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Operated By
UNION CARBIDE NUCLEAR COMPANY
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POST OFFICE BOX X
OAK RIDGE, TENNESSEE

COPY NO. 14

DATE: June 9, 1959

SUBJECT: Radiation Hazards from Recycled Reactor Fuel

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ABSTRACT

The radiation hazards associated with recycled nuclear reactor fuels will greatly complicate the handling and refabrication of these fuels. This problem is most serious with U-233 and plutonium fuels where the presence of U-232 and the heavier isotopes of plutonium contribute energetic alpha, gamma, and neutron radiations at levels many times that from isotopically pure U-233 and Pu-239. This report summarizes present knowledge of the radiation hazards associated with recycled fuel and the additional data needed to make a thorough evaluation of these hazards.

DISCLAIMER

RADIATION HAZARD STUDIES

1.0 Summary

The methods by which future power reactor fuels can be refabricated will depend upon the beta, gamma, and neutron radiation intensities created by isotopes other than the fissile or fertile materials. Activities of fission products can be reduced to very low levels by high-decontamination processes and thus eliminate them as serious radioactive hazards during fuel element fabrication. However, the recycle of fuels through power reactors will increase parasitically produced isotope intensities much beyond those found for the naturally occurring elements. The increase in these radiation intensities can impose handling limitations for fuel element refabrication procedures. These handling limitations on nuclear fuels may be imposed because of one or more of the following reasons: (1) direct contact exposure to the hands may be high, (2) total body exposure at operating distances may be above set limits, and (3) internal exposure hazards due to air contamination may be created. The radioactivity contributing in each of these categories are:

(1) For Surface Doses - Beta particles, gamma rays, and fast neutrons from α-n reactions and/or spontaneous fission.
(2) For Radiation at Distance - Gamma rays and fast neutrons.
(3) For Air Contamination - Alpha emitting dusts.

The importance of many of the isotopic contaminants have been given in several references which appear in the open literature.[1,2,3] However, the concern about isotopic contaminants is a relatively new one and has been accentuated by the desire to achieve high irradiation levels and recycle of power reactor fuels.

The three methods of refabrication which must be considered for reactor fuels are:

Direct Refabrication - Refabrication by methods in which the hands are in direct contact 10-40 hours per week and exposures are limited to 100 mr/40 hr to the total body and 500 mr/40 hr to the skin of the hands.

Semi-remote Fabrication - Refabrication by methods which limit direct contact but in which only distance, structural metals or plastics are used for shielding. The facility may be enclosed in an air-tight hood or dry box and can be directly maintained following removal of the work and minor decontamination.

Remote Fabrication - Refabrication by methods which require that all operations be carried out behind heavy shields and under conditions to protect from alpha activity. If fission products can be removed, as by solvent extraction, and isotopic contaminants are the only sources of penetrating radiation, direct maintenance of the facilities is possible after removal of the fuel and minor decontamination. If high fission product activities are present, remote maintenance is also necessary.

The principal activities and the shielding requirements for the refabrication of several fuels are given in the following table. In all cases it was assumed that the fuels have been decontaminated from fission products such that the fission product activity is less than 20% of the total β-γ activity.

A complete study of the problem of fabricating fuel elements from recycled reactor fuel should include an evaluation of the radiation or contamination hazards of proposed fissile and fertile materials including all transmuted radioactive isotopic contaminants. This project should include the following:

(1) A study of nuclear gamma and α-γ ray spectra for the fissile and fertile materials. The biggest unknown is the α-γ spectra from fissile and fertile compounds.

(2) A study of α-n reactions in order to determine α-n conversion ratios for light elements in metals, compounds, and solutions.

(3) A study of spontaneous fission reactions, including the neutron and gamma ray spectra accompanying spontaneous fission.

(4) A study of the x-ray spectra for all proposed fuel materials.

(5) A study of beta emissions and associated Bremsstrahlung x-ray spectra.

(6) A study of alpha contamination and investigation of new dry-box handling techniques. This is especially true for fuels requiring a gamma neutron-shielded facility.

2.0 Status of Knowledge of Radiation Hazards

The buildup of isotopic contaminants during successive irradiations can greatly affect the handling techniques associated with the recycle of nuclear power reactor fuels. Due to high irradiation and many recycles, the levels of alpha, gamma, or neutron activity may govern the procedures which must be followed during all steps of the fuel cycle. In many cases, especially for uranium (U^{235}-U^{238}) fuels, there are two alternatives: (1) complete decontamination and decay with direct handling or, (2) partial decontamination decay with remote or semi-remote handling. For these

Table I

Estimated Shielding Requirements for Recycled Fuels During Refabrication

Fuel	Major Source of Activity	Scale of Operation	Shielding Requirements, cm of Concrete - Operator 100 cm from work
Slightly Enriched uranium	U-237	50 kg	0
Highly Enriched uranium	U-237	1 kg	0
Plutonium Metal	Pu-240,241,242 (∞ recycle)	1 kg	0*
Uranium + 5% Pu	"	50 kg	0
PuF_4	Pu-240,242,(α-n) (∞ recycle)	1 kg	34
U-233 Metal	1000 ppm U-232 (35 days since) isolation	1 kg	66
Thorium Metal	Th-228	50 kg	45
Thorium + 5% U-233	(as above)	50 kg	88

* With the operator only 30 cm from this material, 16 cm of concrete are necessary.

systems the ultimate choice between alternatives can be dictated by economics. On the other hand, the buildup of activities in plutonium, U-233, or thorium systems follows definite patterns, and these activities cannot be reduced to allowable levels unless excessive decay periods are used. If residual fission product activities are low, it appears feasible to use semi-remote lightly shielded refabrication facilities. Decontamination of such a facility is easy, and direct maintenance of equipment is achievable.

The major problem associated with radiation in natural or slightly enriched uranium fuels is due to U-237. However, it appears economically feasible to allow complete decay before completion of each recycle. In this system a gaseous diffusion cascade may be used for re-enrichment with subsequent reduction of the U-236 content, the major source for production of U-237.

For intermediate or high enriched fuels, where U-237 is also the major problem, the economics and the necessary handling techniques are dictated by the achievable burnup per pass. An economic analysis of decay-cooling of highly enriched reactor cores indicates that unless U-235 burnups in excess of 30% are achievable, the full U-237 decay time cannot be economically justified. However, these economic burnup levels are probably attainable for all intermediate or high enriched fuels with the exception of metallic 20% U-235 enriched fast reactor fuels. The recycle techniques for U-235 also depend on the fraction of the total fuel burned in the nuclear power industry that is burned in fully enriched or seed type reactors.

After U-237 has been allowed to decay to the specified activity, it is then possible to perform any of the necessary recycle operations by direct refabrication techniques.

The use of U^{233}-thorium fuels requires semi-remote handling techniques and/or definitely established schedules with complete decontamination for each scrap recycle. Reactor fuel elements containing U-233 can be fabricated by semi-remote techniques provided complete fabrication can be accomplished in 1-2 weeks or less. If the U-233 contains greater than 200 ppm U-232, then a shielded refabrication facility is necessary. Also, the U-233 must be free of light element contamination in order to reduce the alpha produced neutron background. Thorium can be handled directly during some recycle steps and semi-remotely during others. In many cases it may be justified to handle natural thorium during refabrication of reactor fuel elements and hold back the irradiated thorium for 6-12 years to allow decay of Th-228 to natural thorium backgrounds. The practicality of this procedure will depend on the actual cost of thorium and the inventory charge. Thorium having radiation intensities greater than five times that of natural thorium must also be refabricated in a shielded facility. Also, it appears that economic irradiation levels in thorium must be greater than 5000 g/t and thus very active thorium must be fabricated, probably behind shielding.

Probably the most complex radiation problem is that associated with plutonium recycle. Here the problem is due to significant levels of alpha, gamma- and X-radiation, and neutrons from α-n reactions or spontaneous fission. It is conceivable that total radiation intensities at least six times that of pure Pu-239 will be obtained. The level of higher plutonium isotopes after many recycles can be reduced by discarding a small fraction of the plutonium each cycle. However, the value of the discarded plutonium is probably greater than any savings in fabrication costs resulting from this procedure. Approximately 2/3 of the total radiation intensity of 8-9 R/hr at contact from recycled plutonium will be from fast neutrons. However, these neutrons should not be a problem in an unshielded refabrication facility provided semi-remote operations are possible with the workers at least one meter from the fuel element. Direct contact even with gloves must be avoided except in the case of emergency.

Table II lists the handling limitations or maximum allowable working time for U-233 fuel elements. It is assumed that U-233 will be refabricated semi-remotely and that the workers will be at least an average of one meter away from the work at all times.

Table III lists the maximum permissible concentration of each light element to give a neutron (from α,n reaction) dose of 1000 mr/40 hr week at the surface of a kilogram sphere if this was the only impurity present.

Table IV lists the radiation intensities, required shielding or Th-228 decay times for several Th-228 activity levels in recovered thorium. The most probable Th-228 activity in recycled thorium with an irradiation level of 15×10^{21} ϕt/ pass would be 2×10^{8} d/s/kg Th. Such activity would require 45 cm concrete shielding or 11 years decay.

The total radiation dose due to x-rays or γ components in massive Pu-239 which is transmitted through neoprene (30-35 mils) gloves is approximately 900 mr/hr at contact. The actual measurements of various x- or γ-ray intensities from massive plutonium are given in Table V.

Major radiation problems in handling plutonium are due to α,n reactions and spontaneous fission. Light element contamination in plutonium metal can lead to α,n reactions and the resulting fast neutron flux. Table VI lists the values of the maximum permissible concentrations of several light elements in pure Pu-239, Pu-239 + 13.5 wt % Pu-240, Pu-239 + 1000 ppm Pu-238, and Pu-239 + 13.5 wt % Pu-240 + 1000 ppm Pu-238. The allowable concentrations are based on 1000 mr/40 hrs if the particular light element existed alone.

Table VII lists spontaneous fission neutron fluxes and dose rates at the surface of and one meter from kilogram spheres of various fissile or fertile materials as metal.

Table VIII lists the surface contribution of beta particles as a function of beta energy.

Table II

U-233 Handling Limitations

(Basis: One Kilogram U-233 per Operation)

Dose Measured One Meter from U-233 Piece

U-232 Concentration (ppm)	Maximum Allowable Handling Time Since Final Separation (Days) To Reach 5 mr/hr	Necessary Shielding (To 2 Mr/hr) For U-233 Aged 35 days Since Final Separation Cm of Lead	Cm of Concrete
10	45	1.89	11.4
20	24	3.73	21.0
40	15	5.47	29.7
60	11.5	6.47	34.7
80	9.9	7.18	37.9
100	8.5	7.71	40.6
200	8.0	9.40	48.3
400	4.2	11.0	56.1
600	3.5	12.0	60.5
800	2.9	12.6	63.6
1000	2.7	13.2	66.0
1500	2.3	14.1	70.5
2000	2.0	14.8	73.6
2500	1.7	15.3	75.9

Table III

Maximum Permissible Concentration of Each Light Element (if it existed alone) in U-233 Metal

Basis: Neutron dose of 1000 mr/40 hr week at the surface of a kilogram sphere of U-233, a U-232 concentration of 100 ppm and a time after processing of 3-1/2 months.

Light Element	Maximum Permissible Concentration, ppm
Li	45
Be	4
B	10
C	6,050
N	45,000
O	26,400
F	20
Na	168
Mg	480
Al	240
Si	255
P	4,550
S	5,900
Cl	295
A	125
K	810
Ca	2,100
Sc	1,240
Ti	7,250
V	14,000
Cr	18,500
Mn	24,800
Fe	57,500
Co	60,000
Ni	180,000

Table IV

Thorium Handling Limitations

(Basis: Thorium Billet - 10 cm dia. x 60 cm long)

Dose Measured on Surface and One Meter from Billet

Th-228 Activity[a] d/s/kg/ Th	Th-228 Concentration ppm - in Th	Radiation Intensities		Necessary Shielding for Thorium- (Th-228 Daughter Equilibrium) For 2 Mr/hr One Meter from Billet		Decay Time Following Separation Necessary to Reduce Th-228 Act. to that of Natural Th (yrs)
		At Surface Mr/hr	At One Meter Mr/hr	Cm Lead	Cm Concrete	
4.1×10^{6}	1.35×10^{-4}	163	1.2	0	0	0
2.05×10^{7}	6.75×10^{-4}	815	6	2.86	18.9	4.43
4.10×10^{7}	1.35×10^{-3}	1630	12	4.53	27.1	6.33
8.20×10^{7}	2.70×10^{-3}	3260	24	6.20	34.7	8.25
2.05×10^{8}	6.75×10^{-3}	8150	60	8.40	44.9	10.8
4.10×10^{8}	1.35×10^{-2}	16300	120	10.0	52.3	12.7

[a] Th-228 activity in natural thorium in complete equilibrium with decay daughters is 4.1×10^{6} α d/s/kg.

TABLE V

Massive Plutonium Surface Dose Rates[4]

Component Name	Estimated Dose Rate (no absorber) mrep/hr
M X-rays plus photo and conversion electrons	8820[a]
L X-rays	1340[b]
Intermediate	2 or 3[c]
K X-rays and 127 key gamma	50
Hard and general background components[d]	150
Total	10360

[a] Average photon energy of M radiation for uranium is about 3.3 Kev. Intensity reduced to 8% by about 11 mg/cm^2 cellophane.

[b]

Line	Kev	Mrep/hr transmitted through 30-35 mils of neoprene
ULα	13.6	840
ULβ	16.9	430
ULγ	20.2	70
Total		1340

[c] This component shows lines on the krypton counter spectrometer at 34, 40, 52, and 60 kev.

[d] General background radiation includes ionization caused by fluorescence of neoprene and wall elements and may apply to more than the hard component.

Table VI

Maximum Permissible Concentrations of the Most Important Light Element Impurities in Plutonium

Light Element	Maximum Permissible Concentration of Light Elements in Plutonium, ppm (Based on 1000 Mr/40 hr wk at contact.)			
	Pure Pu-239	Pu-239 + 13.5 wt % Pu-240 (50% Pu-240α)	Pu-239+1000 ppm Pu-238	Pu-239 + 13.5 wt % Pu-240+1000 ppm Pu-238
Li	11	7.3	8	5.8
Be	0.47	0.31	0.32	0.24
B	1.59	1.06	1.1	0.79
C	2,575	1,716	1,680	1,265
N	>50,000	33,500	6,100	5,800
O	6,500	4,330	4,200	3,175
F	5.8	3.83	3.7	2.76
Na	98	66	62	47
Mg	1,350	900	760	590
Al	145	97	83	64
Si	280	166	100	84
Cl	1,100	733	580	328
A	212	142	88	73
K	3,200	2,133	1,110	945
Ca	5,650	3,765	1,900	1,630
Sc	28,250	18,850	7,200	6,400
Fe	>250,000	>165,000	55,000	50,000

Table VII

Spontaneous Fission Dose Rates at Surface And One Meter from Kg Sphere

Fuel	Spontaneous Fission Neutron Flux At Surface n/cm^2 - sec	Spontaneous Fission Dose Rate At Surface (Mr/hr)	At One Meter (Mr/hr)
U-233	0.0124	0.0021	5.75×10^{-7}
U-233 + 20% U-234	0.0588	0.0100	2.74×10^{-6}
U-233 + 20% U-234 + 100 ppm U-232	0.0635	0.0108	2.85×10^{-6}
U-233 + 20% U-234 + 1000 ppm U-232	0.1055	0.0180	4.94×10^{-6}
U-235	0.0206	0.0035	9.6×10^{-7}
50% U-235 + 50% U-236	0.108	0.0184	5.04×10^{-6}
U-238	0.440	0.0750	2.06×10^{-5}
Pu-239	0.667	0.114	3.2×10^{-5}
Pu-239 + 20% Pu-240	5600	920	0.252
Pu-239 + 20% Pu-240 + 10% Pu-242	1.06×10^{4}	1810	0.496
Pu-239 + 1000 ppm Pu-238	75	13	3.56×10^{-2}
Pu-239 + 20% Pu-240 + 10% Pu-242 + 1000 ppm Pu-238	1.061×10^{4}	1810	0.496
Equilibrium Pu[a]	2.44×10^{4}	4160	1.04

[a] 36.1533% (49), 14.7144% (40), 8.8214% (41), 40.3109% (42)

Table VIII

Effect of Beta Particles on Surface Dose Rates

Energy (Mev)	Range in Uranium (cm)	Range in Air (cm)	Surface Dose Rates from Uranium Activity = 1 x 10^9 β's/kg/min (Mr/hr)
0.4	0.00316	47.4	50
0.5	0.00604	90	62
0.75	0.0134	200	94
1.0	0.0206	309	125
1.5	0.0353	530	187
1.8	0.0440	660	225
2.3	0.0586	880	286

Table IX

Internal Hazards of Important Heavy Isotopes

Isotope	MPC_{Air} Values[a] (Based on First Critical Organ) μ c/cc	MPL_{Air} Values[a] (Parent Bases) μ g/meter^3
Th-228 Chain	1.9×10^{-12}	2.31×10^{-9}
Th-232 + daughters	1.4×10^{-13}	1.26
Th-234 + Pa-234	2.0×10^{-8}	8.6×10^{-7}
Pa-233	3.0×10^{-7}	1.47×10^{-5}
U-233	3.5×10^{-7}	36
U-235 + Th-231	7.9×10^{-11}	37
U-238	1.3×10^{-11}	39
Pu-238	6.6×10^{-13}	3.8×10^{-8}
Pu-239	5.9×10^{-13}	9.6×10^{-6}
Pu-240	5.9×10^{-13}	2.6×10^{-6}
Pu-241 + Am-241 + Np-237 m	3.1×10^{-11}	2.75×10^{-7}
Pu-242	6.2×10^{-13}	1.6×10^{-4}

[a]The above values may be increased by a factor of four for occupational exposures based on 40 hrs/week only.

Table IX lists the maximum permissible concentration in air for fissile or fertile materials and their contaminating isotopes. This table can be used to evaluate the degree of particulate removal for dry-box air.

Alpha produced neutrons are also a problem in Pu salts such as PuF_4. The alpha-neutron conversion ratio for PuF_4 is 12 n's/10^6 α's. Using this value the source strengths and radiation intensities for a kilogram sphere of PuF_4 can be estimated. These values are listed in Table X.

It would require ~16" of ordinary concrete to reduce the neutron radiation intensity to 2.5 mr/hr at the outside surface of the shield with the PuF_4 placed one foot behind the shield. There is little known about the alpha-neutron conversion ratios for other plutonium or uranium (in particular U-233) salts.

3.0 Proposed Program for Radiation Hazard Evaluation

The purpose of this subproject will be the evaluation of the radiation or contamination hazards of all proposed fissile and fertile materials, including all their transmuted radioactive isotopic contaminants.

(1) Gamma Radiation

The nuclear gamma radiation spectra for most of the fissile and fertile materials and their contaminating isotopes is known to a great extent but is far from complete. A complete determination of all gamma emissions including alpha produced gamma rays is needed. Gamma dose rates should be computed from the known source strengths and then checked experimentally. Alpha produced gamma radiations can be determined at the same time that alpha produced neutrons are found. This experimental program, which is performed with a Van de Graaff accelerator, is described under α-n Activity. The gamma radiation from fuel elements can be checked by means of a graphite ionization chamber.

(2) α-n Activity

The major experimental effort in the entire subprogram will be devoted to the study of α-n reactions. The fast neutrons created by α-n reactions are much more difficult to combat than gamma rays. Experimental effort will be directed in order to determine:

(a) α-n conversion ratios for light elements either as contaminants in metals or as alloys.

(b) α-n conversion ratios for important compounds (mainly compounds of U-233 and Pu) such as fluorides, chlorides, oxides, nitrates, and oxalates.

(c) α-n conversion ratios for several compounds in solution.

Table X

Effects of α,n Reactions in PuF_4 on Handling Limitations

(Basis: 1 kilogram sphere of PuF_4, ρ = 10 g/cc (assumed))

Source Strength (Mass)	= 2.72×10^7 n/sec-kg Pu
Source Strength (Volume)	= 2.06×10^5 n/sec-cc
Neutron Flux at Surface	= 3×10^5 n/cm^2-sec
Neutron Flux at One Foot	= 1.34×10^3 n/cm^2-sec
Neutron Dose Rate at Surface	= 56 R/hr
Neutron Dose Rate at One Foot	= 250 mr/hr

The results of these experiments should lead to a theoretical or empirical correlation of α-n conversion ratios as a function of α-energy, light element contaminant concentration in metals, light-element alloy structure, compound structure, compound anion, and concentration of cation or anion in solution. After these experimental results have been obtained, the radiation hazards can be evaluated and the allowable fabrication or handling methods can be determined.

There are two methods of approach to the α-n problem; both should be pursued. Approach (1) involves determination of the α,n reaction cross-section as a function of alpha energy and light element. This determination involves bombarding a target of some stable compound of the desired light element with Van de Graaff accelerated α-particles and measuring the number and energy of the neutrons. The α produced gamma rays can also be determined at the same time. The energy-range relationship for the α particle in the particular fuel bearing compound is then required for conversion of the α-n cross-section data to the desired α-n conversion ratio. The neutron dose rates are relatively easy to calculate from the α-n conversion ratios.

Approach (2) involves direct measurement of neutron dose rates from many fabricated shapes of the various fuel materials including all materials in the intermediate recycle steps. This method will only give neutron dose rates but will not determine number (flux) or energy of the neutrons. However, this approach would be a good experimental check for approach (1).

(3) Spontaneous Fission

This study involves a cataloging of spontaneous fission rates and calculation of spontaneous fission dose rates from selected fuel elements. The importance of spontaneous fission in Pu fuels has been indicated in Table VII. Additional experimental work involving improvement of neutron dosimetry may be necessary. Spontaneous fission neutron dose rates from proposed fuel elements can be measured with a Hurst Dosimeter. Spontaneous fission dose rate calculations, including the associated fission gammas, are straight-forward and most of the work will involve cataloging dose rates for various fuel element shapes.

(4) X-ray Activity

A complete determination of the x-ray spectra for proposed fuel material is important. However, x-rays are not of great importance if a lightly shielded facility is necessary for other reasons. On the other hand, x-rays are very important due to the high probability of back scattering if unshielded facilities are used. A very thorough experimental study of x-ray emissions with an x-ray spectrometer should be carried out.

(5) Beta-Activity

Beta activity is of minor importance and can be combated by wearing gloves. However, Bremsstrahlung is an important contributor of x-rays. The most needed material for beta activity is a listing of contributors and some experimental checks. Bremsstrahlung spectra should be determined by an x-ray spectrometer.

(6) Alpha Contamination Problems

Alpha contamination is well known and is controlled by dry-box fabrication methods. However, fuels containing both uranium and plutonium will have to be fabricated using large dry-box facilities. A thorough study of the effects of alpha contamination and the determination of the air cleaning requirements are necessary. A design study of air cleaning requirements will be made. The particular problems associated with an alpha-gamma-neutron facility (for fuels having both alpha contamination problems as well as gamma or neutron radiation) will be emphasized.

(7) Experimental Verification

Long irradiated plutonium, thorium, and U-233 will be processed at ORNL for the Plutonium Isotope Cross-Section, Transuranic and Power Reactor Fuel Reprocessing Development programs. These materials will provide important data on the activities of recycled fuels as well as actual experience in handling through the entire recycle scheme.

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[1] Ullmann, J. W., "Heavy Element Isotopic Buildup," Symposium on the Reprocessing of Irradiated Fuels Held at Brussels, Belgium, May 20-25, 1957, TID-7534, Technical Information Service Extension, Oak Ridge, Tennessee, Book 3: 1080-1107, (1957).

[2] Arnold, E. D., Ullmann, J. W., "Choosing Storage Time to Minimize Processing Costs," Nucleonics, 15:80-3, (1957).

[3] Manowitz, B., Richman, D. M., Zwickler, S., "Heavy Isotope Product in Converter Fuels," Brookhaven National Laboratory, (1957) to be published in Nuclear Science and Engineering.

[4] Schiavone, P. R., Kennedy, W. R., White, T. N., "Surface Dose Rate from Massive Plutonium," Los Alamos Scientific Laboratory, Los Alamos, New Mexico, LA-1574 (1953).

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