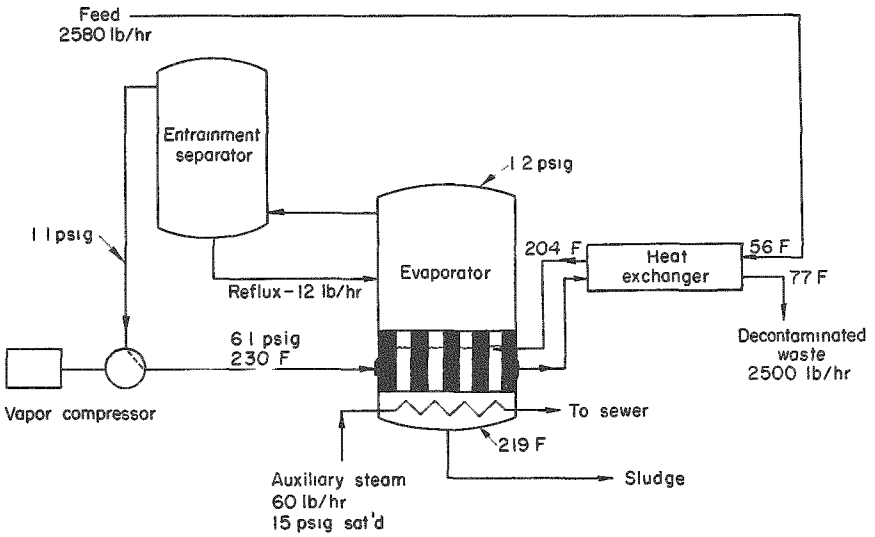
capacity of 285 gal/hr. The evaporator overhead went to a cyclone separator for de-entrainment and then to four stainless steel condensers in parallel. The condensate was continuously monitored. This unit suffered considerably from foaming.

Vapor Compression

At Brookhaven National Laboratory a vapor compression unit (Cleaver-Brooks Mfg Co) was tested. The unit was rated at 85 gal/hr. In this type of evaporator (see Figure 14) the feed is pumped through a heat exchanger, heated by returning distillate and blow-down, and introduced into the hot well of the evaporator. The hot well contents evaporate inside a vertical tube bundle by heat transfer from the vapor condensing on the shell side of the tube bundle. The vapor rises around a baffle and is led to a motor-driven vapor compressor, and finally to the shell side of the hot well. In the compressor the vapor is compressed from 3 to 6 psi, sufficient to raise the condensing temperature about 10 degrees F so that its latent heat may be utilized on the shell side of the evaporator for evaporating the contents of the hot well. The condensate and part of the hot well liquid, called blow-down, are then passed through the heat exchanger and heat is given up to incoming feeds.

Figure 14

Schematic Diagram of Vapor Compression Evaporation[a]



[a]Chem Eng, 62, 194 (1955)

This evaporator was 6 ft high, 2.5 ft in diameter, reduced to 1.8 ft diameter at the hot well section, which was 2.3 ft high. The vertical shell-and-tube evaporator section contained 411 tubes, each of $\frac{3}{4}$-in. outside diameter, with an effective length of 22 ft.

Foam and entrained matter rising with the vapor was partially knocked down by a baffle in the vapor dome, and final de-entrainment was accomplished by filtration through a Fiberglas bed. A similar evaporator with a capacity of 300 gal/hr was later installed at Brookhaven[52] and is still being used.

Double Effect

Mound Laboratory experimented briefly with a double-effect evaporator designed to give overall decontamination factors of approximately 10^8. The first effect was a vertical-tube forced-circulation evaporator, and the second was of the vertical-tube natural-recirculation type. The equipment was designed for processing approximately 100 gal/hr of wastes containing an average of 2 per cent solids. Impingement plates were used in the vapor heads to prevent foaming.

Multiple effect evaporation was also studied at Brookhaven National Laboratory[53,54] Overall decontamination factors of 8×10^6 (10^5 in the first effect) were demonstrated with tracer levels of activity.

Entrainment Separation

The limit to the decontamination factor obtainable by evaporation is determined by the carryover of activity by entrainment and the release of volatile activities. Of the two, entrainment is generally the more important factor. The exact mechanism of entrainment formation is not known, but presumably it is a function of mechanical action of the surface of the boiling liquid and of bubble breakage. Particles formed by either of these mechanisms are carried by the vapor stream if their Stokes' law rate of fall is less than the vapor velocity. Particulate formation, then, primarily should be affected by boil-up rate.

The particulates in the vapor space may be considered to belong to one of three groups:

1. particles whose Stokes' law rate of fall is so small that they are carried along by the vapor velocity and leave the evaporator proper;
2. particles whose Stokes' law rate of fall is roughly equivalent to the vapor velocity (it would be expected that these particles would build up to some equilibrium concentration within the vapor space of the evaporator); and
3. particles whose Stokes' law rate of fall is greater than the vapor velocity. These particles will fall back into the boiling liquid if sufficient free space is provided within the evaporator for them to reach the end of their trajectory. Of course, if

they reach a point where the vapor velocity is sufficiently high while they are still in flight, they may be carried from the system.

Entrainment may then be materially reduced merely by providing sufficient cross section and height above the boiling liquid surface that the larger particles (Group 3) may fall back into the boiling liquid. De-entrainment devices have been utilized to reduce entrainment further and increase the overall decontamination factor. These include cyclones, packed columns, sprays, bubble cap columns, impingement plates, baffles and settling domes. The decontamination contribution of such devices varies from 10 to 100.[55] Deep-bed Fiberglas filters have been used successfully for filtration of vapor (see Chapter 3).

Acid Destruction

Nearly all reactor fuels being chemically processed today are initially dissolved in nitric acid. In addition, the salting agent used in the Purex process is largely nitric acid. Consequently, nitric acid is a major constituent of much of the high-level waste produced, and methods for its destruction have been of interest off and on for over a decade. Chemical and electrochemical methods have been studied.

The reactions of nitric acid with oxalic acid and with formaldehyde are as follows:

Oxalic Acid:

$$2\ HNO_3 + H_2C_2O_4 = 2\ NO_2 + 2\ CO_2 + 2\ H_2O$$

$$2\ HNO_3 + 3\ H_2C_2O_4 = 2\ NO + 6\ CO_2 + 4\ H_2O$$

Formaldehyde:

$4\ HNO_3 + 3\ HCHO = 4\ NO + 3\ CO_2 + 5\ H_2O$ dilute acid 1 N to 8 N

$4\ HNO_3 + HCHO = 4\ NO_2 + CO_2 + 3\ H_2O$ concentrated acid 16 N to 8 N

The reaction with oxalic acid requires heating at 70 to 100 C and is accelerated when catalyzed by potassium permanganate. The formaldehyde reaction goes almost immediately at room temperature with 10 to 16 M nitric acid. Below 5 molar, the solution must be boiled to obtain an almost immediate reaction. The presence of ferric and uranyl nitrates in the concentrations usually found in waste solutions catalyzes the reaction appreciably. In recent work at Hanford, it was found that 95 to 98 per cent of the free

nitric acid in a simulated first-cycle Purex aqueous waste could be destroyed with the use of 1 mole of formaldehyde per four moles of nitric acid.[56] This approaches the limiting theoretical stoichiometry very closely. It was also found that the de-acidified product could be evaporated to about $\frac{1}{3}$ the original waste volume before precipitation was observed. The behavior of fission products during this treatment was established by adding a spike to the feed, and decontamination factors greater than 10^5 were demonstrated.

Hanford has built a small pilot plant unit to test the formaldehyde destruction of nitric acid. The primary reaction vessel is a 4-gal stainless steel pot surmounted by an absorption tower. Several runs have been made. Cold formaldehyde and nitric acid were mixed in the reactor and after a one-half hour delay period the reactor was heated. The maximum pressure generated was 1.7 psig with an initial mixture of 4.6 molar nitric acid and 1.4 molar formaldehyde.[57]

This process has been intriguing to development people for some time because its performance always looks good on paper and small-scale experiments are easily controlled. However, it must be noted that these reaction mixtures may become too vigorous to control and intermediate reaction products may be formed which are temperature and shock sensitive. The process has not yet been put into production use.

Nitric acid may also be destroyed by a number of electrolytic methods. One such process involves electrolytic reduction in alkaline solution; ammonia is produced at the cathode and oxygen at the anode.[58] The process is as follows:

1. Nitric acid waste is neutralized by sodium hydroxide. A gross fission product decontamination factor of 2 to 10 may be obtained by removing the solids formed at this point.
2. The alkaline nitrate waste is electrolyzed in a cell similar to commercial hydrogen-oxygen cells but uncompartmented. Oxygen is scrubbed and released by conventional methods. Some ammonia may be released with the oxygen; however, conditions may be adjusted to keep it in solution. The aqueous product is sodium hydroxide.
3. This caustic solution is evaporated to remove water and also to remove the ammonia which is produced at the previous step. The distillate is dilute ammonium hydroxide that is suitable for direct disposal. The evaporator heel is simply recycled to the neutralization step.

At Hanford still another electrolytic destruction process for nitric acid is being studied in which a two-compartment cell with a Permutit 3142 cation membrane, a stainless steel cathode, and a duriron anode is used.[59]

In a batch electrolysis, 88 per cent of the free nitric acid was destroyed at an overall current efficiency of 57 per cent. The solution volume was decreased by a factor of 3. Neither of these processes have been put into production use either.

Flocculation

Variations of water treatment practice have been studied exhaustively in an attempt to develop a generally useable waste treatment process. It appears that such a system has value in treating large volumes of lightly contaminated wastes, but as a method for handling high-level wastes flocculation is not satisfactory. With the use of a wide range of flocculating agents, overall decontamination factors of about 10 are obtained for mixed fission products. If it is possible to tailor a process for a single radioactive species, much better results can be obtained. Advantages of the process are rather low cost, the ability to handle a wide range of solid content in the feed, and the production of a waste floc volume which is relatively independent of feed solid content. Suitable storage or disposal facilities have to be provided for the radioactive sludge.

A number of water treatment processes were investigated at Oak Ridge to determine their removal efficiencies for strontium-90, cesium-137 and the rare earths from tap water. The characteristic efficiencies of 5 treatment processes are shown in Table 37.(60)

Table 37

REMOVAL OF RADIONUCLIDES FROM WATER BY CONVENTIONAL WATER TREATMENT PROCESSES[a]

Radioisotope	Process Waste Stream Composition (per cent of gross beta 1954-1956)	Per Cent Removal by Treatment Process				
		Chemical[b] Coagulation	Chemical Coagulation Plus 100 ppm Clay	Sand Filtration	Lime-Soda Softening (150 ppm excess)	Phosphate Coagulation (240 ppm dose)
Sr	19.6	3	0-51	4	97.3	97.8
Ce	15.2	91	85-96	-	-	99.9
Trivalent Rare Earths (including Y)	30.4	91	-	87	90.0	-
Cs	29.9	0.5	35-65	50	Not effective	-
Ru	1.9	77	-	-	-	-
	97.0					

[a]K. E. Cowser and R. J. Morton, Treatment Plant for Removal of Radioactive Contaminants from Process Waste Water, Part II: Evaluation of Performance, Hearings, JCAE, 86th Congress of the US, page 547, August, 1959.

[b]Coagulant includes alum, ferrous sulfate or ferric chloride, lime, soda ash or sodium hydroxide, and sodium silicate.

Sand filtration and chemical coagulation with aluminum and iron salts were unsatisfactory for the removal of strontium and cesium. Lime-soda softening and phosphate coagulation were found to be capable or removing more than 90 per cent of the strontium.

Laboratory studies were extended to include actual process wastes at ORNL. Up to 90 per cent removal of gross radioactivity could be obtained with excess lime-soda softening or phosphate coagulation when clay was added for the removal of cesium. Although phosphate coagulation was promising, efficient removals of strontium required accurate control of pH and of the ratio of phosphate and lime dosages.

Some earlier Oak Ridge work reported at Geneva (1955)[61] gave similar results. It was shown that coagulation was most effective for the removal of radioactive ions of valence +3, +4, and +5. Data for a dozen and a half specific isotopes are given in Table 38.

Even earlier data obtained at Argonne similarly showed removals of 90 per cent of mixed fission-product activity from low-level waste for single-stage and up to 97 per cent for two-stage treatments.[62] These data are summarized in Table 39.

Somewhat better results have been shown for the removal of plutonium from laboratory wastes containing up to 25,000 cpm/ml. Wastes containing 100 ppm of fluoride ion, varying amounts of organic and mineral materials, and at a pH which varied from 2 to 13 were studied.[63] Plutonium removals as high as 99.9 per cent were obtained upon using 20 ppm iron (as ferric chloride) and adjusting the pH to 9.5 with lime. The process produces about 20 gallons of wet sludge (3 lb of dry solids) per 100 gallons of waste.

A three-cycle precipitation process was developed through the pilot plant stage at Mound Laboratory.[64] This process was based upon absorption and/or coprecipitation of fission products with ferrous sulfide and ferrous hydroxide-calcium phosphate. In early tests, decontamination factors of 200 to 400 were obtained with cesium; ruthenium was incompletely removed. By increasing the dissolved iron concentration and preconditioning the feed by boiling it with sodium hypochlorite, decontamination factors as high as 4×10^4 were obtainable. Using settling for solids removal and a retention time of one day, volume reduction factors of 15 were found.

A really vast amount of work has been done on this type of processing. A number of representative references are given;[65-71] there are many more. All of these agree generally with the conclusions stated herein. It is difficult to see the justification for very much more work in this area.

Table 38

REMOVAL OF RADIOACTIVE MATERIALS BY CONVENTIONAL WATER TREATMENT PROCESSES[a,b]

Isotope	Removal Range in Per Cent of Initial Activity		
	Chemical Coagulation and Settling	Sand Filtration	Soda-Ash Softening
Cs^{137}-Ba^{137} (Cl)	0-37	10-70	<50
Sr^{89} (Cl)	0-15	1-13	50-95
Ba^{140}-La^{140} (Cl)	1-84	39-99	50-95
Cd^{115} (NO_3)		60-99	50-99
Sc^{46} (Cl)	62-99+	94-99	50-95
Y^{91} (Cl)	1-99+	84-89	50-95
Zr^{95}-Nb^{95} (oxalate complex)	2-99	91-96	50-99+
P^{32} (phosphate)	68-99+		
Cr^{51} (Cl)	0-60		
Mo^{90} (MoO_3)	0-60		
W^{185} (tungstate)	1-96	3-18	<50
Re^{186} (metal)	0-29		
I^{131} (iodide)	0-96		
Ru^{103} (Cl)	43-96		
Pr^{142} (Pr_2O_3)	83-99+		
Ce^{144}-Pr^{144} (Cl)	28-99+		
Pm^{147} (Cl)	4-99+		
Sm^{153} (Sm_2O_3)	44-99+		

[a]Variable chemical dose, coagulants, pH conditions, activity concentrations, and waters.

[b]Straub, C. P., Lacy, W. J. and Morton, R. J., Methods for the Decontamination of Radioactive Liquid Wastes, Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, Switzerland, (1955), Vol 9, p. 24.

Table 39

DECONTAMINATION OF LOW-LEVEL WASTES BY CHEMICAL PRECIPITATION AND SETTLING PROCESSES[a,b]

Chemical Added[c]		Beta Decontamination Factor	
First Stage	Second Stage		
NaOH	(Single-stage Treatment)	3-10	
Lime	(Single-stage Treatment)	5-9	
NaOH, Lime, $FeCl_3$, Active Silica	(Single-stage Treatment)	4-12	
NaOH, Lime, $Al_2(SO_4)_3$, Active Silica	(Single-stage Treatment)	2-10	
NaOH, $FeCl_3$, Active Silica	(Single-stage Treatment)	4-11	
NaOH, Lime, $FeCl_3$	(Single-stage Treatment)	8-12	
NaOH, Lime, $FeCl_3$, Silica Product, Fuller's Earth	(Single-stage Treatment)	5-10	
NaOH, $FeCl_3$, Fuller's Earth, Bentonite	(Single-stage Treatment)	6-21	
NaOH, Lime, Na_3PO_4	(Single-stage Treatment)	12-23	
NaOH, $FeCl_3$	(Single-stage Treatment)	9-15	
		After 1st stage	After 2nd stage
Lime, Na_2CO_3	$FeCl_3$	10-16	21-33
Lime, $FeCl_3$	Lime, $FeCl_3$	7	9
Lime, Na_2CO_3, $FeSO_4$	-	9-12	-
Na_2S, Lime, Na_2CO_3	Lime, $FeCl_3$	5	10
Na_2S, Lime, Na_2CO_3	$FeCl_3$	12-16	18-25

[a]Mixed Fission product activity added at a level of about 100 dpm/ml for single-stage experiments and of about 1000 dpm/ml for two-stage experiments.

[b]Summary Reports Chemical Engineering Division, ANL-4463, p. 80 (1949); ANL-4543, p. 76 (1950); ANL-4720, p. 70 (1951); ANL-4499, p. 85 (1949).

[c]The chemicals tested were applied alone or in various combinations and order of addition. The chemical dosages were in the range normally used in water and industrial waste treatment practice.

Ion Exchange

Certain types of radioactive wastes may be treated by ion exchange. The use of ion exchange resins in connection with maintenance of water quality in reactors has already been discussed. Ion exchange is particularly applicable to aqueous wastes having a total solids content of less than 2500 ppm and preferably less than 1000 ppm. Demonstrated decontamination factors range from 50 for cation resins to about 10^5 for mixed-bed resins, the latter limited by the initial activity of the feed solution used in the experiments. The performance of cationic resins, exhausted cationic resins, and mixed beds is shown in Table 40 for a waste containing 300 ppm total solids.(72)

Table 40

PERFORMANCE OF ION EXCHANGE RESINS[a]

Basis: Waste containing 300 ppm total solids and mixed fission product activity.

Type of Resin	Decontamination Factor	Capacity, Column Throughputs	Volume Concentration Factor
Cation	30	800	130
Exhausted Cation Resin[b]	4-5	>20,000	>3000
Mixed Bed	10^5	55	7

[a]Etherington, H., (Ed) Nuclear Engineering Handbook, McGraw-Hill Book Co., New York (1958), p. 11-96.

[b]Resin exhausted with regard to hardness ions still removes some activity.

Ion exchange resins are high-molecular-weight polymers containing particular ionic groupings as an integral part of the structure. Anion exchangers contain amine groups with an equivalent amount of an anion such as chloride or hydroxyl ion. Cation resins contain phenolic, sulfonic, carboxylic, or phosphonic acid groups with an equivalent amount of a cation such as sodium or hydrogen ion. The polymeric structure is sufficiently cross linked to render it virtually insoluble. A partial list of commercially available resins is given in Table 41.(73)

Table 41

CHARACTERISTICS OF SOME COMMERCIALLY AVAILABLE ION EXCHANGE MATERIALS[a]

Name	Manufacturer	Type	Total Capacity	
			milli-equiv/ g	milli-equiv/ml
Cation Exchangers				
Amberlite IR-100	Rohm and Haas	Phenolic methylene sulfonic	1.75	0.65
Amberlite IR-105	Rohm and Haas	Phenolic methylene sulfonic	2.70	1.00
Dowex 30 (Nalcite MX)	Dow Chemical	Phenolic methylene sulfonic	4.00	1.35
Duolite C-3	Chemical Proc.	Phenolic methylene sulfonic	3.25	1.00
Ionic C-200	Amer. Cyanamid	Phenolic methylene sulfonic	2.70	0.81
Wofatit P	I. G. Farben	Phenolic methylene sulfonic	1.35	0.53
Wofatit K	I. G. Farben	Phenolic methylene sulfonic	2.50	1.00
Wofatit KS	I. G. Farben	Phenolic methylene sulfonic	2.45	0.90
Zeo Karb	Permutit	Sulfonated coal	1.62	0.60
Zeo Rex	Permutit	Phenolic methylene sulfonic	2.70	0.89
Amberlite IR-120	Rohm and Haas	Nuclear sulfonic	4.20	2.15
Dowex 50 (Nalcite HCR)	Dow Chemical	Nuclear sulfonic	4.25	2.20
Alkalex	Research Prod.	Carboxylic	4.95	1.80
Amberlite IRC-50	Rohm and Haas	Carboxylic	10.0	4.20
Duolite CS-100	Chemical Proc.	Carboxylic	3.85	1.11
Permutit 216	Permutit	Carboxylic	5.30	1.70
Wofatit C	I. G. Farben	Carboxylic	7.00	2.50
Montmorillonite		Aluminum silicate	0.8-1.2	
Kaolinite		Aluminum silicate	0.06-0.10	
Glauconite		Aluminum silicate	0.18-0.2	
Permutit	Permutit	Aluminum silicate	1.0-3.0	
Decalso	Permutit	Aluminum silicate		
Zeo Dur	Permutit	Aluminum silicate		
Silica gel		Silicic acid	0.01-0.04	
Anion Exchangers				
Amberlite IR-4B	Rohm and Haas	Weak base	10.0	2.50
Amberlite IR-45	Rohm and Haas	Weak base	6.0	2.0
Amberlite IRA-410	Rohm and Haas	Strong base	2.5	1.0
Amberlite IRA-400	Rohm and Haas	Strong base	2.3	1.00
De Acidite	Permutit	Weak base	9.3	1.5
Duolite A-2	Chemical Proc.	Weak base	7.0	1.20
Duolite A-3	Chemical Proc.	Weak base	6.8	1.10
Ionac A-300	Amer. Cyanamid	Intermediate base	7.4	1.50
Wofatit M	I. G. Farben	Weak base	-	1.20
Alumina		Amphoteric	0.01	
Dowex 2 (Nalcite SAR)	Dow Chemical	Strong base	2.3	0.9
Dowex 1	Dow Chemical	Strong base	2.4	1.0

[a]R. Kunin and R. J. Meyers, Ion Exchange Resins, John Wiley & Sons, Inc., New York (1950).

Contact between a solution and an ion exchanger involves either a batch or column process. In the batch process, the resin is mixed with the solution and agitated continuously or intermittently until equilibrium is reached. After equilibration the resin is removed by filtration. Column processes generally involve a fixed-bed operation. The resin is supported on a porous base while the solution flows through the resin. Moving-bed techniques have also been developed.

The normal fixed-bed cycle comprises several steps:

1. saturation of the exchange bed until breakthrough of ions occurs;
2. elution of the exhausted resin; and
3. rinsing the bed free of regenerant solution.

Contacting a solution with a cation resin will remove the cations and lower the pH. The resin is regenerated by passing acid through it, followed by a water rinse. The use of an anion resin removes only anions, and the effluent in this case becomes more basic.

It is possible to obtain complete deionization by passing the solution through both an anion and a cation bed. Either bed may be used first. Or the resins may be mixed together and used in a single bed (mixed bed). Since the capacity of anion resins is only about half that of cation resins, it is customary to employ about two parts of anion resin to one of cation. To regenerate a mixed bed, it is first necessary to classify the bed by back-washing with water, then to introduce acid into the cation portion of the bed and caustic into the anion portion, and to follow this with a water rinse. Finally the bed is remixed by blowing air through it.

Resin beds should not be less than 30 inches deep, and manufacturers recommend flow rates of about 2 gpm/cu ft. The usual regenerant solutions are 6 N HCl and 4 per cent NaOH.

A number of variables affect the removal of radioactivity by ion exchange resins. These include cross linkage, resin particle size, feed flow rate, pH, concentration of extraneous salts, resin bed composition, bed depth, and temperature.

The degree of resin cross linkage determines its porosity, stability, and solubility.[74] Standard commercial resins, such as 20- to 50-mesh Nalcite HCR, are 8 per cent cross linked. A study which employed Nalcite HCR with 1, 4, 8, and 16 per cent cross linkage for the removal of mixed fission products from tap water which had been adjusted to pH 2.5 showed that the standard resin (8%) gave the best removal of gross activity, cesium, and ruthenium.[75]

Comparison of standard commercial size resins (20- to 60-mesh) with fines (40- to 80-mesh) showed that the total ionic capacity was about the same but that effluents with a little lower activity were obtained with the finer particles. However, for any flow rate there was an expected increase in pressure drop.

It has also been shown that for the removal of gross fission product activity there is no appreciable difference in the decontamination obtained at 2 gal/(min)(cu ft) - the manufacturer's recommended flow rate - and at a rate five times as large. The same study showed that slightly better results were obtained if the feed were adjusted to pH 2.5 (see Table 42).(76)

Table 42

EFFECT OF pH ON REMOVAL OF GROSS BETA ACTIVITY BY CATION EXCHANGE[a]

Feed pH	Before Hardness Breakthrough		After Hardness Breakthrough		Remarks
	Throughput volume, gal/cu ft	Overall integrated DF	Total throughput vol, gal/cu ft	Overall integrated DF	
1.8	5900	10.4	29,000	5.1	Average of 2 runs
2.5	6200	13.6	21,400	5.0	Average of 2 runs
2.5	6100	13.0	260,000	4.1	Flow rate, 10 gal/(min)(cu ft)
2.5	6100	14.5	20,800	5.6	With Alsop filter
4.0	4900	10.7	26,500	3.8	
5.5	4800	2.8	20,400	2.3	Average of 2 runs
8.0	5500	7.0	28,200	2.4	
8.0	6100	6.0	24,300	3.0	With Alsop filter
9.5	5900	18.7	12,300	6.3	With micrometallic filter

Feed solution: Laboratory tap water with added mixed fission product activity (1 to 2 year old). pH adjusted to value shown.

Tap water characteristics: pH 9

total hardness	85 ppm (as $CaCO_3$)
total solids	300 ppm
calcium content	11 ppm
magnesium content	16 ppm

Flow rate: 2 gal/(min)(cu ft)

[a]H. G. Swope and E. Anderson, Cation Exchange Removal of Radioactivity from Wastes, Ind. Eng. Chem., 47, 78 (1955).

Many of the waste disposal applications of ion exchange have to do with activity levels wherein the effect of the radiation upon the resins is negligible. For those uses where significant quantities of radiation are involved, the ability of the resins to withstand radiation damage is of some interest. Parker, et al.,[77] have summarized the results of studies by several investigators[78,79,80] on the effects of beta (absorbed), gamma and X radiation on commercially available organic ion exchange resins (see Table 43).

Table 43

RADIATION DAMAGE TO ION EXCHANGE RESINS[a]

Type of Resin	Per Cent Capacity Loss[b]		
	Wedemeyer[c]	Higgins[d]	Fisher[e]
Strong Acid Nuclear sulfonic Polystyrene			
Dowex 50 (X-8 and X-12)	23	10-20	
Nalcite HCR (X-8)	8		
Amberlite IR-120	9		~12
Permutit Q	2		
Dowex 30		1	
Amberlite IR-105		1	
Amberlite IR-112			~12
Weak Acid-Carboxylic			
Amberlite IRC-50	100		
Permutit H70	100		
Strong Base-Quaternary amine polystyrene			
Dowex 1	44		
Nalcite SAR	37		
Permutit S2	38		
Amberlite IRA-400	42		
Amberlite IRA-410	40		
Weak Base-Weakly Basic amine groups			
Nalcite WBR	20		
Amberlite XE-58	20		
Dowex 3	19		
Amberlite IR-4B	13		
Amberlite IR-45	53		
Permutit Deacidite	3		

[a] G. W. Parker, I. R. Higgins, and J. T. Roberts in Ion Exchange Technology, (F. C. Nachod and J. Schubert, ed) Academic Press, New York (1956), p. 144, Table 16.8.

[b] Per cent of capacity loss per watt-hour (3.8×10^8 r) of energy absorbed per gram of oven-dry resin.

[c] R. E. Wedemeyer, The Stability of Ion Exchange Resins to X-rays, PhD. Thesis, Vanderbilt University (1953).

[d] J. R. Higgins, USAEC, ORNL-1325 (1953).

[e] S. A. Fisher, Effect of Gamma Radiation on Ion Exchange Resins, USAEC, RMO 2528 (1954).

Cobalt-60, tantalum-182, cerium-144, praseodymium-144, and X-ray beams were the radiation sources used. In this tabulation the adsorption of 1 watt-hour of energy by 1 gram of resin (dry basis) is equivalent to 3.8×10^8 r.

As in the case of flocculation, a great deal of work has been done on ion exchange as applied to waste disposal, and the references cited are only representative.

An extension of the ion exchange technique called electrodeionization makes use of synthetic ion exchange membranes of high electrical conductivity. These membranes are permeable to ions of only one charge. Ion exchange membranes are used to divide an electrolytic cell into two or more compartments. In multicompartment units the cells are formed by alternating cation and anion membranes with a cation membrane next to the cathode and an anion membrane adjacent to the anode.(81) Fairly large pilot plant units of this type of equipment have been set up as a part of the saline water program. The British have operated a pilot unit on radioactive wastes. Experimental work has also been done at Argonne National Laboratory and at Oak Ridge National Laboratory. No such unit has been placed in routine operation in connection with waste disposal, however.

Walters et al.(82) have proposed a two-step process for the concentration and electrodeionization of radioactive wastes. The first step - bulk deionization - involves partial decontamination in a multicompartment membrane cell. The second step is the final decontamination of the waste stream by passage through a multicompartment permselective cell containing a mixed bed granular exchanger in the deionization compartments. In the second step, the mixed bed granular exchanger is electrolytically regenerated between permselective membranes. Decontamination factors of 10^5 to 10^6 were obtained from feed solutions of 0.00425 N sodium sulfate containing either long-lived fission products or zirconium-95. Gross activity removal followed closely that of the inactive ions.

Hatch, et al.(83) suggest the use of ion selective membrane (anion membrane) units for pretreatment of nitric acid wastes, i.e., nitric acid removal. Exposure of the membranes to radiation from high-level wastes has not revealed serious limitations with respect to their useful life.

Biological Methods

Removal of radioactivity from liquid wastes by biological methods appears to have limited application for wastes containing low levels of radioactivity. The activated sludge process, trickling filters and sewage oxidation ponds have been investigated. Removals of about 70 to 90 per cent of mixed fission products can be expected by biological treatment.

The degree of removal is essentially independent of feed concentration in the ranges usually encountered in low-level effluents from hospitals, research institutions, and laundries. Iodine and phosphorus removals will depend upon the concentrations of their stable isotopes. Phosphorus removals will vary from 10 to 50 per cent whereas iodine may be expected to vary from zero to 90 per cent. Higher valence ions are more easily removed than those with low valences.(84)

Simulated laundry wastes containing 200-day-old mixed fission products were treated in trickling filters. Ammonium hydroxide and trisodium phosphate were added as supplementary nutrients. About 90 per cent of the gross activity was removed at organic loadings of 250 lb/(acre)(ft)(day). Sludge was produced at a rate of 0.3 lb dry solid/lb BOD removed. Approximate removals of individual isotopes were: cerium, 97 per cent; ruthenium, 80 per cent; strontium, 70 per cent; yttrium, 90 per cent; and zirconium-niobium, 80 per cent. High pH is more effective than low and the effect of recirculation rate was not significant.(85)

The use of a two-stage trickling filter process for the removal of plutonium resulted in removals of 75 to 95 per cent. A sludge volume only $\frac{1}{30}$ that obtained by chemical precipitation treatment of laundry waste was found.(86) Oxidation ponds have given similar results and are useful for handling dilute wastes with variable compositions.(87)

Removal of Specific Fission Products

A considerable amount of work has been done in devising processes for separating specific fission products from waste streams.(88-90) While the industrial use of these isotopes certainly should be, and is being encouraged, it is a fallacy to assume that such utilization will materially change the waste picture. In the first place, even if it were possible to separate each of the hazardous isotopes by degrees sufficient to permit the remainder to be discarded, the separated product would still have to be stored and would become useless from an industrial utilization standpoint long before it had decayed to innocuousness. Secondly, the recovery of specific isotopes for beneficial use is quite a different problem from that of removing them to facilitate waste disposal. For a recovery process, a yield of 90 per cent might be quite satisfactory. If the goal were to permit the residue to be treated as a conventional industrial waste, however, essentially complete removal would be required for all of the elements of concern. For these purposes any plutonium or other transuranic elements must also be removed with comparable efficiency.

One study(91) has concluded that after strontium-90 has been removed by factors of 10^2 to 10^3, the transuranic alpha emitters will become the controlling isotopes from a hazard standpoint. Microscopic losses

ranging to as low as 1 part per 20 billion would have to be simultaneously achieved for the many elements of widely differing chemical characteristics. Achieving this simultaneous high recovery of such chemically diverse products is without precedent in the chemical industry.

The isolation of specific fission products with good recovery and fair decontamination has been demonstrated by the operation of the fission product pilot plant at Oak Ridge, where cesium-137 sources are being made from Purex wastes with an annual output of 200,000 curies of cesium-137.(92)

Conclusion

It can be fairly said that the processes useable in the intermediate treatment of liquid waste discussed herein have had exhaustive development work done upon them. Thoroughly adequate information exists for design and operation of a wide variety of processes and plants. It is difficult to see the justification for additional research on many of them. It would seem that the available information is adequate to allow for the decontamination of large volumes of both low- and high-level waste. More information is needed on some of the methods for reducing wastes to solids, but it would appear that this problem, too, will soon be well in hand.

PROBLEMS FOR CHAPTER 5

1. Assume that the particles entrained into the overhead of an evaporator have the following characteristics:

Group (see page 110)	Fraction of Particles (by number)	Relative Particle Diameter
1	1/3	1
2	1/3	5
3	1/3	10

What decontamination factor may be expected by providing:

a. free space adequate to allow removal of group 3?

b. free space plus baffles (assume that group 3 + 75% of group 2 is removed)?

c. (a) + (b) + Fiberglas filtration which is assumed to remove 50% of group 1 and 99.5% of group 2?

Finally, assume that the evaporation process itself will provide a DF of 10^3. What overall DF is provided by each of the above?

2. Take the waste described in Table 16, page 55, and calculate the concentrations of each significant isotope at 2 years and at 30 years. If the waste were cooled three years, what DF for Sr^{90} would be required so that the volume of water needed to dilute the residual Sr^{90} to tolerance would be just equal to that required to dilute all the others to tolerance?

If the waste were cooled 30 years, what Sr^{90} DF is required for the same condition? For the condition that the volume used for diluting Sr^{90} to tolerance is just equal to that needed for all the others except Cs^{137}?

CHAPTER 6

REDUCTION OF LIQUIDS TO SOLIDS

In this chapter will be discussed methods for reducing liquid wastes to solids. These include adsorption of the activity on natural materials with or without subsequent firing to fix the activity, and various calcining techniques. The former methods are similar in their initial stages to ion exchange, and the latter to evaporation. But whereas ion exchange and evaporation are directed primarily at producing an effluent which is directly discardable, the methods of this chapter are primarily directed at producing a solid which will contain high-level activity in a more easily storable form.

Adsorption on Natural Materials

There has been interest for some time in adsorbing wastes on naturally occurring materials. The adsorption step may be followed by firing at elevated temperature to fix the activity. Some early work along these lines was done by Hatch,[93,94] who had been making use of clays of the montmorillonite group. In this process the waste was passed through a column of extruded clay which adsorbed the radioactivity by ion exchange. The activity was fixed upon the clay by firing it at 1000 C. The capacity for cation exchange has been found to be about 1.2 milliequivalents per gram of clay.

Ten gallons of simulated dilute waste containing 2 curies of strontium-90 were passed in series through three clay columns over a period of twelve days. The effluent was decontaminated by a factor of 2×10^8. In 1953, clay was loaded with activity to 1×10^8 dpm/gram and was then fired at 1000 C for 72 hours to fix the activity. The clay has been subjected to a leaching by sea water ever since. An initial loss to the sea water of 0.08 per cent of the total activity is attributed to surface contamination. Since then, there has been essentially no leaching of activity over a period of 7 years.

This process has been worked on for nearly ten years, but has never been put into production use because the wastes produced by all existing chemical processes are too acid and/or contain too high concentrations of salts to be directly usable. Nitric acid may be removed from Purex wastes by distillation or by electrolysis using permselective membranes prior to the clay-adsorption step. Neither alternative is particularly attractive. Aluminum-bearing wastes must be evaporated and calcined, and the resulting alumina leached with dilute acid to remove the soluble fission products which are then adsorbed on the clay. This work served as the forerunner for most of this type of research but the process does not seem destined to become an important step in waste processing.

Hatch has also been using beds of crushed calcite for removing strontium-90 from 0.05 molar phosphate solutions. After the passage of 1500 column-volumes of solution through a 50-gram test column, decontamination factors ranging from 10^5 to 10^6 are still being obtained.(95) Other carbonate minerals such as siderite ($FeCO_3$), smithsonite ($ZnCO_3$), and rhodochrosite ($MnCO_3$) gave results similar to calcite.

Somewhat similar work is being carried out at Oak Ridge using indigenous conasuaga shale and cesium-137 as tracer. The cesium-137 was adsorbed very effectively on a shale, with a distribution ratio of 500.(96)

At Hanford a method is under consideration for the disposal of aluminum coating wastes by mixing them with sodium silicate to form a stiff semisolid gel.(97) It is proposed that the gel be discharged into a ventilated cavern structure. As water evaporates from the gel, considerable shrinkage would take place. The optimum ratio of sodium silicate to aluminate in the waste is 2 to 5. A large excess of caustic is needed to ensure the formation of a nongranular gel. The cost of disposing wastes in this form is estimated to be $0.10 to $0.14 per gallon, about half that of tank storage.

At Oak Ridge National Laboratory, simulated aluminum nitrate waste solutions have been mixed with limestone, sodium carbonate, and shale to form slurries which were then dried and later fired at various temperatures.(98) It was proposed that in actual practice the mix would be allowed to self-sinter. This has not proved to be feasible, however, and it is understood that this program has been dropped.

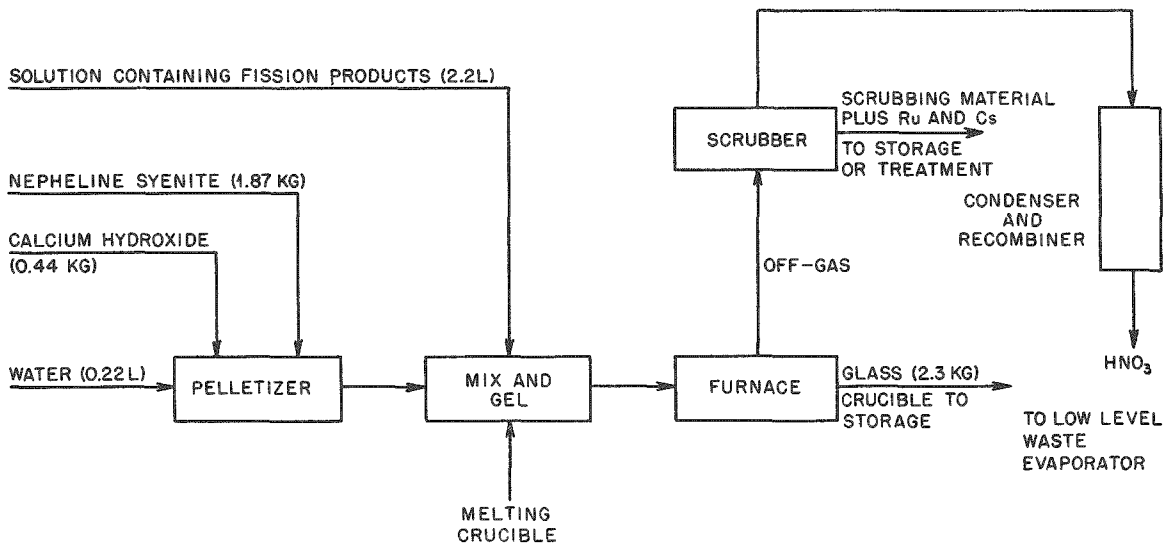
Another use of artificial clays in the fixation of radioactive nuclides has been suggested by Patrick.(99) This method consists of reacting solutions of alkaline silicates or alkaline earth silicates with colloidal gels of aluminum hydroxide and silicic acid. It has been found that the mere mixing of these alkaline silicates with the proper amount of colloidal gels and evaporation to dryness causes the alkaline oxides to combine chemically with the aluminum and silicate to form an insoluble substance exhibiting a pH of 5. The solubility of the compounds has been found to be 10^{-4} to 10^{-5} mole/liter of sodium, cesium, etc. Inasmuch as the fission products often occur as nitrates, the conversion of the latter to silicate form is necessary. The conversion temperatures at which nitrates of sodium, potassium, cesium, and strontium in the presence of colloidal silicates change over to the silicate have been measured. In no case did the temperature exceed 300 C. In other words, the decomposition of the nitrate to the silicate takes place more readily at the lower temperature than the decomposition of the oxide form.

Some of the most encouraging results in developing retentive matrices have been obtained in England[100] and Canada.[101,102] The Canadian process is based on incorporating fission products into a silicate glass matrix made by reacting a concentrated nitric acid waste with nepheline syenite and lime. Nepheline syenite is an igneous rock consisting principally of nepheline ($NaAlSiO_4$) and two feldspars, albite ($Na_2O \cdot Al_2O_3 \cdot 6SiO_2$) and microline ($K_2O \cdot Al_2O_3 \cdot 6SiO_2$). This mixture fuses to form a glass at about 1225 C. Strong acid mixed with powdered nepheline syenite liberates colloidal silica, forming a gel. The viscosity of the mixture is high so that the resulting glass is bubbly, thus exposing a larger surface for leaching. It has been found desirable to use lime as a flux to reduce the fusion point, increase the fluidity, and aid in the incorporation of fission products into the melt.

The process which can be used for nitric acid and nitric acid-aluminum nitrate wastes is shown schematically in Figure 15. Concentrated liquid waste is neutralized with a dry mix of 65 per cent nepheline syenite-35 per cent lime, 1 ml of waste per gm of the dry mix, in a fire clay crucible, and the mixture allowed to gel. The gelled mixture is heated to about 800 C, in order to decompose all the nitrates and to distill off the nitric acid, and then to about 1350 C, to allow the denitrated and dried mixture to fuse. It is anticipated that this process will result in a 50 per cent reduction in volume, from concentrated solution to glass.

Figure 15

Nepheline Syenite Process[a]



[a]Reactor Fuel Processing, 2 (1), p. 41 (January 1959).

If the fission products produced by reactors having a total capacity of 4000 thermal megawatts were incorporated into glass leaching at 3×10^{-7} gram/(sq cm)(day), the cumulative figures for radionuclides leached from the glass would be as shown in Table 44. A rough cost estimate of this process indicates that it should contribute not more than 0.05 mill/kw-hr to the cost of power, assuming the fuel has a burnup of 10,000 Mwd/T. It should be borne in mind that this cost estimate, like so many others, was made by development people. It is the general experience that when reduced to production practice, the actual cost will turn out to be higher by factors of from 2 to 10.

Table 44

CUMULATIVE LEACHED ACTIVITY FROM NEPHELINE SYENITE GLASS CONTAINING FISSION PRODUCTS PRODUCED CONTINUOUSLY AT A CONSTANT RATE[a,d]

Nuclide[b]	Cumulative Leached Material, Curies		
	1 Year	5 Years	25 Years
Sr^{90}[c]	15.2	380	6460
Ru^{106}	30.5	399	400
Cs^{137}	22.6	566	9670
Ce^{144}	73.3	537	540

[a] From 4000 thermal megawatts of installed reactor capacity.

[b] Daughter activities are not included.

[c] The quantity of Sr^{90} in the stored glass after five years would be 1.3×10^{7} curies.

[d] Watson, L.C., et al., The Disposal of Fission Products in Glass, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva (1958). Vol. 18, p. 19.

Another natural material which is useful in adsorbing liquid wastes is vermiculite, an expanded mica. This material takes up liquid much like a sponge. The product is far from unleachable. It is used to immobilize liquid wastes and permit them to be handled as solids. This technique is particularly useful for small laboratory quantities. Concentrated acids generally have to be neutralized before absorption. Some solutions which have been absorbed on vermiculite and the quantity which may be absorbed without evidence of free standing liquid are shown in Table 45.

Table 45

ABSORPTION OF VARIOUS REAGENTS BY VERMICULITE[a]

Reagent	Gallons/cu ft Vermiculite	Remarks
Water	4	
H_2SO_4, Conc	4	Vermiculite darkens
H_2SO_4, 6N	3.5	Viscous mass
H_2SO_4, 3N	4	Viscous mass
HNO_3, Conc	N G	Gas evolved
HNO_3, 6N	N G	Some pressure formed
HNO_3, 3N	3.5	
HCl, Conc	N G	Gas evolved
HCl, 6N	N G	Gas evolved
HCl, 3N	3.5	
H_3PO_4, Conc	N G	Vermiculite partially dissolves
H_3PO_4, 6N	3.5	
H_3PO_4, 3N	-	
$HC_2H_3O_2$, Conc	4	
$HC_2H_3O_2$, 6N	4	
$HC_2H_3O_2$, 3N	-	
HF, 28M	N G	
HF, 15M	N G	Dilute 1:10 with 5% NaOH
HF, 7.5M	N G	Dilute 1:5 with 5% NaOH
Mixture 0.1N HNO_3 0.1N H_2SO_4 0.1N H_3PO_4 1.0N HCl	4	
NaOH (50%)	4	Thick viscous mass formed
NaOH (5%)	3.5	
NH_4OH	4	
Acetone	3.5	Pressure
Ethyl Alcohol	3.5	-
Methyl Alcohol	3.5	Slight pressure
Carbon Tetrachloride	5	Sample dry two months later, vacuum formed.
Kerosene	2.5	Absorbed very quickly. Evaporated on standing. Vacuum formed.

[a]Private Communication from H. G. Swope.

Calcination

There has been continued interest in methods for reducing liquid wastes to solids. While it is true that no great economic advantage will accrue from such a reduction (in fact an additional cost may be incurred), it should be possible to devise ultimate storage methods of solids which are safer than storage of liquids. A number of methods have been tried on a pilot plant scale for immobilizing liquid wastes as solids. Wastes may be simply concentrated until they solidify on cooling and the solids stored in drums. Concentrated wastes have also had Portland cement added so that they set to a solid.[103-105] Wastes are thus immobilized at a volume increase of about two.

A number of methods which combine driving off the water in the waste with thermal decomposition of the contained solids have been under study. These are referred to as calcination processes. They offer considerable promise.

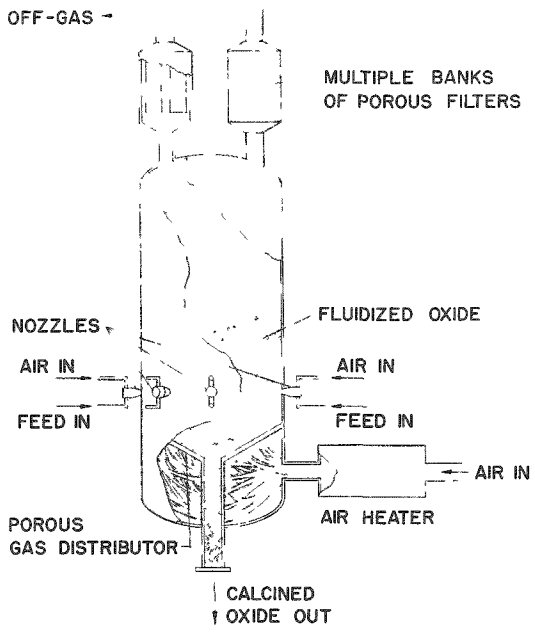
Fluid-bed calcining of wastes is a process which has been under investigation at Argonne National Laboratory and the Idaho Chemical Processing Plant (ICPP) for some time. At the ICPP, a 60-gal/hr pilot plant facility is being built at a cost of $4,000,000 to give this process a thorough test. The process consists of evaporation and calcination of waste solutions to a granular solid consisting of the oxides of the constituent cations in a fluidized bed. A pictorial representation of the fluidized bed calciner is shown in Figure 16. The calciner consists of a cylindrical vessel containing a bed of granular oxides supported on a porous, conical, sintered stainless steel plate. Preheated air is passed through the plate to fluidize the bed, the entire mass behaving much like a vigorously boiling liquid. Waste solution is injected into the bed through pneumatic spray nozzles spaced on the periphery of the reactor in a single horizontal plane. The fluidized bed is heated to 400 to 500 C by means of electric heaters mounted either internally or externally. The reactor is run under a vacuum of 1.3 to 12.5 cm mercury supplied by means of a steam jet exhauster through multiple banks of porous sintered stainless steel filters. These filters are mounted in the top of the reactor to remove entrained oxide particles of greater than 2 to 3 microns. Five filter banks of 2 filters each are used and are manifolded such that blowback of one or more sets of filters may be accomplished simultaneously with off-gas removal through the other sets. Particulate removal may also be accomplished by directing the off-gas through cyclone separators and returning the collected fines to the reactor. The granular product is removed to storage containers by means of a bottom outlet or an overflow pipe.

The volume reduction in converting the nitrate (primarily 2.3M aluminum nitrate) to the oxide is found to range from 6 to 10. The average

bulk density is 0.77 gram/cubic cm. The residual nitrate of the oxide ranges from 2 per cent at a calcination temperature of 320 C to 0.2 per cent at 500 C.

Figure 16

Fluid-Bed Calciner[a]



[a]Jonke, A. A., Petkus, E. J., and Loeding, J. W., A Fluidized Bed Technique for Treatment of Aqueous Nuclear Wastes by Calcination to Oxides, Sanitary Engineering Conference, Cincinnati, Ohio; TID-7517 (December 1955).

Loeding, J. W., et al. Fluidized Bed Conversion of Fuel Processing Wastes to Solids for Disposal, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva (1958). Vol. 18, p. 56; Reactor Fuel Processing, 2 (1) p. 42 (January 1959).

The demonstration pilot plant at Idaho is basically the same, although the off-gas treatment is somewhat different. Gases, upon leaving the calciner, will pass through high-efficiency cyclones, wet scrubbers, adsorber-filters packed with silica gel, and absolute AEC filters before joining the building ventilation air for stack discharge to the atmosphere. Spent silica gel beds will be incorporated with the calcined solids for storage.

Certain components of this proposed off-gas system have been tested at Argonne with diluted ICPP active waste. Results of five different runs are shown in Table 46. At a calcination temperature of 400 C, a ruthenium decontamination factor across the calciner, scrubber, and adsorber of about 10^3 has been obtained. At 500 C these ruthenium decontamination factors have been in excess of 10^4 for the best runs. Nonvolatile fission products have been removed by factors of greater than 10^4. The results are approaching satisfactory limits.(106-108)

While most of the work at Argonne has been with aluminum nitrate-bearing wastes of interest to Idaho, more recent work has also been done using Hanford Purex low-acid waste solutions.

Table 46

PERFORMANCE OF WASTE CALCINER OFF-GAS SYSTEM

(Run Time: 6 hr)

Temp, C	Dilution of ICPP Waste	Feed Activity, Counts/Min		Decontamination Factors, Avg							
				Ruthenium				Nonvolatile Fission Products			
		Gross γ	Ruthenium γ	Across Calciner	Across Scrubber	Across Adsorber	Overall	Across Calciner	Across Scrubber	Across Adsorber	Overall
400	250:1	1.2×10^6	5.0×10^4	1.1	1.8	350	6.6×10^2	390	1.1	81	3.2×10^4
500	250:1	1.1×10^6	4.7×10^4	3.8	b	2900	1.1×10^4	114	3.7	340	1.4×10^5
400	90:1	3.0×10^6	1.3×10^5	1.1	1.4	4200	6.2×10^3	114	3.7	340	1.4×10^5
500	90:1	3.0×10^6	1.3×10^5	3.3	b	390	1.3×10^3	156	3.7	135	5.5×10^4
400[a]	100:1	2.9×10^6	1.1×10^5	1.1	1.3	700	1.0×10^3	52	10.0	101	5.3×10^4

[a] Eighty-five-hour run.

[b] No contamination detectable.

Results to date cover about 200 hours of operation and are quite promising. A granular free-flowing product has resulted from calcining operations performed at 500 C.[109]

Another type of spray calcination is being studied at Hanford where an 8-in. spray, agitated-trough calciner is being tested. Using as feed (1) an acid-killed (formaldehyde-treated) first-cycle aqueous waste, (2) a similar feed with phosphate and borate addition, and (3) a simulated ICPP aluminum nitrate waste solution, encouraging results have been obtained. The effect of calcination time on the volatilization and leaching of fission products from Purex first-cycled aqueous waste was studied at 400 and 800 C. The fraction volatilized did not change markedly for heating times ranging from 5 minutes to 24 hours. For 400 C calcination, the leachability of the residue was also not affected, but at 800 C the leachability decreased greatly from 73 per cent leached after 5 minutes heating to only 3 per cent after 24 hours.

It has been found that the addition of sugar helps to destroy the nitrate. A run made with a slurry feed containing 250 grams/liter sugar gave smooth operation with little or no buildup of dust on the walls. The product powder, predominantly sodium carbonate, had a bulk density of 0.85 gram/cc and fused at 860 C to a compact glassy melt with a density of 2.3 grams/cc. Residual nitrate was low, less than 0.0015 per cent.[57,109,110]

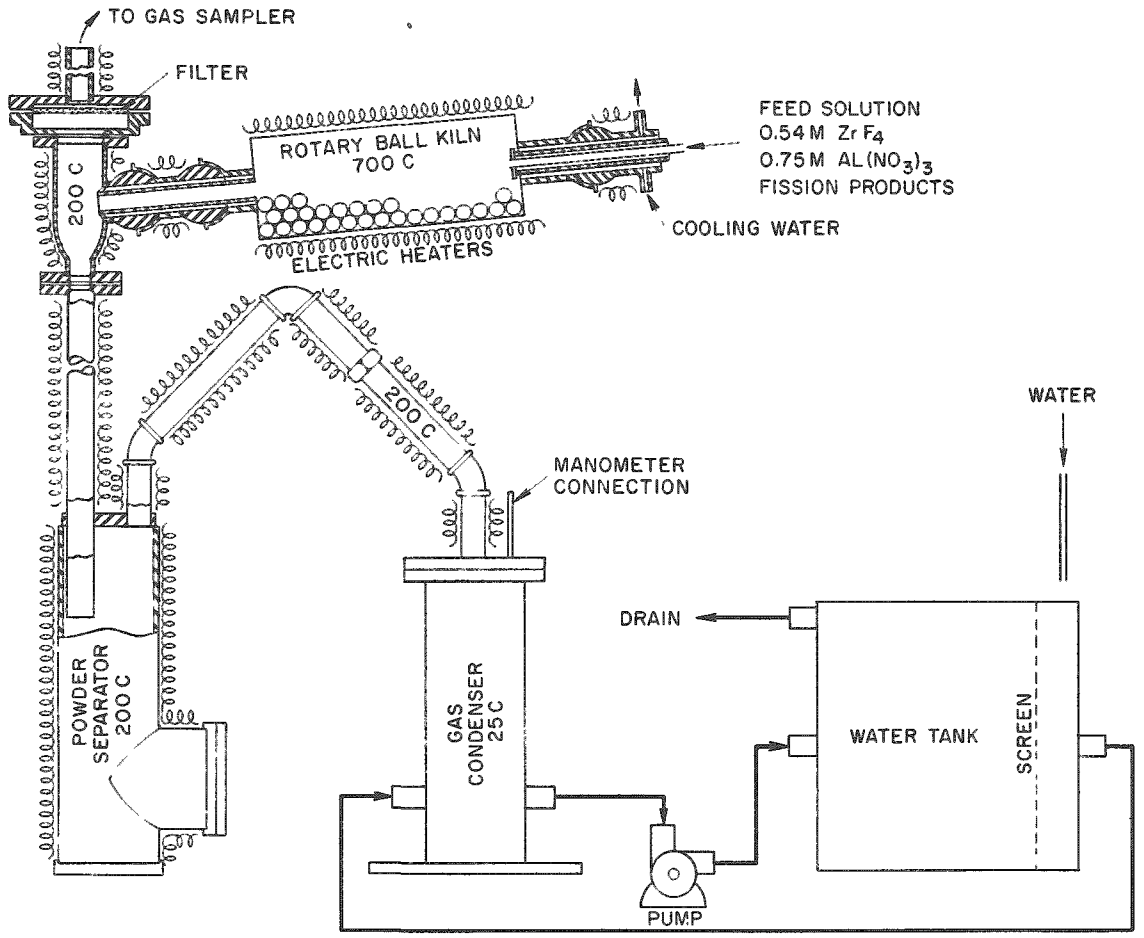
Still a third calcining method is the rotary ball kiln calcination process being studied at Brookhaven.[111] A schematic of this equipment is shown in Figure 17. At least four alloys have been shown to be probably suitable as construction material for the high-temperature calciner vessel: Illium G, Inconel X, Haynes 25, and Nionel. It is concluded that calcination in rotary ball kilns appears to be practical with respect to product quality, mechanical design, off-gas volume, and dust carryover.

None of these calcining methods produces directly a solid which even approaches nonleachability, but they are some of the most convenient methods for reduction of liquids to solids. Some efforts have been made to find ways of incorporating these calcines into glasses. A recent MIT report[112] describes attempts to incorporate calcines from Darex processing of stainless steel-uranium fuel elements into vitreous matrices. The calcine is mixed with lime and silica and fired in a clay crucible.

It would appear that calcining methods are best used merely as ways of removing liquid and decomposing salts, and that vitrification methods which start by mixing raw or concentrated waste with the chosen fluxes are likely to prove the more desirable.

Figure 17

Rotary Ball Kiln Calciner[a]



[a]R. F. Domish, et al., Calcination of High-level Atomic Wastes as a Step in Ultimate Disposal, BNL-535 (December 1958).

PROBLEM FOR CHAPTER 6

1. Assume that at some time in the future there will be 10^6 thermal megawatts of installed reactor capacity. If the waste from these reactors is incorporated into nepheline syenite, what will be the expected annual leach rate for strontium-90 after 1 year and after 100 years? What volume of water would be required in each case to dilute this quantity of Sr^{90} to tolerance? Assume the waste is concentrated to 100 gallons/ton (sp gr 1.2) before mixing with the nepheline syenite and that the fuel has an average burnup of 10,000 Mwd/ton.

CHAPTER 7

RELEASE OF WASTES TO THE ENVIRONMENT

In previous chapters the various methods whereby wastes are either stored or released to the environment have been discussed from the standpoint of waste management at the site. In this chapter the release methods will be considered from the standpoint of their possible effect on the environment.

Assuming that a method for destroying the radioactivity of wastes is never found, it will always be necessary to disperse some small quantities of radioactivity to the environment, meanwhile maintaining the bulk of the activity under strict control. This is true not only because no process of containment can ever be expected to be absolute, but also because processing of all the large-volume, low level and suspect wastes will guarantee absolutely that the development of an economic nuclear power economy will be impossible. It is necessary, therefore, that the diluting power of the environment be used - but it must be used intelligently.

Dispersal takes the form of discharge of gases into the atmosphere and of solids or liquids to the ground, the oceans, or streams, so that mixing with the dispersal medium takes place. Control of the radioactivity is lost as it enters into a complex web of ecological cycles. Cognizance must be taken of the fact that many biological processes result in reconcentration of the activity, sometimes by large factors. Not all of the possible routes are known, let alone understood. The total radiation exposure received by man is a summation of many contributions. These facts make the determination of permissible discharge levels and evaluation of the processes and practices most difficult.

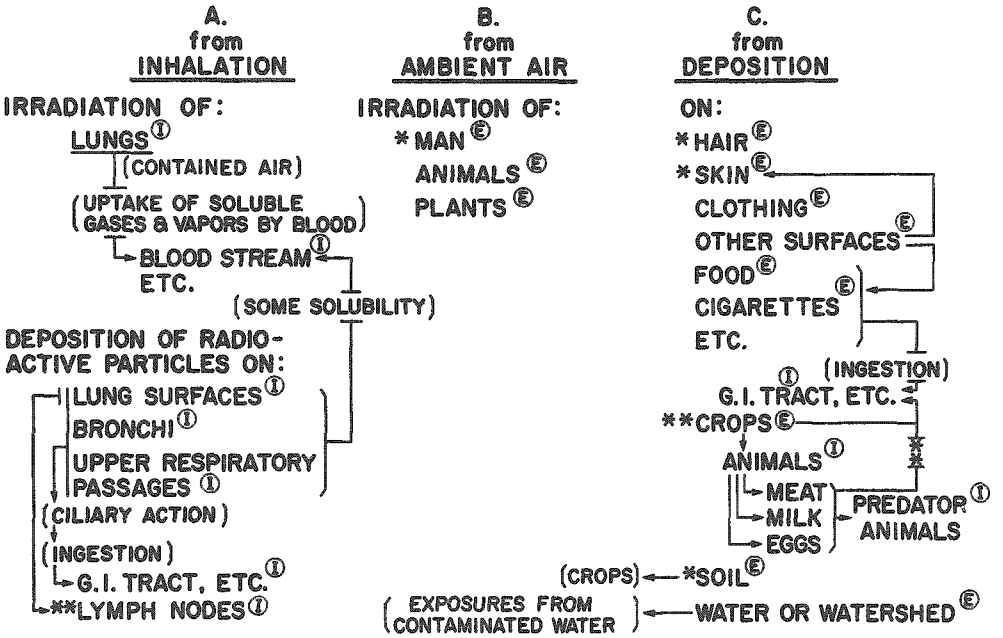
Parker[113] has prepared a series of charts showing some of the possible ways in which radioactivity in the environment may get back to man. These are shown for the atmosphere in Figure 18, for surface waterways and the oceans in Figure 19, and for the ground in Figure 20. It is beyond the scope of this discussion to consider in detail the biological factors involved, but for each of the dispersal media some examples of problems encountered will be given.

Release to the Atmosphere

When radioactivity is released into the atmosphere, the amount which returns to man is a function not only of the dilution and transport in the atmosphere but also of the manner and place of deposition and the relation thereof to a receptor. Since control over potential contamination must be exercised at the source, the meteorological problem includes assessing the probability that material will be carried from the source to a receptor and, if it is, to estimate how much it will be diluted before it gets there.

Figure 18

Exposure Pathways for Radioactive Wastes in the Atmosphere[a,b]

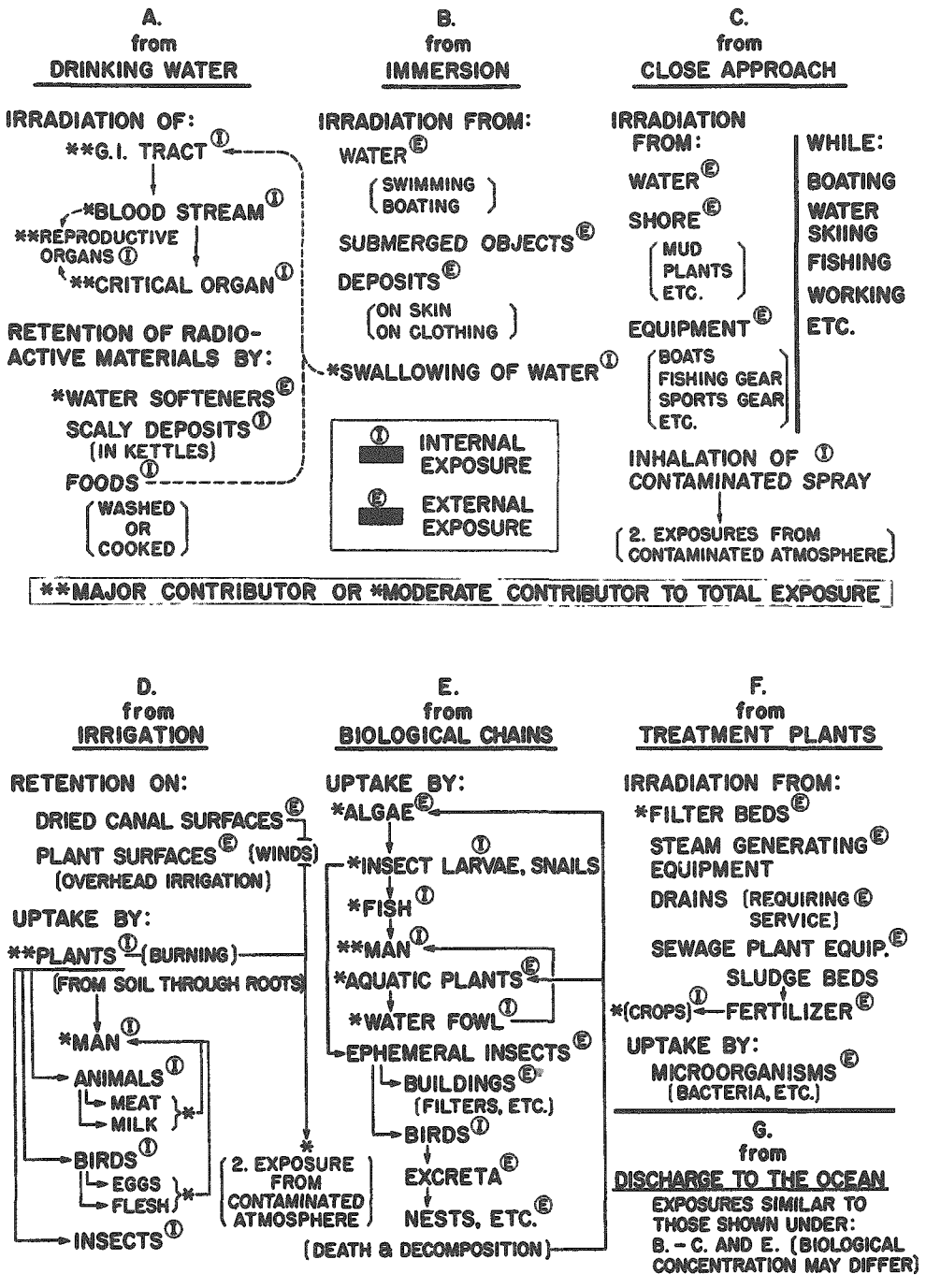


[a]H. M. Parker, Testimony before Joint Committee on Atomic Energy, Hearings on Industrial Radioactive Waste Disposal, Volume 3, pages 2356-2372, August, 1959.

[b]For legend see Figure 19.

Figure 19

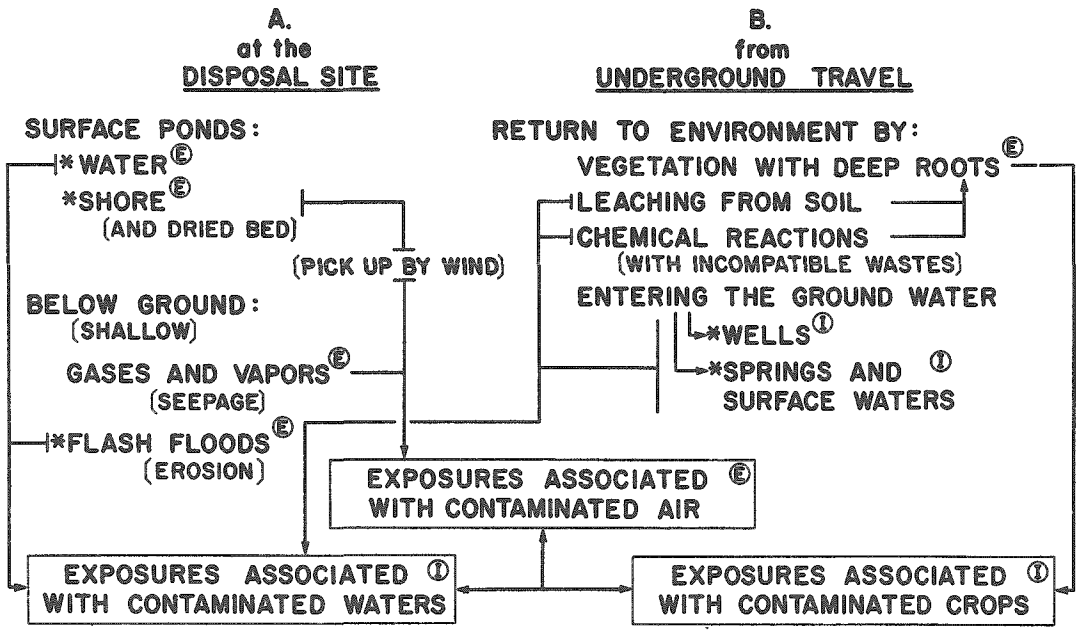
Exposure Pathways for Radioactive Wastes in Surface Waterways[a]



[a]H. M. Parker, Testimony before Joint Committee on Atomic Energy, Hearings on Industrial Radioactive Waste Disposal, Volume 3, pages 2356-2372, August, 1959.

Figure 20

Exposure Pathways for Radioactive Wastes in the Ground[a,b]



[a]H. M. Parker, Testimony before Joint Committee on Atomic Energy, Hearings on Industrial Radioactive Waste Disposal, Volume 3, pages 2356-2372, August, 1959.

[b]For legend see Figure 19.

These problems are complicated and submit only to statistical solutions. The answers obtained are therefore always subject to considerable uncertainty.

The concentration of activity at a fixed point downwind from a stack shows extreme variability due to variability in wind direction, which causes the plume to wander, and to variability in the rate at which dilution takes place. Prediction of concentration at a particular point at a given time is impossible, but prediction of peak concentrations and long-term average concentrations is not quite so hopeless. These concentrations will also vary with terrain and meteorological condition. The lot of the meteorologist is a difficult one at best.

Extensive studies have been made at Hanford(114) which permit prediction of maximum ground level concentrations as a function of wind speed and atmospheric stability. Long-term average concentrations have been predicted for some time from diffusion theory. Hanford data show that measured concentrations are higher than those predicted by theory

and that the greater is the distance from the source, the greater is the error. These empirically derived results are strictly applicable only to Hanford, but they can serve as a guide for other sites.

Studies are also being carried out on deposition of airborne contaminants and on erosion of previously deposited material. These factors are important because the limiting condition for setting maximum permissible concentrations is sometimes the rate at which plants and animals take up waste material from the ground. For example, the permissible level of iodine-131 in pasture grass is 1×10^{-4} μc/gram, based upon a limiting dose of 1 r/day to the thyroid. The concentration of iodine in air necessary to give this concentration in grass is 1×10^{-12} μc/cc, which is a factor of 3000 less than that given for continuous breathing.

Release to Surface Waterways

Surface streams represent an important asset to any nation, both as sources of water and as places into which low-level industrial wastes of many types can be placed. Streams not only represent a source of dilution water, but they also offer some degree of self-purification. This is true for radioactivity as well as for more common types of wastes. The degree of self-purification available varies considerably with the location and the season, but under favorable circumstances it is substantial, and considerable quantities of low-level wastes may be discharged to inland waterways. In conjunction with physical dispersion and dilution which occur due to mixing, there are also chemical and biochemical actions brought about by aquatic plants and animals, by sunlight, and by the hydraulic agitation of the stream. These mechanisms work to purify the water by chemical precipitation, agglomeration, settling action, and biological uptake of organisms. In addition, the stream provides time for radioactive decay and free transportation to the ocean. Storage of long-lived radioisotopes deposited on a stream bed must be regarded, however, as precarious and unreliable. Sudden release of activity from bottom storage can occur during unusual stream conditions such as floods or droughts.(115)

The effect of discharge to streams upon aquatic life is particularly important. This subject has been studied extensively at Hanford.(116) Reactor effluent contains many different radioisotopes and the contribution which each makes to the overall exposure differs with water use, e g., drinking, swimming, boating, industry, irrigation, or production of fish and wildlife Some radioisotopes may be picked up by aquatic forms and become concentrated in certain tissues. The amount to which radiophosphorus and some other isotopes of biological importance are concentrated by Columbia River fish is shown in Table 47. Over 90 per cent of the activity in fish is radiophosphorus, which deposits principally in the hard tissues, such as the bone and scales. No effect on fish population attributable to reactor operation has been discerned. Extensive tests have demonstrated that the concentration of radioactivity in the Columbia River is well below the toxic

level. The limiting factor is not due to radioelements at all, but rather to dichromate which is added to inhibit corrosion. There also appears to be no problem associated with the use of reactor effluent water for irrigation Crops of barley have been grown on experimental plots at Hanford irrigated with the undiluted effluent for 7 years without any significant effect being noticed.

Table 47

ISOTOPES CONCENTRATED SIGNIFICANTLY BY COLUMBIA RIVER FISH[a]

Isotope		Concentration Factor
In decreasing order of abundance in the fish:		If the amount of the isotope in a gram of water is 1, the amount found in a gram of fish (minnows) during the late summer months will be
	Radioactive Half-life	
P^{32}	14 days	100,000 (may reach over 1,000,000 for algae)
Na^{24}	15 hours	100 to 1,000
Zn^{65}	250 days	1,000 to 10,000
Cr^{51}	28 days	10
As^{76}	27 hours	100
Cu^{64}	13 hours	10
Sr^{90}	28 years	1,000

[a]Foster, R F., Research and Development Programs, Related to the Disposal of Reactor Effluent to the Columbia River, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, August, 1959

At the present time the U. S. Public Health Service is involved in determining the fate of radioactivity in streams below such installations as Knolls Atomic Power Laboratory, Pressurized Water Reactor, and Savannah River Project. This project is directed at determining the factors involved in the removal of specific isotopes, either soluble or insoluble, in the stream environment.

Disposal to Ground

Mention has been made several times of the return of low-level wastes to the environment by percolating them into the earth at or near the surface. Such wastes then work their way slowly into the ground water, leaving all or part of the contained radioactivity held either chemically or physically on the soil. Considerable use has been made of this method of disposal at Hanford, Oak Ridge, Savannah River and Chalk River.

The operation of this method of disposal is very dependent upon the geology and hydrology of the particular site.(117) Table 48 summarizes the main hydrological factors affecting the movement of wastes from various sites. The depth to the water table varies considerably, from 200 to 600 feet at Hanford and Idaho to 20 feet at Oak Ridge and Savannah River. Also, the character of the overburden soil varies markedly in composition and in permeability. One of the most important characteristics of ground is its ion exchange capacity. The ion exchange capacities at the various AEC sites range from 5 to 300 grams of exchangeable sodium and calcium per cubic foot of soil. Even the smaller quantity is immense compared to the quantity of radioactive ion likely to be put into the ground.

Brown, et al.,(118) surveyed the experience in ground disposal at three major production sites. Twelve years of practical experience in the controlled disposal of wastes to the ground at the Hanford works, 7 at Oak Ridge, and 4 at Savannah River, have demonstrated the feasibility, safety and economy of the disposal of at least limited volumes of some types of liquid wastes at shallow depth. The total volume and activity which have been disposed to ground at the three sites is shown in Table 49. In addition to 1.2×10^{10} liters of radioactive wastes discharged to ground, Hanford has also discharged more than 1×10^{11} liters of uncontaminated process cooling water to open swamps.

Ocean Disposal

The use of the ocean for the disposal of radioactive wastes has been considered from time to time. At present there is little agitation for the use of even the ocean deeps for the disposal of high-level wastes, although some oceanographers are interested in doing additional research work to determine whether or not this would be feasible The use of the coastal waters and tidal estuaries for the disposal of low-leval wastes is of considerable interest, however.(119,120)

Table 48

HYDROLOGICAL AND GEOLOGICAL FACTORS OF GROUND DISPOSAL AT VARIOUS AEC SITES[a]

	Hanford	Reactor Testing Station, Idaho	Oak Ridge	Savannah River
Main formations involved in waste disposal	(a) Gravel and sand (glaciofluviatile formation) (b) Gravel, sand and silt (Ringold formation)	Basalt	Shale (Conasuaga formation)	(a) Fine sand and silt (formations of Tertiary Age) (b) Gravel and sand (Tuscaloosa formation)
Depth to water table (feet)	About 200	About 200 to 600	About 20	About 20
Permeable sediments above water table (feet)	About 200	About 100 (alluvial sediment overlying basalt)	About 20	About 20
Permeability (gallons per day per square foot)	(a) 10,000 to 60,000 (b) About 400	Erratic; about 10,000 to 20,000 (?).	Low; 1 to 10	(a) 1 to 100 in various beds (b) About 1,000
Average rate of travel (feet per day)	(a) 15 (b) About 1	20 (?)	About 1	About 1
Place of emergence at surface	Columbia River	Snake River	Small creeks, thence to Clinch River	(a) Small Creeks, thence to Savannah River (b) Savannah River
Distance to point of emergence	About 10 miles	About 100 miles	About 400 feet	(a) About 500 feet (b) A few miles

[a]Theis, C. V., The Disposal of Low- and Intermediate-level Radioactive Wastes to the Ground, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, August, 1959.

Table 49

GROUND DISPOSAL AT THREE MAJOR U. S. PRODUCTION SITES[a]

	Cumulative Total Volume, Liters	Cumulative Total Gross Beta-emitter Activity, Curies
Savannah River	5.0×10^8	2.4×10^2[b]
Oak Ridge	2.6×10^7	1.0×10^5
Hanford	1.2×10^{10}	2.4×10^6

[a]Brown, R. E., et al., The Storage of High-level Radioactive Wastes to the Ground, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva (1958), Vol. 18, p. 95.

[b]Not including 2300 curies of I^{131}.

The fate of the radioactive material introduced into the marine environment depends upon the following considerations: (1) the physical and chemical form in which the material occurs, (2) initial mechanical dilution of the waste by the receiving water, (3) advection of the wastes away from the source region by currents and simultaneous turbulent diffusion, (4) uptake of the activity by suspended silt and bottom sediments which removes some of the material from the water and restricts further dispersion, and (5) concentration of activity by various parts of the biota, including shellfish and finfish important to man as a source of food. Some important fission and corrosion products are concentrated by certain marine organisms by factors of 100 to 10,000. Research is now in progress on each of these basic phenomena in restricted waterways and coastal waters.

There are three separate areas of the sea to be considered in waste disposal: estuaries, coastal waters, and deep sea. In the United States the estuaries are the kind of which the bottom water moves inshore, so there would be a tendency for radioactive waste materials to remain in the estuary and not be dispersed. These harbors and inshore waters in general also contain many bottom-living animals which concentrate radioactive materials, so the dispersal of radioactive materials from the estuaries takes place more slowly than the dispersal of the water itself.

In coastal waters, i.e., waters within 100 miles of the coast, there are two different situations - the shallow coastal waters of the Gulf and Atlantic coasts, and the relatively deep waters which exist quite close to shore off the Pacific coast. Finally, there is the deep sea, which lies below a virtual screen or curtain called the thermocline. It is isolated from the

rest of the ocean and very few of man's food organisms come directly or indirectly from it. Although there is no clear theory on this, it is generally believed, and there is quite a bit of evidence for believing, that the deep waters are isolated from the surface layer for periods of the order of hundreds to thousands of years. If this is true, then the use of ocean depths for disposal of some high-level waste may be possible, but other methods of disposal appear more promising.

The National Academy of Sciences has several working committees established to prepare recommendations on specific disposal problems of concern to the AEC. The reports of two of these groups have been completed and are included in the record of the Hearings on Industrial Waste Disposal which have been referenced many times herein.

The most substantial use of the sea for the disposal of radioactivity has been made by the British at Windscale in Cumberland, where they have been discharging activity into the Irish Sea through a pipeline which extends about two miles beyond the high water mark. An intensive program to monitor the effects of discharging wastes thusly has been carried out, and the results obtained through 1956 have been published.(121,122) The average rate of activity discharge is reported to be 2500 curies per month over five years. This rate has been increasing. The values for the year 1956 are given in Table 50.

Table 50

DISCHARGE OF LIQUID RADIOACTIVE WASTE TO THE SEA FROM WINDSCALE WORKS[a]
(For the year 1956)

Isotope	Mean Discharge Rate (Curies per Month)
Total beta activity	6100
Sr^{89}	106
Sr^{90}	150
Ru^{103}	120
Ru^{106}	2700
Pu^{239}	3.8

[a]Dunster, H. J., The Disposal of Radioactive Waste to the Sea During 1956, IGS-R/R-2 (January 1958).

The following general pattern of dispersal of activity in the sea off Cumberland has been shown. The discharged effluent is rapidly diluted with large volumes of sea water and is carried to and fro along the coast over a distance of a few kilometers by the oscillating tidal stream. Beyond the range of this movement, the activity is removed by the process of eddy diffusion, and the concentration falls to negligible levels in a few tens of kilometers. Much of the radioactivity becomes absorbed onto fine particles of sand, mud, and organic matter in suspension, and some of this is deposited onto the sea bed. The length of the pipeline is such that the concentration of deposited activity on the seashore is about 50 times lower than that on the sea bed near the effluent outlet. The fish in this area, of which plaice are the most important, live on the sea bed and consequently take up activity from the water, from the particles, and from the invertebrates which form their food. The activity level in the edible parts of fish is many hundred times less than that in the sea bed sand and is probably not greatly different from that of the surrounding water. Some of the activity reaching the shore is absorbed not by the sand but by seaweed, and, since this weed is used almost directly as a food, stringent limits have had to be placed on its activity. Most of the weed harvested in Cumberland grows within about 20 kilometers of the effluent outlet, and there is no substantial dilution by weed from other areas. Consequently, maximum permissible discharges are limited primarily by the activity of this weed and by the need to safeguard the small group of people (the Welsh) who regularly eat it in quantity.

The results of this experimental program have shown that it would be safe to release some 15,000 curies per month of fission products at a point about 3 kilometers off this part of the Cumberland coast. Further work now in hand suggests that this figure could reasonably be increased to over 45,000 curies per month. These high figures are possible because the marine processes in this area cause sufficient dispersal to prevent any continuing buildup from year to year of the activity levels, either in the vicinity of the outlet or in the more widespread regions of the Irish Sea.

In November, 1959, a conference on the Disposal of Radioactive Wastes was held in Monaco under the joint sponsorship of the International Atomic Energy Agency and of UNESCO. The subject which caused the most controversy at this meeting was the dumping of wastes into the sea. The Russians, particularly, objected to the practice. Other countries, especially with close interest in the North Sea, also expressed opposition. It was clear that everyone is agreed high-level wastes should not go into the sea. There was some feeling that the British operation at Windscale and those planned for Dounreay and Winfrith Heath are pushing the limits of low-level wastes pretty hard. The British presented their position fully and defended it ably.

The other use of the sea for waste disposal has been the dumping of very low-level radioactive solid trash. Some of this has been done off both

coasts of the United States, and attempts are being made to use the Gulf of Mexico also. All of these present and proposed operations have been under some considerable political pressure.

At the July 29, 1959, Hearings on the Disposal of Low-level Radioactive Wastes at Sea, held by the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy, AEC General Manager A. R. Luedecke said that the AEC has thus far disposed of at sea the following quantities of radioactivity (at the time of disposal):(123)

1. In the Atlantic Ocean, about 800 curies plus that in the reactor structure of the dismantled Seawolf prototype, which is estimated at 33,000 curies induced in stainless steel. The contained activity will be released to the sea through corrosion at an estimated rate of 2 to 3 curies a year. The major disposal areas are 230 miles and 150 miles southeast of Sandy Hook, both off the continental shelf at a depth greater than 1000 fathoms.

2. In the Pacific Ocean, about 14,000 curies at a site 48 miles west of the Golden Gate and about 60 curies at a site 53 miles west of Port Vicente, Calif. Both sites are also at depths greater than 1000 fathoms.

Luedecke said that licensed private organizations and other government agencies have also disposed of a total of about 2600 curies in the Atlantic Ocean, 102 curies in the Pacific Ocean, and 10 curies in the Gulf of Mexico.

PROBLEM FOR CHAPTER 7

1. Make such assumptions as seem reasonable and calculate possible concentrations of activity in sea water from dumping operations in the Atlantic and Pacific Oceans.

CHAPTER 8

ULTIMATE DISPOSAL

The term "ultimate disposal" must mean that a method is devised whereby concentrated high-level radioactivity may be stored in such a manner that the integrity of the environment is guaranteed for all time. This means, practically, that the storage method must be adequate for the better part of a millenium. The method (or methods) developed must have a cost which can be borne by the nuclear power economy. Such a solution to the problem has not yet been found or at least has not been proved. Proving the point may take a generation or so. Methods which have been used or suggested for containment of high-level wastes include tank storage of liquids, storage of liquids or solids in salt mines, and disposal of liquids to very deep wells.

Tank Storage

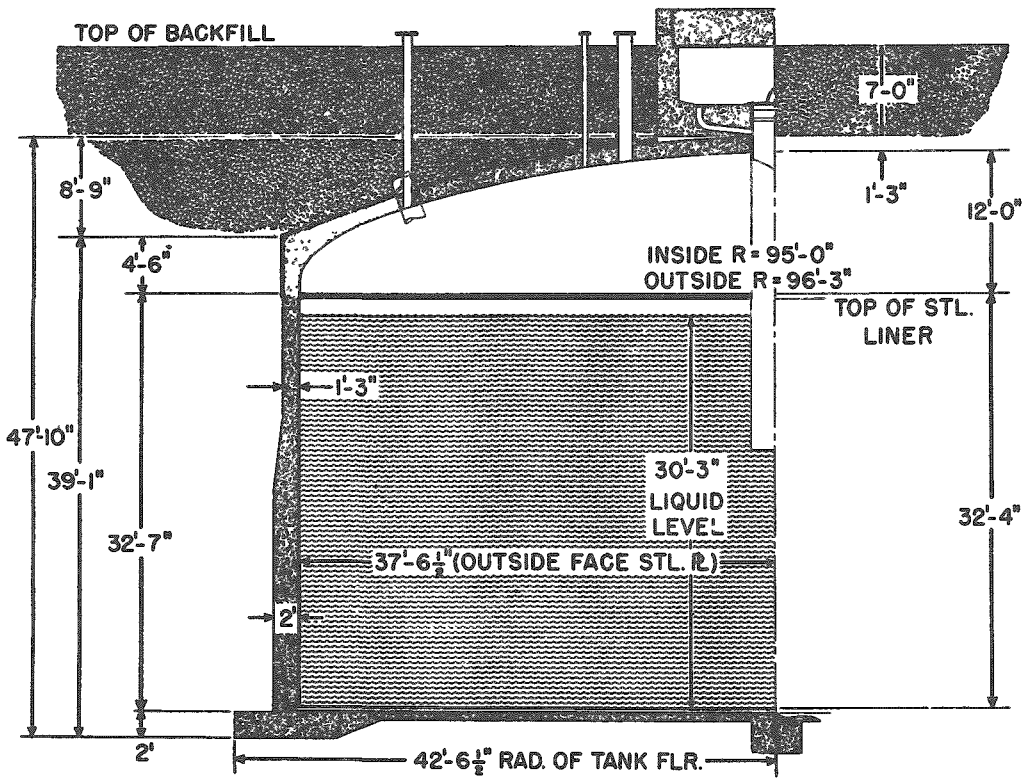
Tank storage must be considered under this heading if for no other reason than the fact that to date all of our "ultimate storage" has been done by this method. Few, if any, now suggest that liquid storage in tanks is a satisfactory "ultimate" answer. But, it has been suggested that tanks would be satisfactory for storing wastes so concentrated that they have set to a solid on cooling.

Some information on the cost, operation, and availability of tanks has already been given (Chapter 2). Storage tanks are designed to contain liquids safely for an indefinite period of time. Provision must be made for removal and transfer of the liquid in case of leakage. Shielding is accomplished by constructing the tanks underground. The tanks must withstand, therefore, an external earth pressure. The tank design must also take into account the elevated temperatures of the liquid and sludge, the temperature differential between them, and internal pressures caused by the liquid itself and the vapor pressure of the boiling liquid.

The design selected at Hanford is a cylindrical concrete tank with a flat bottom and an ellipsoidal dome roof.[124] The sides and bottom are covered with an inner steel liner. A cross section of such a tank is shown in Figure 21. The cylindrical shape adapts itself well to changes in temperature. The inner steel liner is more leakproof than would be a tank of concrete; yet the concrete forms a second line of defense if a leak occurs in the steel tank. In case both leak, test wells are provided to determine this fact. The reinforced concrete outer tank resists not only the external earth pressure but the internal liquid and vapor pressures. An ellipsoidal dome was chosen for the roof because it requires no internal supports. The lack of ground water at Hanford means that columns are not required to resist the uplift of water pressure under the tank.

Figure 21

Cross Section of Hanford Waste Storage Tank[a]



[a] O. H. Pilkey, et al., The Storage of High-level Radioactive Wastes Design and Operating Experience in the United States. Record of Hearings on Industrial Radioactive Waste Disposal, JCAE, Vol 1, page 418, August, 1959.

An ellipsoid has a much lower rise than a hemisphere, so that the amount of excavation is reduced. It is also a good structural shape to withstand the pressures both internal and external.

The inner steel tank was designed largely to specifications of the American Water Works Assoc. Better welding was required. Complete penetration butt-welded joints were used throughout and spot radiographing was done. The steel thickness on the more recently constructed tanks is $\frac{3}{8}$-inch.

The general specifications for the reinforced concrete tank were taken from the Portland Cement Assoc.(125) They were designed for full internal pressure, ignoring the inner steel liner. The concrete was

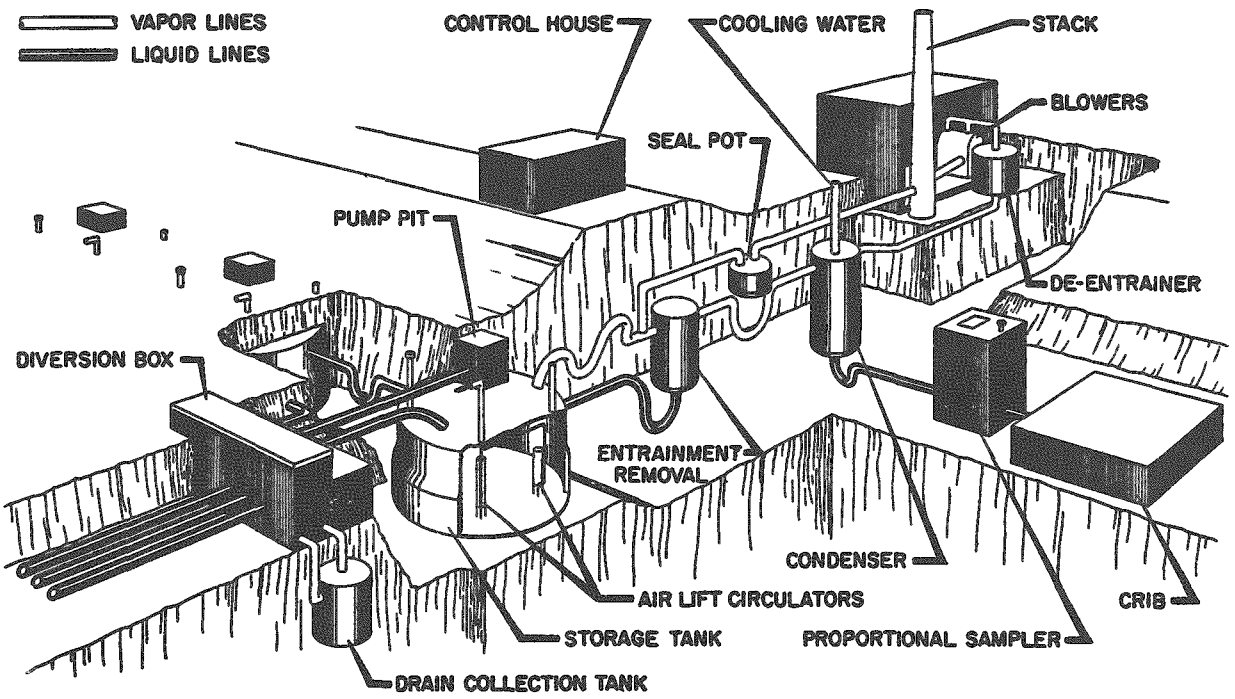
designed to withstand the external pressure of the earth, but this pressure was not used to offset the internal liquid pressure. A maximum wall thickness of 2 feet was selected.

The dome roof was also designed to Portland Cement Assoc. standards.(126) It was designed to withstand an external load of six to eight feet of earth plus its own weight plus a live load of two 35,000-lb tractors. A number of nozzles enter the tank through the dome to allow for temperature, pressure, liquid level and radiation measurement, and for sampling and liquid transfer.

A tank farm contains a good deal of equipment besides the waste storage tank proper. A schematic of a waste facility layout is shown in Figure 22. A seal pot provides protection against pressure surges. Vapors from the tanks are put through a deentrainment device and then condensed. Gases are then filtered before disposal to a stack. Condensed liquids may be returned to the tank or sent to ground disposal.

Figure 22

Waste Storage Facility Layout at Hanford[a]

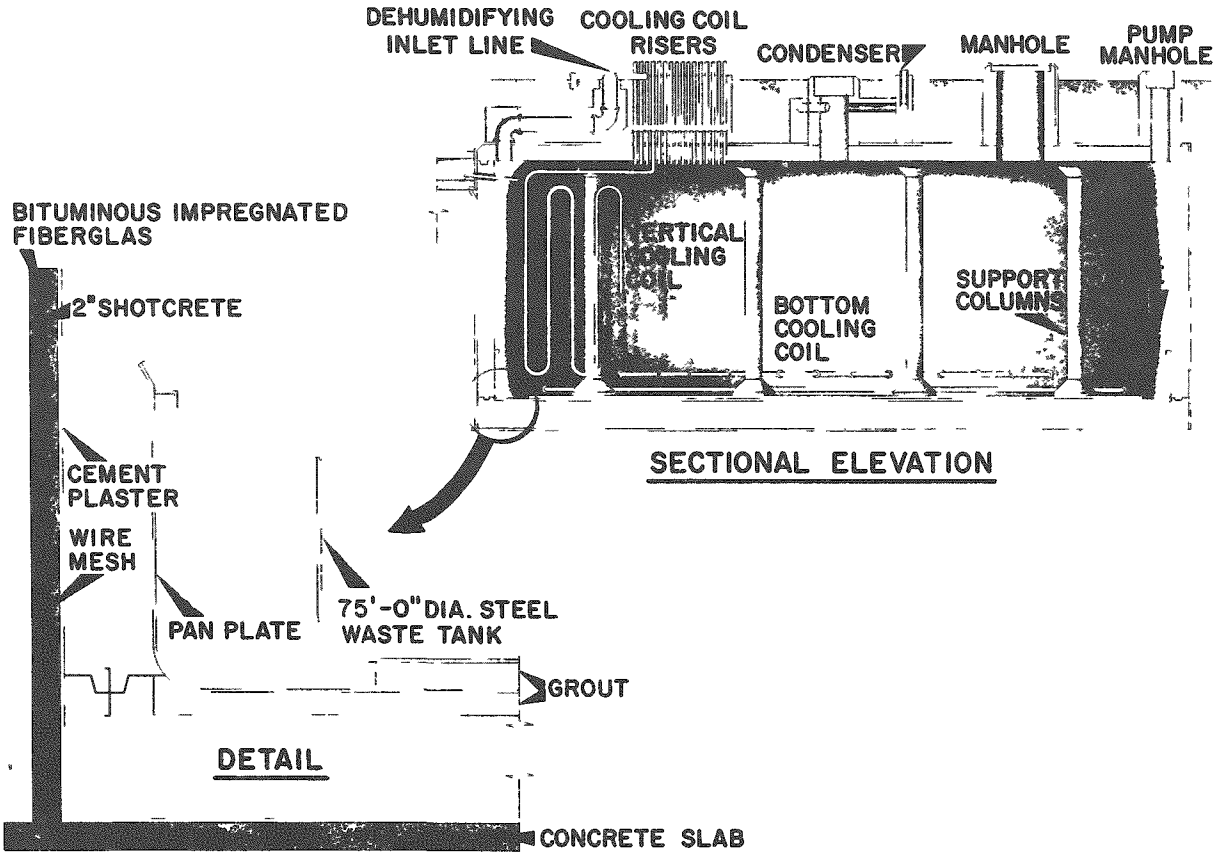


[a] O. H. Pilkey, et al., The Storage of High-level Radioactive Wastes Design and Operating Experience in the United States. Record of Hearings on Industrial Radioactive Waste Disposal, JCAE, Vol 1, page 413, August, 1959.

The tanks built at Savannah River are different in a number of respects (see Figure 23), brought about largely by the fact that the water table lies close to the surface of the ground and by the greater population density of the surrounding area. A "cup and saucer" design is utilized to insure collection of any leakage. Monitoring equipment is installed in the free space which exists between the outer steel-lined concrete shell and the steel tank. The inner tank is constructed of $\frac{1}{2}$-inch steel. It has a steel cover which, together with the earth covering, is supported internally by twelve steel-sheathed concrete columns.

Figure 23

Cross Section of Savannah River Waste Storage Tank[a]



[a] O. H. Pilkey, et al., The Storage of High-level Radioactive Wastes Design and Operating Experience in the United States. Record of Hearings on Industrial Radioactive Waste Disposal, JCAE, Vol 1, page 423, August, 1959.

Heat is removed by a series of cooling coils and the wastes are not allowed to boil. To remove heat from the first liquids put into a tank, there is a horizontal coil installed in the bottom of the tank consisting of

circular steel coils containing 15,000 sq ft of area. More efficient vertical coils containing an additional 9000 sq ft are installed higher in the tank, and there is a spare set near the top of the tank. It is easy to see why Savannah River tanks are more expensive than those installed at Hanford.

Operating experience with waste storage has been good. Some of them have now been in use for over 15 years and none has ruptured or even leaked seriously. Such operating problems as have occurred are associated with the boiling phenomenon and with radiolytic decomposition. While the tanks normally boil rather gently, at times they are known to burst into a violent surging boil, releasing vapors at rates up to twenty times that normally expected. Pressure surges up to 2 lb/sq in. are noted at such times. Provision for such pressure surges has been included in the design criteria of the newer tanks. This surging is believed to be due to the fact that much of the heat-producing activity is carried to the bottom of the tank by the sludges which settle out. Because of the hydrostatic head of the stored liquid, much of the released heat can be stored near the bottom of the tank without boiling taking place. If something occurs to start an upward flow of the hot liquid, a bubble of gas rising for instance, the pressure on the liquid is reduced; boiling can then start, which will increase the upward flow bringing more superheated liquid to the surface. Once started, vigorous boiling will occur until the reservoir of heat is dissipated. In the course of these surges, no damage to the tanks has occurred. There is at least one recorded instance, however, where the bottom of a tank bowed upward a distance of three or four feet and then subsided back essentially to its original position after a day or so.

Several years ago, when wastes which had been cooled a little less than 90 days prior to processing were sent to the tank farms, copious quantities of brown fumes were noted to escape from the tanks. This was thought to be associated with radiolytic decomposition. Since increasing the cooling time back to the more common values, this has not been a problem

Although there have been no recorded tank ruptures, there have been several attempts made to try to decide just how serious a tank rupture would be. If a rupture is assumed, the geological environment becomes most important. At Hanford, which has the most favorable of environments, quite uncatastrophic results are expected.(127) Two field-scale tests were performed to simulate loss of Redox wastes through a leak in an underground storage tank. One test used simulated neutralized Redox waste which had not undergone self-concentration. It contained a precipitate equal to about 5 volume per cent. The waste entered the ground through a one-inch hole in a 7 x 7-ft test plate without giving any indication of developing a flow restriction, either by a waste-soil reaction or by the precipitate plugging the soil. The maximum flow recorded was 450 gal/hr. Visual examination showed that the precipitate had dispersed in the soil, forming a saucer-shaped

pattern about 2 ft deep in the center with a radius of 7 ft. The precipitate-free wastes penetrated to a depth of 5 ft in the center and in the end had spread to about an 11-ft radius.

The second test employed a synthetic solution simulating Redox waste self-concentrated to 85 per cent of the original volume. This waste contained about 20 volume per cent of a rapidly settling precipitate. Only 50 gal of this waste entered the ground before plugging occurred and all measurable flow stopped. The plugging was apparently due to the settling of the precipitate in the bottom of the standpipe. Visual examination of the soil beneath the plate indicated that clear wastes had spread to a radius of about 3 ft.

On the other hand, a mathematical study was made at Oak Ridge of a hypothetical plant located in the Ohio River Valley. A series of less favorable postulates for the manner in which the waste would behave once it was in the ground was used, and this study did predict catastrophic results.(128) Depending on the assumptions, river concentrations 100,000 times MPC persisting for 30 to 60 days, river concentrations 100 times MPC for ten years, or river concentrations about twice MPC for a century were calculated. The assumptions were undoubtedly too pessimistic, but the fact that such calculations can be made using, at least, plausible assumptions makes it doubtful that tank storage can be accepted as a method of ultimate disposal for more than a very few highly favorable situations.

But it also seems clear that tank storage will have a place in overall waste management to store wastes during their first years, since it is easier to remove the radioactive decay heat from a liquid waste than from a solid.

Deep Well Disposal

The petroleum industry has developed an extensive technology of injecting fluids into deep wells. The American Petroleum Institute was requested to appoint a small committee to consider the feasibility of disposing of radioactive liquids in this way. A report of this subcommittee gave the following conclusions:(129)

1. "Radioactive waste can be disposed of safely by injection, through deep wells, into porous rocks. There is, obviously, a limit to the amount and activity of waste that can be stored in any particular formation without danger. This limit cannot be accurately determined on the basis of available information, but the data required for estimating the safe storage capacity of a reservoir can be obtained experimentally.

2. "Major problems in the disposal of radioactive waste through deep wells will be those of confining the waste, dissipating the heat generated, and protecting the disposal system against damage by corrosion or radiation. These will present difficulties, but no insurmountable obstacles are indicated.

3. "A disposal plant might comprise, in addition to the injection wells, a number of auxiliary wells which would serve various functions, with facilities for chemically treating, filtering, and storing waste and diluent water. Although reliable cost estimates cannot be made on the basis of information now available, preliminary consideration suggests that the cost of disposal might run less than a dollar per gallon of high-level undiluted waste.

4. "A modest program of laboratory work and theoretical study would clarify many of the problems. If such a study confirms the preliminary conclusion that deep-well disposal might be economic under favorable circumstances, a concurrent program of field testing and exploration should be the next phase of the development of the process to be undertaken."

Storage in Salt Deposits

A number of natural geological formations have been suggested as possible media for the storage of wastes. The one which seems to offer the most promise is rock salt. Salt has considerable compressive strength. At normal temperatures its movement is controlled by plastic flow and deformation occurs only slowly when salt is under pressure below the yield point. These unique characteristics give salt remarkable geological features. Excavations in salt are practically always dry. Because of its plasticity any fractures in salt close rapidly. Large spaces may be mined out; even at depths of 1,000 feet, two-thirds of the salt area may be removed without perceptible deformation of the pillars.(130)

The location of salt deposits is widespread in the United States. Deposits of rock salt underlie 400,000 square miles of the United States and they represent some of the few naturally occurring dry environments in the eastern part of the country. The volume of high-level waste that will probably be produced in the year 2000 is computed to be about 168 acre-feet or less than 10 per cent of the salt space now being mined out annually.

Although laboratory experiments have been conducted on the behavior of salt in relation to nuclear wastes, all of the principal factors cannot be conclusively determined in laboratories but must be established by means of field experiments. A research program coordinated through ORNL is underway to determine the feasibility of using salt formations as an ultimate

storage place for either liquid or solid wastes.[131 132] This research program includes heat calculations, chemical compatibility physical stability of the system, design of the field experiment, test of laboratory models of the field experiment, and the field experiment itself which will be carried on in a mine at Hutchinson, Kansas

Calculations on the storage of waste solutions as liquids in spherical cavities in salt show that, for a 10-ft diameter sphere and one-year-old wastes, the maximum rise in temperature at the center of the sphere is 530 F at 1000 hr and 315 F at the surface of the sphere at 1500 hr. The maximum temperature rise for a 6-year-old waste at the center of the sphere is 100 F at 4500 hr and 65 F at the surface at 7000 hr. It is concluded that the thermal problem in salt disposal will cause no serious difficulties if the size of the cavities and the age and dilution of wastes are controlled.

Laboratory tests so far have shown that the structural properties of rock salt are not greatly altered by high radiation, although high temperatures increase the creep rate for both irradiated and unirradiated samples. Chemical interaction of liquid wastes with salt produced chlorine and other chlorine compound gases, but the volumes were not excessive. The migration of nuclides through the salt and the deformation of the cavity can only be studied in the field.

The suggestion for the use of salt and this experimental program both contemplate the use of salt spaces for holding either liquids or solids.

CHAPTER 9

FUTURE PROBLEMS

A number of attempts have been made to assess the magnitude of the disposal problem which the world may face during the next half century. Estimates have been made by Hatch,[133] Glueckauf,[134] Lane,[135] this author,[136] and others. These estimates all tend to agree that the maximum plausible worldwide use of nuclear energy will result in the burnup of approximately 1,000 tons of fissionable material (and the production of 1,000 tons of fission product) per year near the end of the century. This is equivalent to approximately 2×10^6 megawatts of installed reactor capacity. Assuming this production rate, it is possible to calculate the equilibrium quantity of any particular isotope. Only a few are of serious importance from the standpoint of ultimate disposal. In Table 51 there are given the quantities of eight isotopes, which would have accumulated in fifty years. Also given are the maximum permissible concentration in water and air, and from these there are calculated the volumes of water and air required to dilute the isotope in question to tolerance.

Table 51

50-YEAR ACCUMULATION OF LONG-LIVED ISOTOPES AND REQUIRED DISPERSAL VOLUMES

Basis: 2.2×10^6 Mw Installed Reactor Capacity = 3 tons Fission Products/day.

Isotope	Accumulated Quantity in 50 Years, Curies	Maximum Permissible Concentrations[c]		Volume Required to Dilute to Tolerance	
		Water μc/ml	Air μc/ml	Water cu mi	Air cu mi
Zr^{95}	1.3×10^{11}	4×10^{-3}	4×10^{-7}	7.8×10^{3}	7.8×10^{7}
Ce^{144}	1.1×10^{11}	4×10^{-2}	7×10^{-9}	6.6×10^{3}	3.8×10^{9}
Ru^{106}	1.0×10^{11}	0.1	3×10^{-8}	2.4×10^{2}	8×10^{8}
Pm^{147}	5.1×10^{10}	1	2×10^{-7}	12	6×10^{7}
Sr^{90}	8.6×10^{10}	8×10^{-7}	2×10^{-10}	2.6×10^{7}	1×10^{11}
Cs^{137}	8.1×10^{10}	1.5×10^{-3}	2×10^{-7}	1.3×10^{4}	9.7×10^{7}
Tc^{99}[a]	2.0×10^{7}	3×10^{-2}	3×10^{-6}	0.2	1.6×10^{3}
Pu^{239}[b]	2.8×10^{6}	1.5×10^{-6}	2×10^{-12}	4.5×10^{2}	3.4×10^{8}

[a] Decay neglected.
[b] Based on a loss of 0.1% in processing.
[c] From National Bureau of Standards Handbook 52 (1953).

Strontium-90 is the controlling isotope and the dispersal volume of water needed, 2.6×10^7 cubic miles, is about eight per cent of the entire world ocean volume. This comparison is given only to emphasize once again the inadequacy of dispersal as a disposal method for this quantity of waste

Presently used solvent extraction processes produce about 1000 gallons of high-level waste per ton of uranium processed The total volume of waste formed initially is a function of the average fuel burnup If this is assumed to be 10,000 Mwd/ton, the annual processing rate will be 73,000 tons (200 tons/day). This is equivalent to 73,000,000 gallons/year. Interim storage, even charged at $1/gallon, would amount only to 0.015 mill/kwh. Either immediately or after a decay storage of some years, most or all of the water in these wastes could be separated from them. Using only known techniques such as already have been described herein, the liquid wastes could be evaporated, calcined to solids and the overhead vapor further decontaminated by ion exchange. Such a process might be expected to give a decontamination factor of 10^{10} and to cost less than $0.10/gal. This operation would add less than 0.002 mill/kwh to the cost of power.

The initial waste volume can be reduced by concentration, by the use of higher degrees of uranium burnup, or by the selection of processes which produce a smaller waste volume initially. There will be, however, an irreducible volume which must be accepted. Taking as the most optimistic assumption that the inert salts can be held to a weight equal to 9 times the fission products themselves, 30 tons per day of concentrate will be formed at the 1000 ton/year rate. At a density of 100 lb/cu ft, the volume is 600 cu ft/day - 220,000 cu ft/year. This is not an impractical number. If it were to cost $1000/cu ft to store this safely the annual bill would be about $200,000,000. Even this is only 0.04 mill/kwh and if the storage cost were reduced to $100/cu ft (a good goal to aim at), this item would add only 0.004 mill/kwh to the cost of power. It cannot be truly said that waste disposal is going to pose an economic roadblock to the industrial utilization of nuclear energy.

On the other hand, there does remain the technical problem of devising a safe resting place for the high-level waste - one which can be provided for $100/cu ft or even $1000/cu ft. This problem has not yet been solved. Some of the work directed toward solving the problem has been described herein. Since it is difficult to state categorically ahead of time that any particular method selected will be adequate for several centuries of storage, it may become desirable to use a system with several lines of defense. Such a system might consist of:

(1) forming the waste into a solid which is as nearly unleachable as possible;

(2) surrounding this solid with a package which itself offers some promise of long-term integrity; and

(3) storing in a place such that the likelihood of contacting the waste with water is low.

The first of these is being investigated in detail and shows reasonable promise of success. The second has not received adequate attention. Various packaging techniques must be tried out and subjected to rigorous long-term corrosion testing. The third is the subject of considerable speculation and not enough work. The more arid parts of the world naturally suggest themselves, and several of them are being used as disposal sites. These areas tend to be remote from population centers (areas of high electrical power demand), and it would be desirable to discover ways of storing wastes compatible with the proximity of people.

There would appear to be no reason to be pessimistic about the chances of solving this problem.

PROBLEM FOR CHAPTER 9

1. Making reasonable assumptions for the ranges to be expected for the following quantities:

a) fuel burnup, and

b) waste volume/ton uranium processed,

construct a table or graph showing the $/gallon of waste which can be spent for waste disposal if 2 per cent of the total power cost is allocated to this item.

BIBLIOGRAPHY

1. Harold Etherington, editor, Reactor Engineering Handbook, page 11-28, McGraw-Hill Book Co., New York (1958).

2. Leslie Burris, Jr. and Ira G. Dillon, Estimation of Fission Product Spectra in Discharged Fuel from Fast Reactors, ANL-5742 (July 1957).

3. J. O. Blomecke and Mary Todd, U^{235} Fission Products as a Function of the Thermal Neutron Flux, Irradiation Time, and Decay Time, ORNL 2127, Vol I, 1957; Vol II, 1958.

4. C. W. Klassen, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 4, page 2544 (August 1959).

5. Hanson Blatz, editor, Radiation Hygiene Handbook, First Edition, Section II, McGraw-Hill Book Company, New York (1959).

6. National Bureau of Standards, Handbook 52, Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water, (March 1953).

7. National Bureau of Standards, Handbook 69, Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air and Water for Occupational Exposure (June 1959).

8. I. Pullman, Physical Properties of Ionizing Radiation which Affect the Population of the United States, Nat'l. Acad. of Sci., April 18, 1956 (Limited Circulation), National Academy of Sciences, Washington 25, D.C.

9. F. L. Culler, Nature of Radioactive Wastes, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 32, August, 1959.

10. Reactor Fuel Processing, 3 (1), 4 (January 1960).

11. D. A. Holaday, The Nature of Wastes Produced in the Mining and Milling of Uranium Ores, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 72, August, 1959.

12. Industrial Radioactive Wastes: Mining and Milling of Uranium Ores, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 674, August, 1959.

13. E. C. Tsivoglou et al., Waste Characteristics for the Resin-in-Pulp Uranium Extraction Process, Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva (1958), Vol. 18, p. 174.

14. Nature of Wastes from the Uranium Milling Industry, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 58, August, 1959.

15. Reactor Fuel Processing, 3 (1), 42 (January 1960).

16. M. A. DeSesa, Interim Report on Investigations into the Problem of Radioactive Pollution of Uranium Mill Effluents, WIN-101 (Dec. 15, 1958).

16a. L. B. Emlet, Statement on Waste Management at the Oak Ridge and Paducah Production Plants, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 515, August, 1959.

17. N. C. Jesson, Nature of Wastes in Fuel Element Manufacture, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 116, August, 1959.

18. Radioactive Waste Management Operations at the Hanford Works, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 199 et seq., August, 1959.

19. Summary-Analysis of Hearings, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, page 11, August, 1959.

20. A. L. Bethel et al., Presentation on Shippingport Atomic Power Station (PWR) Waste Disposal Facilities, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 620, August, 1959.

21. T. J. Iltis and M. E. Miles, Radioactive Waste Disposal from U.S. Naval Nuclear Powered Ships, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 924, August, 1959.

22. Radioactive Wastes from Reactor Operation, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 122, August, 1959.

23. R. J. Christl, Waste Management Savannah River Plant Atomic Energy Commission, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States Vol 1, page 826, August, 1959.

24. J. R. Horan, Radioactive Waste Disposal Management at the National Reactor Testing Station, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 584, August, 1959.

25. R. L. Doan, Statement of the Phillips Petroleum Company Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1, page 610, August 1959.

26. Management of Radioactive Wastes at Argonne National Laboratory. Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 1. page 708. August, 1959.

27. W. A. Rodger and P. Fineman, A Complete Waste Disposal System for a Radiochemical Laboratory, Nucleonics 9 6, 50-61 (December 1951)

28. E. A. Coppinger and R. E. Tomlinson, Heat Problems in the Disposal of High-Level Radioactive Wastes, Chem. Eng. Progress 52, 10, 417 (October 1956).

29. Hanson Blatz, editor, Radiation Hygiene Handbook, McGraw-Hill Book Co., New York (1959), Section 20.

30. Thomas, Hill, and Abersold, Industrial Engineering Chemistry, 41 2409 (1949).

31. A. G. Blasewitz and W. C. Schmidt, Treatment of Radioactive Waste Gases, Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva (1958), Vol. 18, p. 187.

32. Progress Report on Waste Studies on Waste Concentration Studies V: Engineering Results on the BNL Semi-Works Vapor Filtration Vapor Compression Evaporator, BNL-121 (T-24) (August 15, 1951)

33. I. Langmuir and K. Blodgett. Smokes and Filters. OSRD-3460 (January 1944).

34. R. Dennis et al., Dust Collectors Tested in the Field. Chemical Engineering (May 1954).

35. R. C. Smith, An Electrostatic Precipitator for the Collection of Uranium Tetrafluoride Dusts, K-1229 (April 30, 1956).

36. R. C. Liimatainen and W. J. Mecham, Removals of Halogens, Carbon Dioxide, and Aerosols from Air in a Spray Tower, Journal of the Air Pollution Control Association, 6 (1) 17 (May 1956).

37. D. C. Hampson, E. H. Hykan, and W. A. Rodger, Basic Operational Report of the Argonne Waste Incinerator, ANL-5067 (February 1953).

38. Tariff No. 10, Published Interstate Commerce Commission Regulations for Transportation of Explosives and other Dangerous Articles, Bureau of Explosives of the Association of American Railroads, H. A. Campbell, Agent, 30 Vesey Street, New York 7, N.Y.

39. Motor Carriers Explosives and Dangerous Articles Tariff No. 8, Tariff Bureau of the American Trucking Associations, Inc., F. G. Freund, Agent, 1424 - 16th Street N.W., Washington 6, D.C.

40. Transportation of Explosives and Other Dangerous Articles, Civil Air Regulations, Available from Supt. of Documents, US Government Printing Office, Washington 25, D.C.

41. Official Air Transport Restricted Articles Tariff, E. F. Johnson, Agent, National Airport, Washington, D.C.

42. Title 46, Part 146, Code of Federal Regulations, Federal Register, July 17, 1952, page 6460 ff and December 31, 1952.

43. Water Carrier Tariff No. 6, Bureau of Explosives, H. A. Campbell, Agent, 30 Vesey Street, New York 7, N.Y.

44. U.S. Postal Guide, 1951 Edition, page 51, Part I.

45. Problems Associated with Transportation of Radioactive Substances, NAS-NRC Publication No. 205 (1947).

46. Federal Register, November 8, 1947, page 7328.

47. F. R. Bruce, Statement for the Hearings, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol 3, page 2345, August, 1959.

48. E. R. Irish and R. E. Tomlinson, Retention of Fission Products in Tanks, Paper presented at Fuel Processing Symposium, Brussels, Belgium, May, 1957.

49. F. Birch, Thermal Considerations in Deep Disposal of Radioactive Waste, NAS-NRC/Pub-588 (July 1958).

50. P. Fineman and D. M. Mohler, A Survey of Waste Evaporation of AEC Sites, ANL-SL-615 (1952).

51. KAPL-594 (August 28, 1951).

52. Chem. Eng., 62, 194 (1955).

53. Progress Report on Waste Concentration Studies BNL-49 (T-16) (November 15, 1949) (Restricted).

54. Brookhaven Conference Report, Waste Processing and Evaporation, BNL-59 (C-12) (March 27-28, 1950).

55. Progress Report on Waste Concentration Studies, Brookhaven National Laboratory, BNL-1-10 (March 1, 1949); BNL-90 (T-20)(October 1, 1950); BNL-121 (T-24) (August 15, 1951); Waste Processing, 2. Evaporation, Brookhaven Conference Report, BNL-59-(C-12) (March 27-28, 1950).

56. Reactor Fuel Processing, 1 (3) 30 (July 1958).

57. Reactor Fuel Processing, 2 (3) 31 (July 1959).

58. H. W. Alter, et al., Electrolytic Recycle Method for the Treatment of Radioactive Nitric Acid Waste, KAPL-1721 (June 26,1957).

59. Reactor Fuel Processing, 2 (1) 40 (January 1959).

60. K. E. Cowser and R. J. Morton, Treatment Plant for Removal of Radioactive Contaminants from Process Waste Water. Part II: Evaluation of Performance, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, Joint Committee on Atomic Energy, 86th Congress of the United States, page 547, August, 1959.

61. C. P. Straub, W. J. Lacy, and R. J. Morton, Methods for the Decontamination of Radioactive Liquid Wastes, Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva (1955), Vol. 9, p. 24.

62. Summary Reports; Chemical Engineering Division, Argonne National Laboratory ANL-4463, page 80, 1950; ANL-4499, page 85, 1950; ANL-4543, page 76, 1950; ANL-4720, page 70, 1951.

63. C. W. Christenson, M. B. Ettinger, G. G. Robeck, E. R. Herman, K. C. Kohr, and J. F. Newell, Removal of Plutonium from Laboratory Wastes, Ind. Eng. Chem. 43, 1509-1516 (1951).

64. C. S. Lowe, et al., Decontamination of Liquid Wastes by Iron Sulfide, Iron Hydroxide, and Calcium Phosphate Precipitations; Final Report, AECD-3632 or MLM-662 (Rev) (December 28, 1953).

65. R. F. McCauley, R. A. Lauderdale, and R. Eliassen, A Study of the Lime-Soda Softening Process as a Method for Decontaminating Radioactive Waters, NYO-4439 (September 1, 1953).

66. J. B. Nesbitt, W. J. Kaufman, R. F. McCauley, and R. Eliassen, The Removal of Radioactive Strontium from Water by Phosphate Coagulation, NYO-4435 (February 15, 1952).

67. W. L. Wilson, P. A. F. White, and J. G. Milton, The Control, Conveyance, Treatment and Disposal of Radioactive Effluents from the Atomic Weapons Research Establishment, Aldermaston, J. Brit, Nuclear Energy Conference, 1, 149 (July 1956).

68. F. R. Farmer, The Problem of Liquid and Gaseous Effluent Disposal at Windscale, J. Brit, Nuclear Energy Conference, 2, 26 (January, 1957).

69. E. Glueckauf and T. V. Healy, Chemical Processing of Fission Product Solutions, Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva (1955), Vol. 9, p. 635.

70. W. W. Schulz and T. R. McKenzie, The Removal of Cesium and Strontium from Radioactive Waste Solutions, Sanitary Engineering Conference, Cincinnati, Ohio, TID-7517, December, 1955.

71. R. E. Burns and M. J. Stedwell, Volume Reduction of Radioactive Waste by Carrier Precipitation, Chem. Eng. Prog., 53, 93-F (1957).

72. H. Etherington, ed, Nuclear Engineering Handbook, McGraw-Hill Book Co., New York (1958), page 11-96.

73. R. Kunin and R. J. Meyers, Ion Exchange Resins, John Wiley and Sons, Inc., New York (1950).

74. R. Kunin and R. J. Meyers, Ion Exchange Resins, John Wiley and Sons, Inc., New York (1950), page 61.

75. H. G. Swope in Ion Exchange Technology (F. C. Nachod and J. Schubert, ed), Academic Press, Inc., New York (1955), p. 477.

76. H. G. Swope and E. Anderson, Cation Exchange Removal of Radioactivity from Wastes, Ind. Eng. Chem., 47, 78 (1955).

77. G. W. Parker, I. R. Higgins, and J. T. Roberts in Ion Exchange Technology, (F. C. Nachod and J. Schubert, ed) Table 16.8 Academic Press, New York (1956), p. 144.

78. I. R. Higgins, Radiation Damage to Organic Ion Exchange Materials, ORNL-1325 (1953) (Confidential).

79. S. A. Fisher, Effect of Gamma Radiation on Ion Exchange Resins, RMO-2528 (1954).

80. R. E. Wedemeyer, The Stability of Ion Exchange Resins to X Rays PhD. Thesis, Vanderbilt University, 1953.

81. W. R. Walters in Ion Exchange Technology, (F. C. Nachod and J. Schubert, ed) Academic Press, Inc., New York (1956), p. 494.

82. W. R. Walters, D. W. Weiser, and L. J. Marek, Concentration of Radioactive Aqueous Wastes - Electromigration through Ion-Exchange Membranes, Ind. Eng. Chem., 47, 61 (1955).

83. L. P. Hatch, W. H. Regan, B. Manowitz and F. Hitman, Processes for High Level Radioactive Waste Disposal, Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva (1955), Vol. 9, p. 648.

84. W. E. Dobbins, Biological Methods for the Removal of Radioactivity from Liquids, Cleveland Nuclear Eng. Science Congress, 1955.

85. W. E. Dobbins, G. P. Edwards, and W. Gulevich, Removal of Radioactivity from Laundry Wastes by Trickling Filters, NYO-4641 (May 1955).

86. J. F. Newell, C. W. Christenson, and E. R. Mathews, Laboratory Studies on Removal of Plutonium from Laundry Wastes, Ind. Eng. Chem., 43, 1516-1519 (1951).

87. E. R. Herman, E. F. Gloyna, Summary of Investigations on the Removal of Radioisotopes from Waste Water by Oxidation Ponds, Sanitary Engr. Conference, Cincinnati, Ohio, TID-7517 (December 1955).

88. L. L. Ames, Jr., et al., Removal of Sr from Wastes by Calcite-Phosphate Mechanism, Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, (1958), Vol. 18, p. 76.

89. A. Raggenbass, et al., A Pilot Unit for the Separation of Cesium-137, Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva (1958), Vol. 18, p. 210.

90. R. L. Moore and R. E. Burns, Fission Product Recovery from Radioactive Effluents, Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva (1958), Vol. 18, p. 231.

91. H. A. C. McKay, Alpha-emitters in Fission Product Wastes, AERE C/R 2034 (FPSD/p 19), Harwell, Berkshire (1956).

92. A. F. Rupp, The Oak Ridge National Laboratory Fission Product Pilot Plant, Hearings on Industrial Radioactive Waste Disposal, Joint Committee on Atomic Energy, 86th Congress of the United States, Vol. 3, page 2335, August, 1959.

93. L. P. Hatch, W. H. Ragen, B. Manowitz and F. Hitman, Processes for High-level Active Waste Disposal, Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, Switzerland (1955), Vol. 9, p. 648.

94. L. P. Hatch, Fixation of Radioactive Wastes in Stable Solids, Hearings on Industrial Radioactive Waste Disposal, Joint Committee on Atomic Energy, 86th Congress of the United States, Vol. 3, page 1839, August, 1959.

95. Reactor Fuel Processing, 2 (1) p. 41 (January 1959).

96. R. L. Blanchard, B. Kahn, and G. G. Robeck, Laboratory Studies on the Ground Disposal of ORNL Intermediate-level Radioactive Wastes, ORNL-2475 (April 11, 1958).

97. Reactor Fuel Processing, 1 (2) p. 31 (April 1958).

98. T. N. McBay, R. L. Hamner, and M. P. Hayden, Fixation of Radioactive Wastes in Clay Flux Mixes, Sanitary Engineering Conference, Cincinnati, Ohio; TID-7517 (December 1955).

99. W. A. Patrick, Use of Artificial Clays for Removal and Fixation of Radioactive Nuclides, Sanitary Engineering Conference, Cincinnati, Ohio; TID-7517 (December 1955).

100. C. B. Amphlett and D. T. Warren, AERE C/R-1686 (1956).

101. R. W. Durham, Disposal of Fission Products in Glass, paper presented at the 2nd Nuclear Engineering and Science Congress, Philadelphia, Pa., March, 1957.

102. L. C. Watson, et al., The Disposal of Fission Products in Glass, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva (1958). Vol. 18, p. 19.

103. Chemical Engineering Division Summary Report, ANL-4720 (1951), p. 43.

104. KLX-1377, Disposal of Radioactive Wastes in Cement, (June 1952).

105. J. A. Kaufman and N. B. Garden, A Method for the Solidification of Radioactive Liquid Wastes, UCRL-8684 (March 1959).

106. A. A. Jonke, E. J. Petkus, and J. W. Loeding, A Fluidized Bed Technique for Treatment of Aqueous Nuclear Wastes by Calcination to Oxides, Sanitary Engineering Conference, Cincinnati, Ohio; TID-7517, (December 1955).

107. J. W. Loeding, et al., Fluidized Bed Conversion of Fuel Processing Wastes to Solids for Disposal, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva (1958), Vol. 18, p. 56.

108. Reactor Fuel Processing, 2 (1), p. 42 (January 1959).

109. Reactor Fuel Processing, 2 (4), (October 1959).

110. Reactor Fuel Processing, 2 (1) p. 40 (January 1959).

111. R. F. Domish, et al., Calcination of High-level Atomic Wastes as a Step in Ultimate Disposal, BNL-535 (December 1958).

112. M. I. Goldman, et al., Fixation in Vitreous Matrices of High-activity Fission Product Wastes from Aqueous Reprocessing of Spent Stainless Steel-Uranium Fuel Elements, NYO-4444 (February 1, 1960).

113. H. M. Parker, Testimony before Joint Committee on Atomic Energy, Hearings on Industrial Radioactive Waste Disposal, Vol. 3, pages 2356-2372, August, 1959.

114. G. R. Hilst, Testimony before Joint Committee on Atomic Energy, Hearings on Industrial Radioactive Waste Disposal, Vol. 2, pages 1579-1604, August, 1959.

115. H. A. Thomas, Jr., Testimony at the Hearings, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol. 2, page 1001, August, 1959.

116. R. F. Foster, Testimony at the Hearings, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol. 2, page 1010, August, 1959.

117. C. V. Theis, Testimony at the Hearings, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol. 2, page 1116, August, 1959.

118. R. E. Brown, et al., The Storage of High-level Radioactive Wastes to the Ground, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva (1958), Vol. 18, p. 95.

119. D. W. Pritchard, Environmental Factors Affecting the Disposal of Radioactive Waste Materials into Tidal Estuaries and Coastal Bodies of Water, Testimony at the Hearings, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol. 2, page 1271, August, 1959.

120. R. Revelle, Research Needed to Evaluate the Effects of Radioactive Material in the Marine Environment, Statement for the Record, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol. 2, page 1299, August, 1959.

121. H. J. Dunster and F. R. Farmer, A Summary of the Biological Investigations of the Discharges of Aqueous Radioactive Waste to the Sea from Windscale Works, Sellafield, Cumberland, IGS-R/R-3 (January 1958).

122. H. J. Dunster, The Disposal of Radioactive Waste to the Sea During 1956, IGS-R/R-2 (January 1958).

123. The Forum Memo to Members, Atomic Industrial Forum, Inc., 6 (9) (September 1959).

124. O. H. Pilkey, et al., The Storage of High-level Radioactive Wastes Design and Operating Experience in the United States, Record of Hearings on Industrial Radioactive Waste Disposal, JCAE, Vol. 1, pages 394-422, August, 1959.

125. Portland Cement Association, Bulletin No. ST-57, Circular Tanks without Pre-stressing.

126. Portland Cement Association, Bulletin No. ST-55, Design of Circular Domes.

127. Reactor Fuel Processing, 2 (1), p. 44 (January 1959).

128. Health Physics Annual Progress Report for the Period Ending July 31, 1959, ORNL-2806, page 106.

129. T. V. Moore, Problems in Disposal of Radioactive Wastes in Deep Wells, Report on Subcommittee on Radioactive Waste Disposal, American Petroleum Institute, October, 1958.

130. W. B. Heroy, Disposal of High-level Waste in Natural Salt Formations, Testimony at the Hearings, Hearings on Industrial Radioactive Waste Disposal, JCAE, 86th Congress of the United States, Vol. 3, page 2028, August, 1959.

131. F. L. Parker, et al., Status Report on Waste Disposal in Natural Salt Formations, ORNL-2560 (Sept. 2, 1958).

132. F. L. Parker, et al., Status Report on Waste Disposal in Natural Salt Formations, II, ORNL-2700 (April 23, 1959).

133. L. P. Hatch, Am. Scientist, 41, 410 (1953).

134. E. Glueckauf, Long-term Aspects of Fission Product Disposal, Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, Switzerland (1955), Vol 9, p. 3.

135. J. A. Lane, Determining Nuclear Fuel Requirements for Large-scale Industrial Power, Nucleonics 12(10), 65 (1954).

136. W. A. Rodger, Radioactive Wastes - Treatment, Use, Disposal, Chem. Eng. Prog., 50, 263 (1954).