

UNIT OPERATIONS USED TO TREAT PROCESS AND/OR
WASTE STREAMS AT NUCLEAR POWER PLANTS*

H. W. Godbee

A. H. Kibbey

Oak Ridge National Laboratory
P. O. Box X
Oak Ridge, Tennessee 37830

MASTER

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ABSTRACT

An attempt is made to identify the main sources of low-level radioactive wastes (LLW) that are generated at light water reactor (LWR) nuclear power plants in the United States. To place the LWR waste problem in perspective, rough estimates are given of the annual amounts of each generic type of LLW [i.e., Government and commercial (fuel cycle and non-fuel cycle)] that is generated. Most of the wet solid wastes arise from the cleanup of gaseous and liquid radioactive streams prior to discharge or recycle. Many different chemical engineering unit operations have been, are being, and likely will be used to treat process and/or waste streams at LWR plants. These include adsorption, evaporation, calcination, centrifugation, compaction, crystallization, drying, filtration, incineration, reverse osmosis, and solidification of waste residues. The treatment of these various streams and the secondary wet solid wastes thus generated (e.g., filter cartridges, filter sludges, and spent ion-exchange resins) is described. The dry wastes from all LWRs have similar physical and chemical characteristics in that they can be classified as compactible, noncompactible, combustible, noncombustible, or combinations thereof. The various treatment options for concentrates or solid wet wastes, and for dry wastes are discussed. Among the dry waste treatment methods are compaction, baling, and incineration, as well as chopping, cutting and shredding. Organic materials [liquids (e.g., oils or solvents) and/or solids], could be incinerated in most cases. The filter sludges, spent resins, and concentrated liquids (e.g., evaporator concentrates) are usually solidified in cement, or urea-formaldehyde or unsaturated polyester resins prior to burial. Incinerator ashes can also be incorporated in these binding agents. Asphalt has not yet been used as a solidification agent in the United States, but it probably will be used in the near future. This paper presents a brief survey of operational experience at LWRs with various unit operations, including a short discussion of problems and some observations on recent trends. Such information is a valuable aid in retrofitting old equipment and in selecting new. Some areas appearing to need more research, development, and demonstration are specifically pointed out.

UNIT OPERATIONS USED TO TREAT PROCESS AND/OR WASTE STREAMS AT NUCLEAR POWER PLANTS

H. W. Godbee and A. H. Kibbey

The requisite cleanup or decontamination of radioactive process and/or waste streams at light water reactor (LWR) nuclear power plants is obtained by the combination of a number of chemical and physical separations methods or unit operations. These clean-up operations generate radioactive wastes that must be prepared for disposal. Additionally, all plants generate radioactive trash and some discarded or failed equipment which also must be processed for disposal. To place the LWR waste problem in perspective, a rough summary of the low-level radioactive waste (LLW) volumes generated in all U.S. nuclear activities (i.e., Government and commercial) and sent to shallow land burial is presented in Table 1. As shown, LWR operations account for almost half of the commercial waste ($\sim 2.8 \times 10^4 \text{ m}^3/\text{y}$ out of over $5.6 \times 10^4 \text{ m}^3/\text{y}$) and are about equal to the Government LLW generation rate. Better utilization of unit operations to reduce the amounts of LWR waste that must be shipped for offsite burial has recently become a prime concern. Impetus in this direction has come from the closing of the commercial burial grounds at West Valley (New York), Maxey Flats (Kentucky), and Sheffield (Illinois), and the new limitations that have been placed on waste acceptance at Barnwell (South Carolina). Since most of the U.S. nuclear power plants are located in the East and Midwest, a significant saving in the costs of transport to and burial at the

Table 1. Estimated accumulated LLW^a volumes buried and present amounts shipped to burial annually

Source	Total accumulation ^b		Present annual burial rate	
	(m ³)	(ft ³)	(m ³)	(ft ³)
<u>Government</u>	1.4 x 10 ⁶	50.8 x 10 ⁶	~2.8 x 10 ⁴	~1 x 10 ⁶
<u>Commercial^c</u>	4.5 x 10 ⁵	15.8 x 10 ⁶	>5.6 x 10 ⁴	>2 x 10 ⁶
<u>Fuel Cycle:</u>			3.4 x 10 ⁴	1.2 x 10 ⁶
Reactor Operation			~2.8 x 10 ⁴	~1 x 10 ⁶
Fuel Fabrication			~5.7 x 10 ³	~2 x 10 ⁵
Other Steps			small	small
<u>Nonfuel Cycle:</u>			~2.2 x 10 ⁴	~7.8 x 10 ⁵
<u>Institutional</u>			~1.9 x 10 ⁴	~6.7 x 10 ⁵
Medical			~1.8 x 10 ⁴	~6.4 x 10 ⁵
Academic			~7.5 x 10 ²	~2.6 x 10 ⁴
<u>Industrial and Other Research (including pharmaceutical)</u>			~3 x 10 ³	>1.1 x 10 ⁵
Total	1.9 x 10 ⁶	66.6 x 10 ⁶	~8.4 x 10 ⁴	~3 x 10 ⁶

^aLow-level waste (LLW) comprises that radioactive waste which is not spent fuel or high-level waste [as defined in "Siting of Fuel Reprocessing Plants and Related Waste Management Facilities," Federal Register 35, No. 222, 17530 (November 14, 1970)] and which contains less than 10 nCi of transuranics per gram of material.

^bAccumulation times: Government \geq 4 decades.
Commercial \geq 2 decades.

^cIncludes all categories listed below; mill tailings are excluded.

Beatty (Nevada) or Richland (Washington) sites could be realized through volume reduction techniques.

Some early stream cleanup and waste treatment practices at LWRs have been abandoned or modified as new and improved technologies have been developed. This paper endeavors to point out some historic pitfalls and to describe and assess general present practices at LWRs. A passing comparison is made with common practice in other segments of the nuclear industry (i.e., defense, institutional, and industrial activities) to show where extensions of known technology could be applied. An attempt is made to indicate possible future trends in the unit operations used at LWR plants by taking into account pertinent nuclear power plant experience in Canada, Europe, and other foreign countries.

The first generation of small nuclear power plants [<300 MW(e)] sometimes used relatively unsophisticated off-the-shelf equipment that had been widely applied to clean up operations in other industries that do not suffer from the constraints of remote operation and maintenance that are imposed by radioactivity. Additionally, due to the lack of experience in a new industry, streams were not accurately characterized, especially under upset conditions. As a result, equipment was frequently underdesigned and the amounts of waste generated often greatly exceeded expectations. Lack of proper instrumentation, as well as a deficiency of sampling ports and devices, made process control difficult. Some of these difficulties continued to persist in the next generation of larger [~ 500 to 1000

MW(e)] LWR plants. Consequently, there have been many instances of evaporators being undersized and/or failing because of corrosion; filters being easily plugged and/or failing to remove fine particulates; ion-exchange beds and resin transfer lines plugging and being insufficiently shielded; solidification systems that either failed to solidify completely or else set prematurely, and often were incapable of being operated remotely; and centrifuges failing because of the action of abrasive particles. The separation of components in stream cleanup and waste treatment systems, as opposed to skid-mounting, is another concept that evolved out of early experience that forced adoption of stronger measures to minimize personnel exposures.

In LWR nuclear power plants, the liquid streams have various amounts of dissolved plus suspended solids and varying amounts of radioactivity associated with them, depending upon their source within the plant. Corrosion products in the coolant stream become activated in the internals of the reactor core, producing such radioactive species as ^{58}Co , ^{60}Co , ^{54}Mn , ^{51}Cr , ^{58}Ni , and ^{59}Fe . Defective fuel and uranium present on the cladding of fuel elements (tramp uranium) also contribute radioactive fission products such as ^{90}Sr , ^{134}Cs , ^{137}Cs , ^{131}I , and ^{85}Kr . Generally speaking, relatively significant fractions (i.e., about one-fourth) of the activated corrosion products (especially iron and nickel) tend to be present as suspended solids, and fission products tend to be present dominantly as soluble forms. Adequate facilities and equipment to

collect and process radioactive liquid streams enable the nuclear industry to hold releases of radioactive material in liquid effluents within applicable regulatory limits.¹ These limits are most readily met by minimizing the volume of liquids discharged and/or by decontaminating the liquids to a high degree before discharging them. In LWR plants, as in the other steps of the nuclear fuel cycle, over half the LLW is generated from this routine cleanup of liquid process or waste streams that are recycled within or discharged from the plant. The solid wastes thus generated are comprised of filter sludges (sometimes including filter aids) as well as spent ion-exchange resins, reverse osmosis (RO) membranes, and filter cartridges. At these plants the largest volume of LLW is usually generated during refueling and/or maintenance operations. Normally, at the so-called deep-bed (referring to beds of bead resins used in primary coolant cleanup) boiling water reactor (BWR) plants, the largest LLW volume fraction is solidified ion-exchange resin regenerant solution (Na_2SO_4). Filter/demineralizer (e.g., Powdex) type BWRs do not regenerate their resins, and dewatered filter/demineralizer sludges account for a large volume fraction of their LLW. At pressurized water reactor (PWR) plants, solidified boric acid concentrates from the coolant and chemical-and-volume-control system are the major LLW.² Disposable cartridge filters, rather than precoat filters, are widely used in PWR plants.³ The spent filter cartridges, while small in volume, contain a large portion of the activity removed from the process and wastewater streams

Some PWRs use condensate polishers (resin beds) in the secondary loop to clean the water for recycle, and these PWR regenerant solutions are similar to those from a BWR. In the event of primary to secondary system leakage, radioactive fission products collect on the condensate polisher resins but normally they are not radioactive. In general, PWRs generate less LLW (volume/MW) than BWRs.⁴ All nuclear power plants generate some waste oils.

In this paper, the LLW from LWR plants is treated in two broad categories, namely dry and wet wastes. Dry radioactive wastes generally contain trash comprised of such items as paper, rags, plastics, rubber, wood, glass, and metals that can be classified as combustible or noncombustible and compactible or noncompactible, or as combinations thereof. As mentioned earlier, wet wastes derive in a large part from cleanup of aqueous process or waste streams prior to recycle or discharge. Some of the more conventional physical and chemical methods used to treat radioactive liquid process and/or waste streams in U.S. plants include centrifugation, evaporation, filtration, and reverse osmosis. These unit operations effectively decontaminate the bulk of the waste water to activity levels that can be discharged directly to the environment or recycled within the facility. The radioactive contaminants are, however, concentrated in the liquid or solid residues (wet wastes) that often require further conditioning such as volume reduction or immobilization prior to storage or disposal. The wet wastes, which include ion-exchange resin slurries, filter cartridges, filter sludges, evaporator

concentrates, oils, solvents, and other miscellaneous materials, can sometimes be classified according to combustibility (e.g., resins, oils, and other organics), but compactibility per se rarely applies; any volume reduction, if possible at all, is usually accomplished by removing associated water by physical means such as decantation, filtration, centrifugation, or evaporation.

The amounts and characteristics of the low-level radioactive wastes generated at any nuclear facility, as well as the subsequent handling procedures required, have direct bearing on the choice of an appropriate treatment or conditioning method. Some of the physical and chemical volume reduction (VR) methods suggested for dry LLW, including most waste metals, and for wet waste concentrates and residues are listed in Table 2. For most organic materials (solid and liquid) some type of incineration appears to be a logical choice. The VR methods that seem most promising for aqueous wastes that require some type of immobilization or solidification are: calcination, crystallization, drying, thin-film evaporation, or an appropriate form of incineration. All VR methods must ultimately take into account any secondary wastes (e.g., off-gas scrubber solutions) that may be generated in the process, as well as any treatment that may be required to convert such secondary wastes to a form suitable for storage and/or disposal. There are both similarities and some marked differences in the LLW treatment methods used at Government installations and those used in commercial fuel cycle plants or private institutions, as will be touched on in the following discussion.

Table 2. Some of the volume reduction methods suggested for use with low-level radioactive dry wastes and wet waste residues

PHYSICAL METHODS		CHEMICAL METHODS
Compression		Calcination
Baler or Compactor		Incineration ^{a,b}
Crystallization		Acid digestion
Evaporator		Agitated hearth
Tray		Controlled air
Dismantling		Cyclone drum
Drying		Fluidized bed
Fixed or Fluidized Bed		Microwave/Gas plasma
Evaporation		Molten glass (Joule heating)
Hot carrier fluid		Molten salt
Thin film		Multiple hearth
Melting		Pyrolysis (Controlled air)
Size Reduction		Rotary kiln
Chopping	Grinding	Slagging pyrolysis
Cutting	Shredding	
Crushing		
Smelting		

^aTaken, with modification, from FMC Corporation, Engineered Systems Division, Selection of Waste Treatment Process for Retrieved TRU Waste at Idaho National Engineering Laboratory, Final Report, Contract K-1010, FMC Document No. R-3689, prepared for EG&G Idaho (November 10, 1977).

^bAlso useful for organic liquids.

The foregoing considerations are reflected in the evaluation of the individual unit operations that are described below for treating dry wastes and wet wastes, respectively.

DRY WASTE TREATMENT

Noncompactible and/or noncombustible LLWs often require special treatments depending upon their particular individual characteristics. They can range in size from small pieces of scrap or waste metal to very large pieces of discarded equipment. The radioactive contamination may be deposited only on the surface, or it may be distributed throughout the entire structure. These wastes, to date, have represented a relatively small fraction of all the LLW generated, but increasing decommissioning activities are steadily adding to their generation rate.⁵ For example, a large LWR is expected to generate about one-third as much LLW [$16,700 \text{ m}^3$ ($6 \times 10^5 \text{ ft}^3$)] during decommissioning as in an expected 40 years of normal operation [$5.1 \times 10^4 \text{ m}^3$ ($1.8 \times 10^6 \text{ ft}^3$)]. Practically all of this decommissioning waste will be noncompactible and/or noncombustible.

The huge total volume of dry compactible and/or combustible waste generated in all radioactive operations suggests the need for suitable volume reduction methods. This is especially true for installations that have little or no accommodation for onsite storage. As mentioned before, the closing of the West Valley, Maxey Flats, and Sheffield disposal sites has resulted in sharply increased waste transportation costs and, in turn, has provided greater

incentive toward volume reduction. A line diagram showing options for the management (pretreatment, collection, treatment, volume reduction, immobilization or solidification, packaging, container handling, and storage) of dry LLW in nuclear facilities is given in Fig. 1.

Physical Methods for Dry Waste

Small contaminated noncompactible and/or noncombustible waste items usually require no special treatment before packaging for disposal. Larger pieces of equipment sometimes require surface de-contamination by chemical means, or perhaps by electropolishing hydrolaser, or sandblasting, prior to size reduction by dismantling or torch-cutting. An electropolishing, or a sandblasting, technique can sometimes be used to reduce metal surface contamination to the point where reuse or recycle is possible instead of discard. Among the physical means most commonly employed at Department of Energy (DOE) sites for treating dry compactible and/or combustible wastes are chopping or shredding, baling, and compaction. Commercial facilities normally use in-drum compaction for these wastes. Bulk volume reduction factors of 2 to 3 for chopping or shredding, and 3 to 10 for baling and compaction, are usually attainable.

Chemical Methods for Dry Waste

The dominant chemical method used for treating dry LLW is incineration. In the U.S. commercial fuel cycle only one small nuclear power plant (Yankee Rowe)⁶ now uses an incinerator for

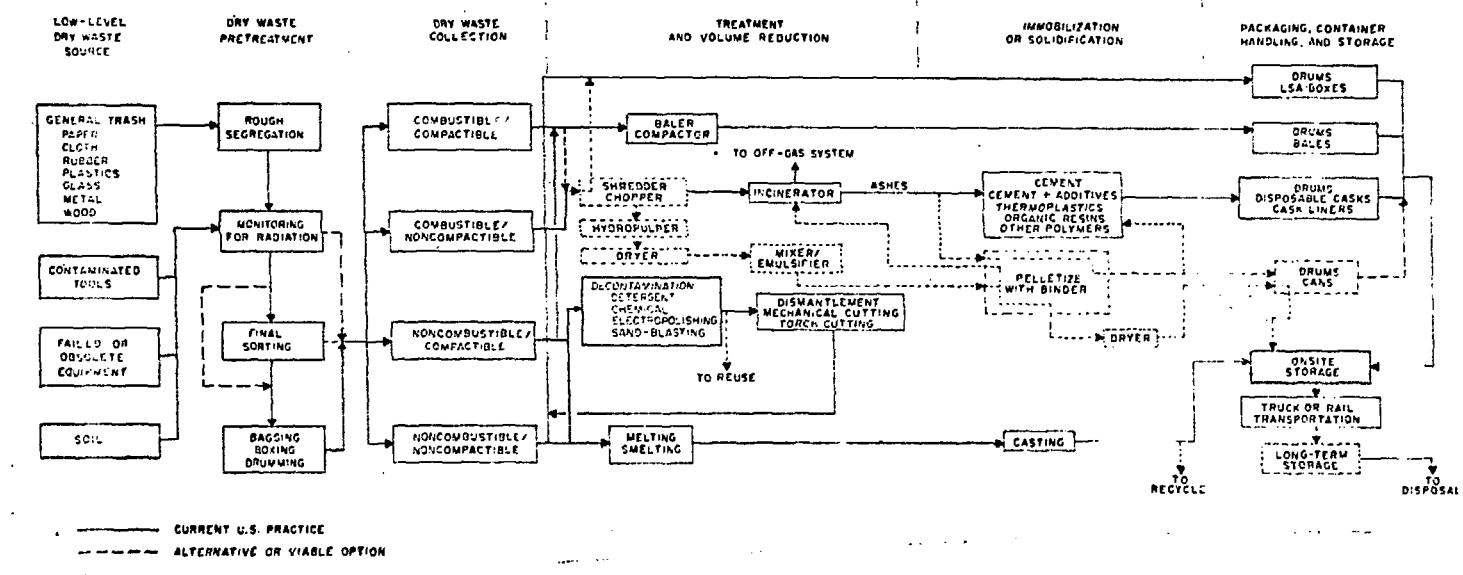


Fig. 1. Flow diagram illustrating the management of dry low-level radioactive wastes in nuclear facilities.

combustion of "essentially nonradioactive" wastes although several fuel fabrication plants use incineration routinely.² A number of commercial institutions (e.g., hospitals or medical schools) incinerate their LLW, but obtaining a license to do so has apparently been quite difficult.⁷ However, a recent survey by the Nuclear Regulatory Commission (NRC) indicates that incineration of scintillation counting fluids, which comprise the major volume portion of institutional wastes, is among the most viable alternatives for disposing of these wastes.⁸ Several DOE sites have had active incineration R&D programs over the last few years, each investigating a selected type of incinerator. In 1977, an exhaustive study and evaluation of these and other commercially-available incinerators were made in an effort to facilitate decision-making with respect to treatment of retrieved Idaho National Engineering Laboratories (INEL) transuranic (TRU) wastes.⁹ Even though the radiochemical properties of Government wastes may be somewhat different from those of combustible commercial wastes, the same incineration methods might be applicable if some modifications are made. Treatment of the off gases-from burning commercial radioactive wastes may be more complex because they are likely to contain higher concentrations of volatile radioactive fission and activation products than TRU wastes. The radioactive heavy metals in TRU wastes form nonvolatile oxides. The nature of the retrieved INEL wastes (especially the high soil content) imposes unique constraints on the treatment process that are not necessarily required in processing other radioactive wastes. A brief description

of the incineration processes studied at each DOE site (the types of waste each is meant to treat and some possible limitations) is given in ref. 10.

Fluidized-bed dryer/incinerator

A fluidized-bed dryer/incinerator that uses separate vessels for drying and incineration (Fig. 2) has been developed by Aerojet Energy Conversion Company. In this system, aqueous waste is sprayed into the dryer vessel containing a starter bed of granular material, such as fine sand, which is fluidized by a stream of preheated air. The bed is further electrically heated to an operating temperature of \sim 450 to 480°C. Water is evaporated and salts that are unstable at these temperatures are decomposed, leaving anhydrous, free-flowing solids in the bed. The off-gas from the calciner vessel contains steam and other gases as well as fine solid particles. A cyclone separates most of these solids from the gases and drops them into a product storage hopper. To maintain a constant bed depth as more waste is dried, a portion of the bed is periodically removed through a screw conveyor, cooled, and moved to the product storage hopper. Hot gases from the cyclone pass through a wet scrubber/waste pre-concentrator, where heat from the gases is used to preconcentrate feed to the dryer up to about 28 wt % solids. At the same time, virtually all remaining particulates are scrubbed from the gases. The off-gas from the preconcentrator then passes through a condenser. Most of the off-gas (air at this point) is routed through a gas

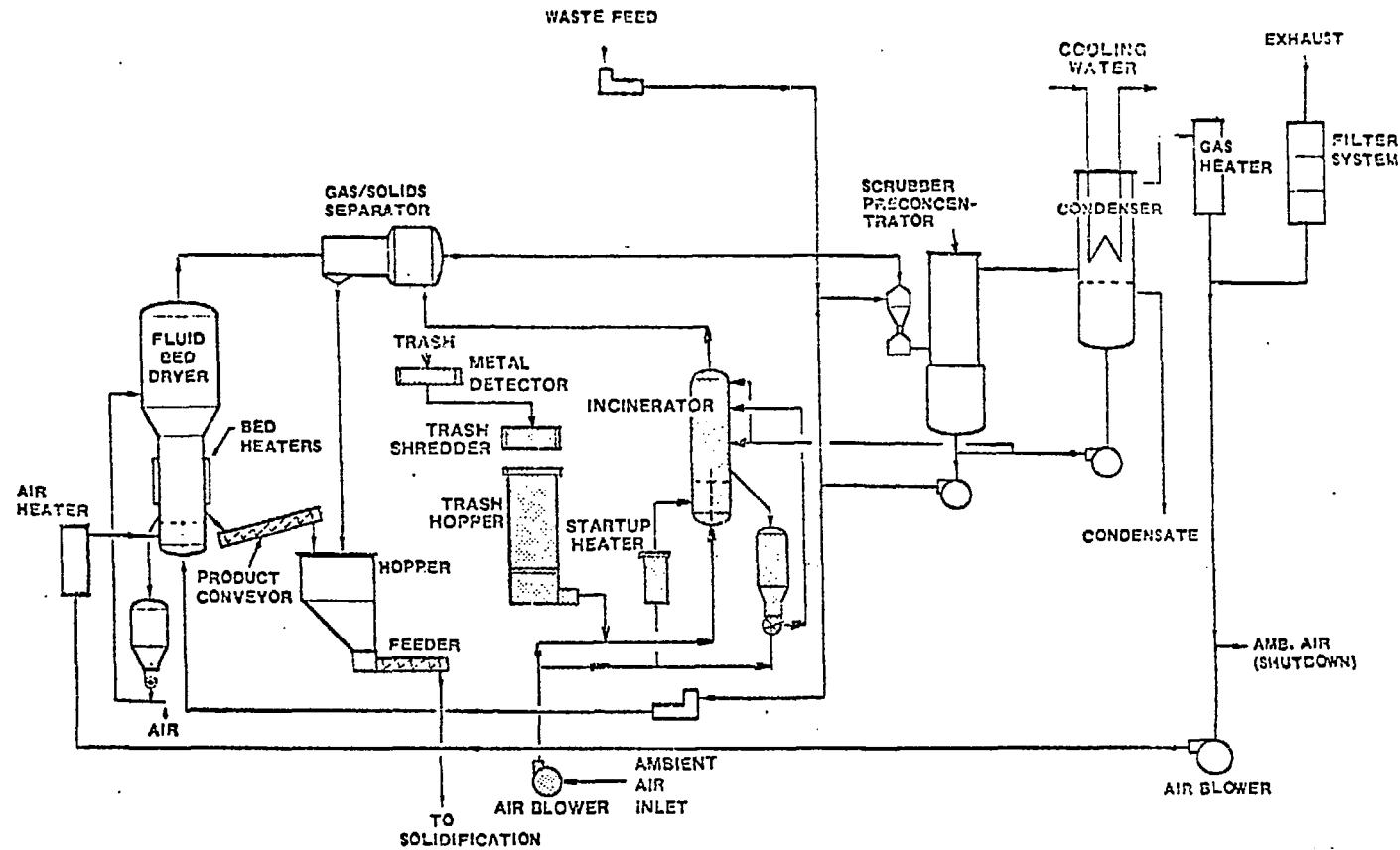


Fig. 2. Flow diagram of the fluidized-bed dryer/incinerator developed by the Aerojet Energy Conversion Company.

heater prior to recompression and recycle to the dryer. A small portion of the exhaust air is continuously bled from the system via an absolute filter and a charcoal absorber, monitored for radiation and discharged to the atmosphere.

While aqueous wastes are volume reduced in the fluidized-bed dryer, dry combustible wastes and contaminated oils are volume reduced by burning in a separate, fluidized-bed incinerator (Fig. 2). The bed in the incinerator is, as in the dryer, an inert material such as sand. The bed is preheated to $\sim 540^{\circ}\text{C}$ with hot air. Combustible wastes pass through a metal detector, are shredded, and blown into the incinerator. Combustion takes place at a temperature of $\sim 790^{\circ}\text{C}$. Both solid and gaseous products of combustion are removed as an overhead stream and are treated in the off-gas system described above for the fluidized-bed dryer. Since essentially no material is added to the bed, no removal of bed material during operation is normally required.

A full-scale, prototype, fluidized-bed dryer/incinerator has operated successfully for over 3,500 h with nonradioactive aqueous wastes (e.g., solutions of Na_2SO_4 and $\text{Na}_2\text{B}_4\text{O}_7$) and for over 500 h on combustible wastes. This unit can dry aqueous wastes at a rate of 75 to 450 l/h and burn dry combustible wastes at ~ 220 kg/h. Volume reduction factors achieved are: 5 for 20 wt % Na_2SO_4 solutions; 10 for 12 wt % H_3BO_3 solutions; and 70 for uncompacted, dry, compressible wastes. The decontamination factor (DF) for ^{131}I across the dryer and off-gas cleanup system was determined to be 10^5 .

for all chemical forms of iodine tested. Several units similar to the prototype have been ordered for LWRs, but these will not be operational for 2 to 5 years.

Fluidized-bed calciner/incinerator

A volume reduction system that is both a fluidized-bed calciner and incinerator has been developed by Energy, Incorporated/Newport News Industrial Corporation. Liquid (aqueous and organic) and dry, combustible, solid wastes as well as dewatered resins have been treated in a prototype unit. The system is similar in principle to the one described above. However, three major differences are: the Newport News unit uses a single vessel as calciner or incinerator; it uses in-bed combustion to heat the bed; and it operates under slight vacuum whereas the Aerojet unit operates under slight pressure. Kerosene, or a similar fuel, is injected into the bed and burned. By using an excess of fluidizing air, essentially all of the solid products formed during calcination at $\sim 400^{\circ}\text{C}$ or incineration at $\sim 1000^{\circ}\text{C}$ are removed from the bed with the off-gas. The bulk of these solids are removed from the off-gas in a dry cyclone and fall into a product storage hopper. Particulate solids remaining in the off-gas stream are removed by a quench tank and wet cyclone/scrubber arrangement. The off-gas passes through a condenser and demister to remove residual moisture before passing through high-efficiency particulate air (HEPA) filters and an iodine absorber prior to discharge.

The commercial nuclear power industry has no operating experience with the fluidized-bed calciner/incinerator. Several units have been ordered for installation at LWRs. These units are designed to treat ~90 kg/h of dewatered resins; ~65 kg/h of shredded, dry combustible wastes; or ~20 kg/h of 25 wt % Na_2SO_4 solutions. Volume reduction factors expected are: 5 for 25 wt % NaSO_4 solutions; 10 for 12 wt % H_3BO_3 solutions; 20 for dewatered resins; and 80 for shredded, dry, combustible wastes. Although no commercial nuclear power industry experience exists with such units, considerable experience has been gained at INEL where a similar type calciner is used to treat high-level radioactive aqueous wastes.¹¹

Volume Reduction Factors for Waste Incinerators

Some typical volume reduction factors that can be expected for incinerator units which are suitable for treating low-level radioactive wastes are given in Table 3. Volume reduction factors of 20 to 45 are achievable with most dry combustible wastes. However, higher volume reduction factors (i.e., 70 to 80) have been reported for the fluidized-bed incinerators of the types described above. As noted in the table, volume reduction of some noncombustibles may be realized by melting in place of incineration. Wet solid wastes, when incinerated, will usually be reduced in volume by a factor between 2 and 20, depending upon the basic chemical composition of the waste. Aqueous concentrates, which are actually calcined rather than incinerated, have shown that volume reduction factors in the

Table 3. Some volume reduction (VR) factors^a expected for incinerator units used with low-level radioactive wastes

Waste type	VR factor	Waste type	VR factor
Combustible/Compactible	20 to 45	Aqueous Concentrates ^b	5 to 10
Combustible/Noncompactible	20 to 45	Special Aqueous Solutions ^b	~10
Noncombustible/Compactible	(c)	Oils	>100
Noncombustible/Noncompactible	(c)	Other Organic Liquids	>100
Filter Cartridges	(d)	Membranes (UF and RO)	(d)
Spent Resins	15 to 20	Biological	~20
Slurries and Sludges ^b	2 to 5		

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^aVolume reduction factors for incineration step only; solidification of ash is not considered, and secondary wastes from off-gas cleanup are neglected.

^bThese wastes are not actually incinerated but calcined to achieve VR in some units.

^cThese wastes are not actually incinerated but melted to achieve VR in some units.

^dNot usually applied to this waste type.

range of 5 to 10 can be readily attained. Oils and other organic liquids normally show volume reduction factors greater than 100 when burned. At the present time, only small fractions of the wet low-level wastes generated in the nuclear industry in the United States are actually incinerated. The granular solids and/or ashes from a dryer, calciner, and/or incinerator may be rendered less mobile by incorporation in a binder or solidification agent. These agents include asphalt, cement, cement/asphalt, cement/polymers, glass, polyethylene, as well as urea-formaldehyde, unsaturated polyester, and epoxy resins.

WET WASTE TREATMENT

Wet wastes in this paper include emulsions, solutions, slurries, and sludges of both inorganic and organic materials contaminated with low levels of radioactivity. As mentioned previously, they arise during the treatment of process or dilute waste streams as nuclear facilities move toward a practice of "maximum recycle or reuse" (of water and other solvents) or "near-zero release" (of radioactivity or other potentially hazardous materials). To be effective, maximum recycle or reuse requires streams that are essentially free of impurities which interfere with the process and are low in radioactivity which can increase in-plant personnel exposure. To be achievable, near-zero release requires effluent streams that have been highly purified (decontaminated) or detoxified. A line diagram showing options for the management of wet LLW in nuclear facilities is given in Fig. 3.

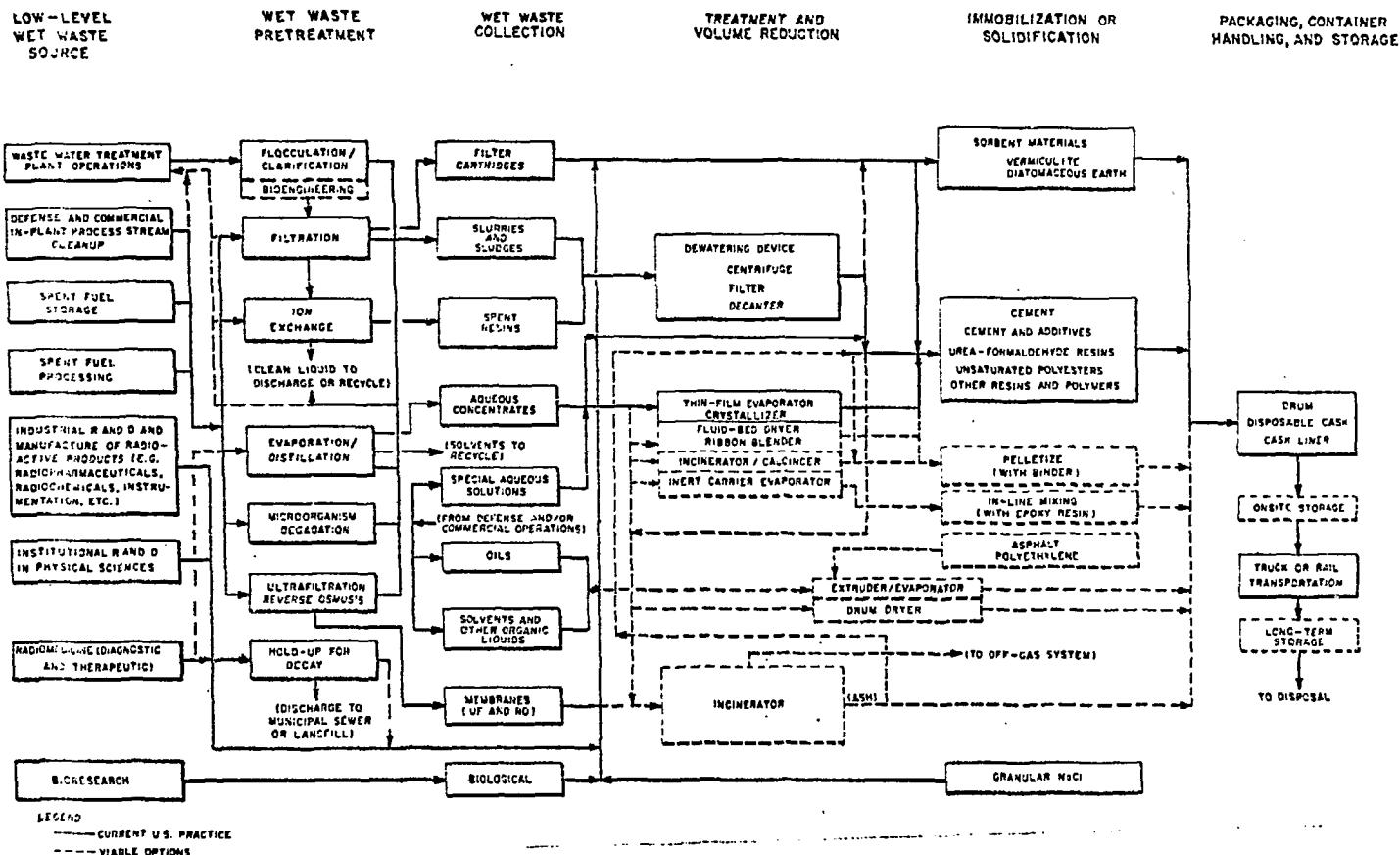


Fig. 3. Flow diagram illustrating the management of wet low-level radioactive wastes in nuclear facilities.

Among the unit operations commonly used in the cleanup of in-plant process streams and LLW liquids are: absorption, biodegradation, clarification, coagulation, evaporation, filtration, flocculation, ion exchange, precipitation, reverse osmosis, sedimentation, and ultrafiltration. The small quantities of radioactivity present in the dilute process and waste streams are concentrated in various types of wet sludges and precipitated salts, ion-exchange resins, and evaporator concentrates that must be stored and conditioned for disposal. These unit operations also serve to reduce the volume of waste that must be prepared for transport to a burial ground.

Physical Methods for Wet Waste

Presently, at LWR plants, the most frequently used unit operations which employ physical or mechanical methods for treating process and LLW liquids are evaporation and filtration. Used to a lesser extent are centrifugation, reverse osmosis, and ultrafiltration.

Evaporation

Evaporation is the process by which the volatile and nonvolatile components of a solution or slurry are separated via boiling away the volatile component. It is a unit operation that has wide application at nuclear facilities for reducing both waste volumes and the amount of radioactivity in liquid effluents. Evaporation can be used on solutions or slurries having widely different compositions and concentrations; however, it is most effectively used on radioactive liquids having high concentrations of solid impurities. Evaporators may be

categorized in a number of ways depending usually upon the classifier's interest. Since a measure of evaporator performance, as judged by the DF achieved,^{12,13} is the prime interest with radioactive liquids, a division that seems to bring out this point has been selected. The types of evaporators that are used or proposed for use at nuclear facilities have been roughly divided into four main categories:¹³ natural circulation (NC), forced circulation (FC), spray film (SF), and submerged U-tube (SU). Examples of these types are illustrated in Figs. 4 and 5. The results of a recent survey¹³ show that a mean system (feed-to-condensate) DF of about 10^4 can be expected for nonvolatile fission and corrosion products treated in single-effect NC and FC evaporators and of about 10^3 in SF and SU types. The mean system DF expected for iodine is about 10^3 in the NC and FC types and about 5×10^2 in the SF and SU types. Evaporators of the types illustrated in Figs. 4 and 5 are expected to yield concentrates containing 20 to 25 wt % solids. The solids content of the concentrate can be at least doubled by using a wiped-film evaporator or an evaporator/crystallizer.

The wiped-film evaporator is considered as a forced-circulation type since it uses mechanical energy to improve heat transfer. These machines are also called agitated-, scraped-, thin-, and turbulent-film evaporators. The heating surface consists of a single, large-diameter, cylindrical or tapered tube. The liquid being concentrated is spread out into a thin, highly turbulent film by the blades of the rotor. Wiped-film evaporators are usually operated in a once-through

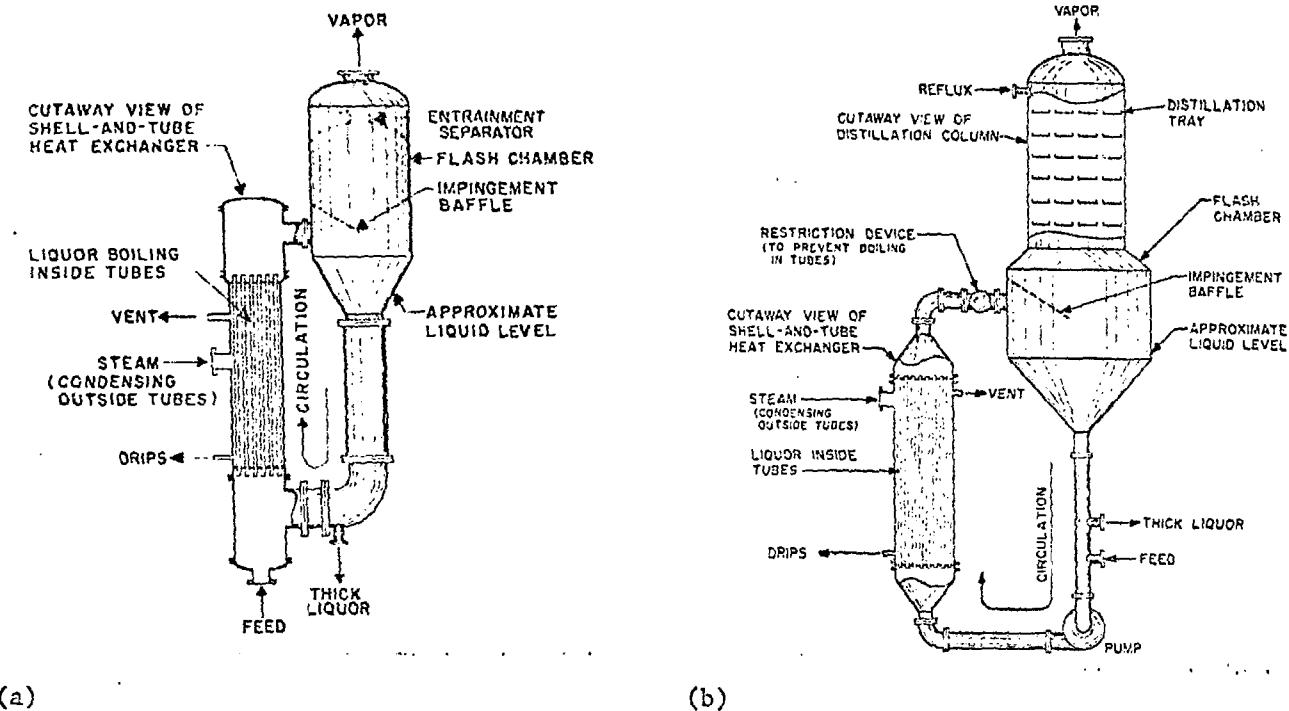
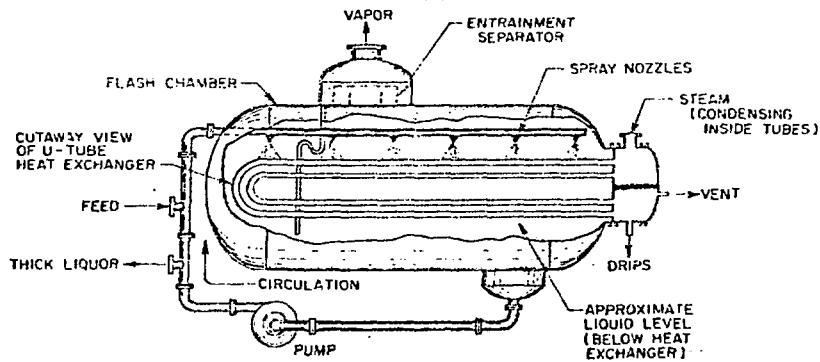
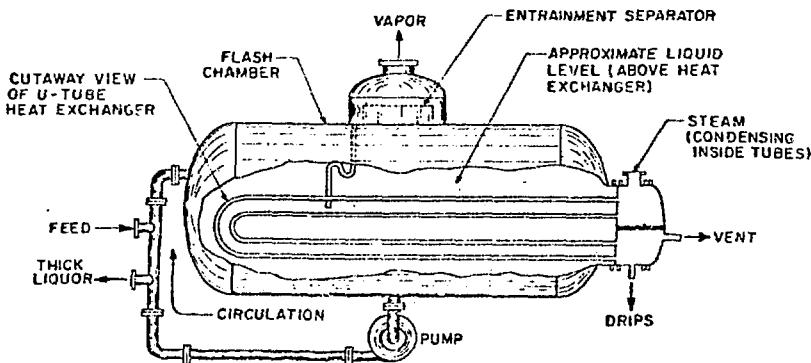


Fig. 4. Typical evaporators used at LWR plants. (a) natural-circulation, rising-film, long-tube vertical evaporator with an external heater and (b) forced-circulation evaporator with an external, vertical, single-pass heater and restriction device to prevent boiling in tubes.



(a)



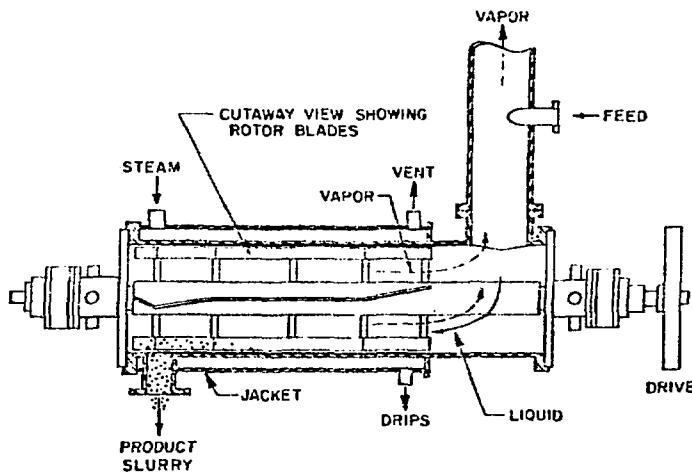
(b)

Fig. 5. Typical evaporators used at LWR plants: (a) spray-film evaporator with horizontal U-tube heater and (b) submerged U-tube evaporator.

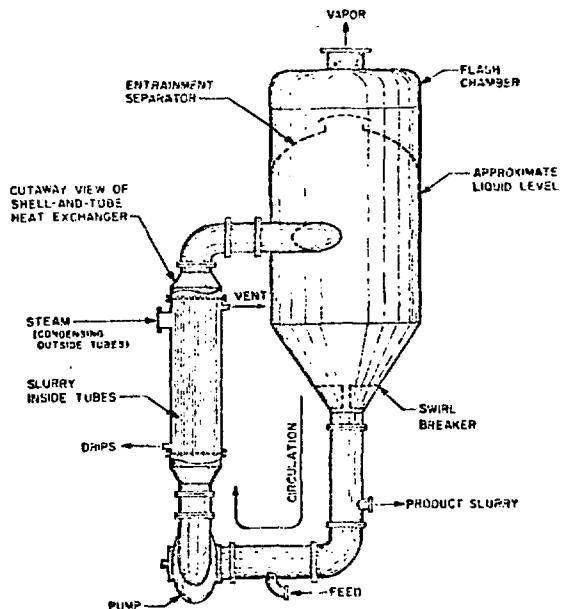
mode. They are available in either horizontal (Fig. 6a) or vertical models. They have been studied extensively at Savannah River for reducing the volume of the high-level aqueous waste stored there. Since 1951, roughly 50 units have been used to treat radioactive liquids.¹⁴ Evaporation techniques for the crystallization of salts have been used for centuries. Forced-circulation evaporators are well suited to concentrating salting liquors, and so function as evaporating crystallizers (or crystallizing evaporators). In this study, such a unit is referred to as an evaporator/crystallizer since whether it is an evaporator or a crystallizer is largely a question of the shape of the solubility curve of the material on which it operates. An evaporator/crystallizer is illustrated in Fig. 6b. Evaporator/crystallizers have been used since 1974 to reduce the volume of high-level aqueous waste at Hanford. Several have been purchased recently for treating radioactive liquid wastes in LWRs.¹³

Filtration

Liquid filtration refers to a process in which undissolved particulate solids suspended or carried by a liquid are separated from the liquid by forcing it through a porous body. The mixture of solids and liquid to be separated is often called the feed, feed slurry, influent, or prefilt. The solids to be separated are sometimes termed contaminant, crud, dirt, or suspended solids. The liquid passing through the porous body is generally called the effluent or filtrate. The porous body is variously named the filter



(a)



(b)

Fig. 6. Evaporators yielding slurry products: (a) wiped-film evaporator and (b) evaporator/crystallizer.

medium, medium, or membrane. The term *septum* is used sometimes to mean the filter medium. However, *septum* is also used frequently to mean the support for the filter medium or the partition between the prefilter and filtrate. Filters of various types are widely used throughout the different systems of nuclear facilities to remove suspended solids from liquid streams that are to be recirculated (recycled) within or discharged to the environment. The need for removal of these solids may stem from requirements for low radioactivity level or freedom from unwanted (interfering and abrasive) materials. For example, radioactive suspended solids may be removed from a stream to reduce the level of personnel exposure during maintenance and refueling operations or to avoid unnecessary wear on pump seals. Occasionally, the suspended solid represents a material of value whose recovery is desirable (e.g., resin slurries). Often, suspended solids are removed because they can interfere with a subsequent operation or process (e.g., ion exchange or evaporation). Frequently, they are removed to prevent their abrasive attack on sensitive instrumentation located downstream. The use of filtration at LWR plants is described in ref. 3.

Filtration and filters may be categorized in a number of ways, several examples of which are given in ref. 3, depending usually upon the classifier's interest. Since the characteristics and amounts of wet wastes generated by filters are among the prime interests of this study, a division that seems to bring out these points has been selected. The many types of filters that have been used for liquids

in nuclear facilities have been roughly divided into two main categories: disposable and reusable. Under each of these main categories, several types of filters are included. Most of the types which are in use or are proposed for use in nuclear facilities are listed in Table 4. A typical disposable type (cartridge) is shown in Fig. 7a and a reusable type (pressure precoat) in Fig. 7b.

Centrifugation

Like filtration, centrifugation is another unit operation used to remove suspended solids from radioactive liquid streams. Centrifuges are mechanically driven rotating devices which employ centrifugal forces to separate solids from liquids. By taking into account the centrifugal force developed, the range of throughputs normally obtained, and the solids concentration that can be handled, three main types can be identified:¹⁵ namely, solid-bowl centrifugal (or centrifugal filter) and tubular- or disk-bowl centrifuges. In the United States, centrifuges have been installed at several LWR plants (e.g., Millstone 1, Monticello, and Vermont Yankee) to dewater resins and/or filter sludges. The defense high-level waste solidification process under development at Savannah River uses a centrifuge to separate the sludge and salt slurry from the waste tanks into a salt solution and sludge.

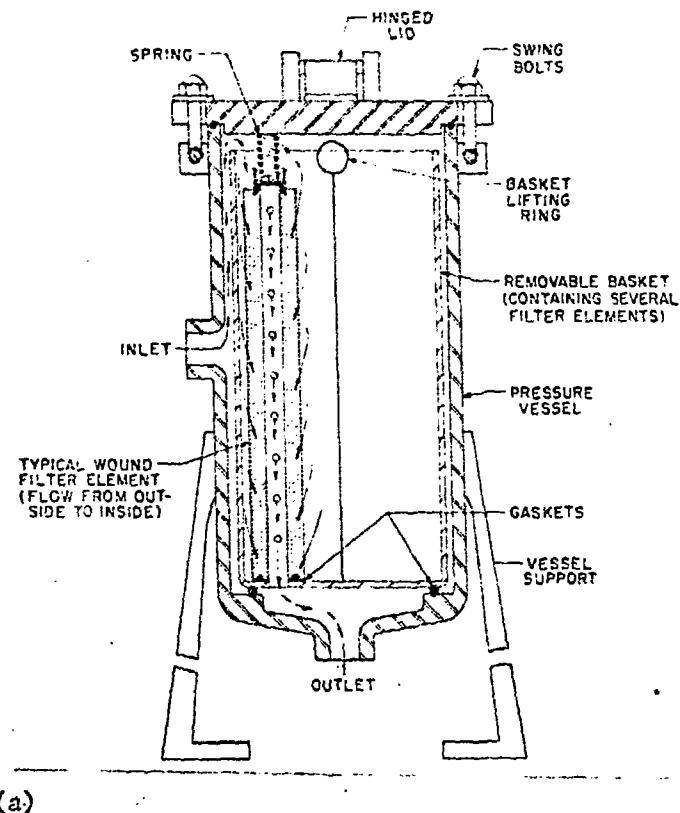
Reverse osmosis

The phenomenon of osmosis may be defined as the spontaneous passage of liquid from a dilute to a more concentrated solution

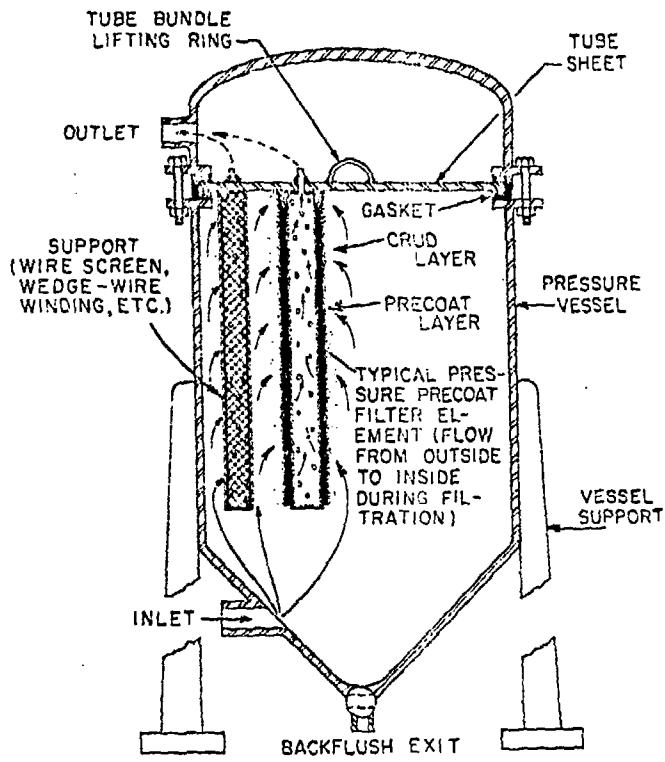
Table 4. Filters for liquids in nuclear facilities

Type of filter	Type of filter
<u>Disposable</u>	<u>Reusable without precoat</u>
Pleated paper cartridge ^a	Partially-cleanable metallic ^a
Pleated wire screen ^a	Porous ceramic ^a
Wound cartridge ^a	Stacked etched-disc ^a
Woven mesh bag ^a	<u>Reusable with precoat</u>
<u>Reusable deep bed</u>	Backflushable tubular bundle ^a
Crushed coal	Centrifugal discharge ^a
Ground walnut shells ^a	Clamshell ^a
Sand	Flat bed ^a
<u>Reusable magnetic</u>	Pressure leaf
Magnetite bed ^a	Rotary vacuum
Electromagnetic ^a	

^aFilter types used or proposed for use at LWR plants in the United States.



(a)



(b)

Fig. 7. Typical filters used at LWR plants: (a) disposable-cartridge filter illustrating liquid flow from outside to inside of element and (b) tubular-support pressure-precoat filter.

across a semipermeable membrane which allows the passage of the solvent but not the dissolved solids. This transfer continues until the pressure, defined as the osmotic pressure, is large enough to prevent any net transfer of the solvent to the more concentrated solution. If pressure [$\sim 2 - 10 \text{ MPa}$ ($300 - 1,500 \text{ lb/in.}^2$)] is applied on the concentrated side at higher than the osmotic pressure, the phenomenon of reverse osmosis (RO) takes place, wherein the solvent flows from the concentrated side to the more dilute side. The semipermeable membrane normally used today is a modified cellulose acetate film. The membranes have pore sizes less than $0.001 \mu\text{m}$ and thus can retain organic molecules with molecular weights between 1,500-500.¹⁶ Various types of RO units with varying degrees of prefiltration requirements for acceptable operation are on the market today. There are spiral-wound, hollow-fiber, and internal- or external tubular designs with feed pretreatment for suspended solids ranging from prestraining to filtering out five-micron particles to prevent fouling. Reverse osmosis units are usually composed of a number of stages so that the permeate (solution that passes through the membrane) from one stage is the feed to the next stage. That portion of the feed which is rejected by the membrane is called blowdown, brine, or concentrate. To prevent hydrolysis (which causes deterioration) of the membrane, the temperature of the feed should be relatively low (less than 50°C) and the pH should be neutral or slightly below (i.e., 4 to 7). Applications of RO units to radioactive liquids are reviewed in ref. 16.

In the United States, RO is used or has been evaluated by several LWR plants. At R.E. Ginna, a 0.13-l/s (2-gpm) unit is used on laundry waste. The 0.13-l/s (2-gpm) pilot-plant unit tested at H. B. Robinson 2 led to the installation of a 3.2-l/s (50-gpm) full-scale unit at Brunswick (both plants belong to Carolina Power & Light) for treating floor drain waste. The unit has seen limited operation. A number of RO units are on order by LWR plants (e.g., Comanche Peak, LaSalle, and Zimmer). The Idaho Chemical Processing Plant (ICPP at INEL) has used a 1.6-l/s (25-gpm) unit on the fuel-storage-pool water. To recover water from sanitary sewage for recycle to cooling towers, the Rocky Flats Plant (RFP) evaluated RO. The results with the 1.3-l/s (20-gpm), 2-unit (brine from the first is fed to the second), pilot plant led to the construction of a full-scale plant which is now undergoing shakedown. The plant has three parallel lines and is designed to operate at 9.5, 19, or 28 l/s (150,300, or 450 gpm).

Ultrafiltration

The process of ultrafiltration is related to reverse osmosis. Contaminated water is forced through the membrane under pressure; the ultrafiltration membrane, however, is considerably more porous than the RO membrane. As a result, all soluble species [except those of high (about 100 or higher) molecular weight] pass through the membrane with the water. Colloids, suspended solids, and high-molecular-weight organic molecules do not pass through the membrane. Because of the porous nature of the ultrafiltration membrane, the

process can be operated with high fluxes at relatively low pressures [$\sim 0.2 - 1.4$ MPa (25-200 lb/in.²)] in contrast to the high pressures required by RO. This advantage stems from the fact that the osmotic pressures of colloids and high-molecular-weight organics are extremely low. Mound Laboratory has evaluated ultrafiltration using a 30-gpm pilot plant and has a program which would lead to the demonstration of a liquid waste treatment system utilizing ultrafiltration, reverse osmosis, and adsorbents. The data collected would be used to design a full-scale waste treatment facility utilizing these three unit operations, either in combination or singly. A pilot-plant ultrafiltration unit has been installed at Tsuruga Nuclear Power Plant (Japan) for testing on actual in-plant streams.

Chemical Methods for Wet Waste

Presently, the most frequently used unit operations which employ chemical processes for treating low-level radioactive liquids are coagulation/flocculation and ion exchange. Used to a lesser extent are incineration (in the United States) and biological treatment. Since coagulation/flocculation and biological treatment are not used at U.S. nuclear power plants, they are not discussed in this paper.

Ion exchange

Ion exchange refers to a process in which a reversible interchange of ions of the same sign (+ or -) takes place in a stoichiometrically equivalent proportion between an electrolyte solution and

a solid phase (ion exchanger) in contact with the solution. A multitude of natural and synthetic materials have ion-exchange properties, for example, slightly soluble ionic crystals, slightly soluble inorganic polymers such as aluminosilicates (zeolites), organic polymers of a carbohydrate type such as cellulose, or of a polypeptide type such as protein, and synthetic organic polymers such as sulfonated cross-linked polystyrene. Of these, the important commercial ion exchangers are the natural and synthetic zeolites and the synthetic organic polymers. The ion-exchange process may be carried out as a batch or column operation. Column operation is essentially a large number of batch operations in series and is the system used in treating radioactive process and waste liquids.¹⁷⁻¹⁹

The ion-exchange system most frequently employed in LWR plants is the mixed-bed system, which consists of a stationary bed containing mixed anion and cation resins. The liquid stream is fed from the top of the bed, and percolates downward through the bed during operation of the process. When ionic contaminants begin to appear in the effluent in significant quantities (breakthrough), the bed is considered exhausted or loaded and the feed is discontinued. The bed is then regenerated (followed by rinsing with deionized water) or else it is discarded and replaced with fresh resins. These beds contain resins with a particle size range of 0.4 to 0.7 mm. For removal of both cation and anion contaminants from liquid waste, two separate ion exchange beds in series, a cation bed ahead of an anion bed, are sometimes used instead of a mixed-bed system. The

separate-bed arrangement may be preferred for the purpose of removing specific radionuclides. Regeneration is much simpler with a separate-bed than with a mixed-bed unit. Inorganic ion exchangers are rarely regenerated.

Decontamination factors achieved in ion-exchange treatment of radioactive waste liquids vary in accordance with the type and composition of the stream, the radionuclides in the liquid, type of exchanger, regeneration methods, and operational procedures. With synthetic organic resins, DFs have been reported as low as 2 and as high as 10^5 . Some of the myriad uses of ion exchange at nuclear facilities are reviewed in refs. 17-19.

Incineration

The incinerators described previously have burned, or with proper changes could burn, combustible liquids. A modification of the feed system, or an additional one, may be required for liquid incineration. At the Savannah River Plant (SRP), pilot-plant studies showed that quiescent, open-pan burning in a two-chamber unit can be an effective method for treating contaminated solvents. At the ICPP (INEL), low-level contaminated solvent from the solvent extraction process is steam distilled to remove most of the contamination, then burned in a solvent burner.

Physicochemical Methods for Wet Waste

While coagulation/flocculation, precipitation, and sedimentation are individual processes, they are interrelated, and are often carried

out in a single vessel. Although an ion-exchange column filled with bead resins of ~ 0.5 mm does some filtering, filtration is not its intended function. However, when these resins are finely ground ($>90\%$, <0.03 mm) they can function as both filter media and ion exchangers in a single vessel. Such operations, in which chemical and physical processes take place more or less simultaneously in the same vessel, are referred to as physicochemical (or combined) methods in this paper.

Coagulation/flocculation/sedimentation

Although coagulation/flocculation/sedimentation is widely used at U.S. nuclear research and production facilities, it is not used at U.S. nuclear power plants, as stated earlier.

Filtration/demineralization

Consideration of the improved kinetics in ion exchange with small particle sizes has led to the development of fine powdered ion-exchange resins. Since the particles are so small ($>90\%$, <0.03 mm), the powders can also serve as filter media. Use of such powdered resins involves coating the elements of a standard, tubular-support, pressure-precoat filter (see Fig. 7b) with thin layers (~ 3 to 12 mm) of resin. In this arrangement, the resin functions both as a filter and as an ion exchanger. Such units are used extensively at LWR plants for cleaning in-plant streams that have low suspended solids loadings and low ionic strengths (e.g., coolant water).^{3,18,19} The spent powdered resins are not regenerated.

CONCLUSIONS AND RECOMMENDATIONS

There are several areas in the treatment of low-level radioactive wastes that could benefit from more intensive research, development, and demonstration (RD&D). Among them are volume reduction methods, organic liquid and waste metals treatment, and waste solidification techniques. The unit operations and combinations of them used in waste treatment and the methods used in waste solidification systems should be improved. Because the amount of LLW being generated in the United States is expected to continue to increase despite efforts to minimize its generation, the many variables involved in volume reduction systems need to be defined (i.e., a data base needs to be built). Interrelationships of the variables need to be evaluated (i.e., systems analyses carried out) if optimization of volume reduction is to be realized. Too often in the past, secondary wastes which arise from the treatment of primary wastes have been ignored. Also, the trade-offs between smaller volume and increased radioactivity level have often been neglected in the assessment of volume reduction processes for LLW. For correct perspective, unbiased studies should include any immobilization techniques that may be applied to the residues from a volume reduction process. In the following discussion, the recommended RD&D of treatment methods for dry wastes is again considered separately from that of wet wastes.

Dry Wastes

The dry, combustible and/or compactible radioactive wastes generated in nuclear facilities are similar in that they all contain such things as cloth, rubber, plastics, paper, and wood. The proportions of these wastes vary widely depending upon the activity being conducted at a facility, and a treatment procedure that may be suitable in one case may not necessarily be applicable in another. However, more conscientious segregation of the dry wastes would be beneficial in all instances because it simplifies the choice of a treatment method.

Compaction, shredding, chopping, cutting, and other size reduction methods need refinement and/or development to improve their versatility, especially in remote operations. The problem of airborne radioactive particulates associated with such operations often remains unresolved. Future decommissioning of nuclear facilities will increase the need for improved size reduction methods as well as the demand for trained crews to perform the operations.

In the United States, there has been only limited experience with the incineration of dry low-level β - γ contaminated wastes. The practice was tried and abandoned in the early days of operation at the Shippingport Nuclear Power Plant. Also, after less than satisfactory performance with contaminated wastes, the old incinerator at Yankee Rowe Nuclear Power Plant has been restricted to burning essentially nonradioactive trash. The licensing of incinerators for commercially-generated radioactive wastes is under jurisdiction of the NRC. The

volatility of some fission and activation products (e.g., $^{129,131}\text{I}$, ^{106}Ru , ^{85}Kr , ^{14}C , and ^3H) could pose problems in meeting release requirements if an off-gas system is not properly designed and installed. Not enough is known about the composition of the off-gases that could be expected from the combustion of the various types of LLWs that have been previously described in this paper. A study should be made on the effects of eliminating the use of problematic materials [e.g., polyvinylchlorides (PVCs)] whenever possible, and should include other possible ways to accomplish off-gas cleanup (e.g., more efficient scrubbing and use of catalysts or sorbents).

Although incineration technology has advanced significantly in recent years, the treatment of the off-gases from combustion of β - γ contaminated wastes needs thorough demonstration. An evaluation should be made of the secondary wastes that are generated in achieving sufficient off-gas cleanup. Up to now the data pertinent to the design and performance of off-gas systems for use in β - γ waste combustion have been gathered primarily by private vendors of incineration equipment and are therefore not readily available. Independent studies are needed on off-gas treatment systems so that licensing procedures could be simplified.

The trade-offs involved in choosing an incineration process for β - γ contaminated wastes from all sources need precise identification. An evaluation of the abilities and limitations of available incinerator systems in accommodating a wide range of anticipated LLWs should be made. Furthermore, a study to determine the advantages

and disadvantages of large centrally or regionally located incinerators versus smaller onsite units to process commercially-generated radioactive wastes would be useful. The desirability of having more than one type of incinerator (e.g., one for dry trash and one for liquid wastes) at a single site, and definition of the many parameters involved in making such decisions, would be of value. A paper study of the incineration and/or other processes used throughout the world for successfully treating power plant, medical, bio-research, and other low-level β - γ wastes deriving from institutional and industrial applications of radioisotopes could be a valuable guide.

A recurring problem that is closely associated with drying and incineration processes is the difficulty frequently encountered in transferring dry powders or ashes (e.g., hoppers clog, screw conveyors jam, and moving belts slip). The development of reliable remote methods for achieving quantitative transfers would be a substantial contribution to present waste treatment practices.

Wet Wastes

The variety and complexity of wet LLWs preclude easy solutions to the diverse problems that their treatment presents. An attempt is made here to group the wastes in such ways that common treatment methods can be applied.

The volume reduction of wet, low-level, solid wastes (e.g., spent resins, zeolites, and diatomaceous earth filter sludges) is

usually accomplished by physical means (e.g., filtration or centrifugation) that remove associated water. The presence of abrasives (e.g., diatomaceous earth) in wet wastes can cause undue wear on the metal surfaces they contact during a dewatering process. Choice of properly designed equipment (e.g., case-hardened steel for centrifugal bowls) to match the properties of the solids being dewatered can improve performance and minimize maintenance which, in turn, reduces the volume of secondary wastes. Filtration remains more of an art than a science. A study of the uses of filtration in the nuclear industry, with special attention to performance (e.g., can the amount of precoat used be reduced or can cartridge filter change-outs be minimized) would be helpful in reducing wet waste volumes. Studies directed toward finding better ways of drying wet solid radioactive wastes should be pursued. These might include a physicochemical process such as microwave drying.

As pointed out above, proper equipment design and proper choice of a process for treating a wet waste can reduce the volume of waste for disposal. This also applies to the evaporation of dilute aqueous wastes in nuclear power plants. A high decontamination factor ensures greater possibility of water recycle with minimal secondary waste generation (e.g., spent resins from condensate cleanup). Since the use of evaporators is so widespread in the nuclear industry, an evaluation of the performance of the various types that are used would be an aid in reducing waste volumes. Evaporators and crystallizers are used routinely to reduce the volume of liquid LLWs. Operating

experience shows that evaporators and crystallizers routinely give system DFs (feed to condensate) equal to 10^2 to 10^4 . Theoretical considerations indicate that these DFs should exceed 10^6 . Experiments are needed to determine the reasons for these low DFs and to find ways to raise them. Engineering correlations are needed to describe mist formation versus physical properties and operating conditions. With knowledge of how mists form, designs could be chosen to minimize mist formation. Experimental studies are needed to determine the factors limiting demister performance. If the phenomena which limit demister performance are understood, improved designs can be found. More studies are needed on multiple-effect evaporators [i.e., several evaporators (or effects) connected so that the vapor from one effect serves as the heating medium for the next] to determine if they can give improved (higher) DFs and better energy utilization. Operating experience also shows that an evaluation is sorely needed of the factors (e.g., corrosion, maintenance, controls, etc.) that influence the availability and reliability of evaporators and crystallizers used to treat radioactive liquid wastes.

The oxidation of wet radioactive organic wastes (solid and liquid) has not been adequately studied in the United States. These materials can be greatly volume reduced by some form of incineration, but again proper choice of process and equipment is essential to maximize overall volume reduction. Incineration is already being used in certain limited cases (e.g., for animal carcasses and scintillation fluids at some institutions). However, the rigid specifications imposed on

the releases of radioactivity and particulates in the off-gas make licensing a difficult process, which discourages wider application of this technique. The problems associated with burning all types of wet LLWs should be carefully examined with regard to combustion characteristics and off-gas composition. For example, the recent NRC study⁸ on disposal of scintillation fluids could be extended to include more combustible wet wastes (e.g., ion-exchange resins, lubricating oils, and organic coolants). Some incineration processes may be more amenable to treating these wastes than others, and an unbiased assessment is essential. Some additional study along these lines would benefit not only the power industry, but the health service community and Government installations as well.

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