

# DISPOSAL OF RADIOACTIVE WASTES

Vol. 1

CONFERENCE PROCEEDINGS,  
MONACO,  
16 - 21 NOVEMBER 1959

# DISPOSAL OF RADIOACTIVE WASTES

## I

PROCEEDINGS OF THE  
SCIENTIFIC CONFERENCE ON THE DISPOSAL OF RADIOACTIVE WASTES  
SPONSORED BY  
THE INTERNATIONAL ATOMIC ENERGY AGENCY AND  
THE UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL  
ORGANIZATION,  
WITH THE CO-OPERATION OF  
THE FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED NATIONS,  
AND HELD AT THE OCEANOGRAPHIC MUSEUM OF THE PRINCIPALITY  
OF MONACO,  
16—21 NOVEMBER 1959

INTERNATIONAL ATOMIC ENERGY AGENCY  
KÄRNTNER RING, VIENNA I, AUSTRIA  
1960

DISPOSAL OF RADIOACTIVE WASTES, IAEA, VIENNA, 1960  
STI/PUB/18  
Printed in Austria

## FOREWORD

Almost every human activity creates some kind of waste. Whether it is harmful, inconvenient, neutral or even positively useful in some other activity depends largely on its nature, which can often be changed by some fairly simple chemical process so as to neutralize harmful wastes, render inconvenient wastes useful, and so on. Radioactive "waste" can be extremely harmful or useful, again depending on its form and the way it is handled; but its essential nature cannot be changed or destroyed by any means at present under the control of man. Furthermore, the harmful waste of today may well become the useful raw material of tomorrow.

As more and more countries embark on programs of nuclear research and nuclear power, the quantities of radioactive material to be disposed of are rapidly increasing and the problems of safeguarding humanity on the one hand and of storing possibly useful material on the other are assuming great importance.

It was for these reasons that the International Atomic Energy Agency and the United Nations Educational, Scientific and Cultural Organization combined their forces in sponsoring and organizing, with the co-operation of the Food and Agricultural Organization of the United Nations, a large scientific conference devoted to the subject of the disposal of radioactive wastes.

The Conference was held from 16 to 21 November 1959 at the Oceanographic Museum in Monaco, in deference to the leading position of this institution in the field of oceanography, which is an extremely important discipline in relation to the disposal of wastes into the sea. A total of 283 scientists attended, representing 31 countries and 11 international organizations.

The sponsoring organizations wish to record their sincere gratitude for the generous hospitality accorded the Conference and all its participants and staff by H.S.H. Prince Rainier III, his Government and the People of Monaco.

It is with the consciousness of offering scientific information of great value to the future progress of an extremely important field of knowledge that I now commend these Proceedings to the earnest attention of all workers in that field.



Director General  
International Atomic Energy Agency

# CONTENTS OF VOLUME I

## A. NATURE OF RADIOACTIVE WASTES

The origin and nature of radioactive wastes in the United States Atomic Energy Programme . . . . .	3
F. R. Bruce (United States of America)	
Management of waste from the use of radioisotopes . . . . .	51
L. R. Rogers (United States of America)	
Origin, nature, disposal and control of radioactive wastes arising from the use of radioisotopes . . . . .	63
A. W. Kenny (United Kingdom)	
Problems to be expected in disposing of fission products from a possible nuclear power programme . . . . .	73
G. K. Dickson (United Kingdom)	
Quantité et activité des produits de fission obtenus en France dans les années à venir compte tenu du développement de l'énergie atomique . . . . .	89
J. Guirlet et J. M. Lavie (France)	
Forecasting the quantity and activity of fission products in France in future years in the light of atomic energy development (Translation of foregoing paper) . . .	10
J. Guirlet and J. M. Lavie (France)	

## B. TREATMENT AND PROCESSING OF RADIOACTIVE WASTES

Disposal into the atmosphere, temporary storage, handling and transportation of wastes

Disposal of waste into the atmosphere . . . . .	111
H. F. Schulte (United States of America)	
On the waste disposal potentiality of the atmosphere . . . . .	125
S. H. Small and P. B. Storebø (Norway)	
Economic aspects of air and gas cleaning for nuclear energy processes . . . . .	139
L. Silverman (United States of America)	
Etude de la fixation et de la migration de cations radioactifs dans un échangeur d'ions naturel . . . . .	181
L. Baetslé (Belgique)	
Study of the fixation and migration of radioactive cations in a natural ion-exchanger (Translation of foregoing paper) . . . . .	200
L. Baetslé (Belgium)	
Temperature distribution in radioactive solid wastes. Part I. Beta-active solids . .	213
D. A. Kotewale and A. K. Ganguly (India)	
Conditionnement des déchets radioactifs solides . . . . .	225
P. Cerré (France)	
Processing and pre-treatment of solid radioactive waste (Translation of foregoing paper) . . . . .	232
P. Cerré (France)	
Une solution de stockage dans le sol des boues radioactives de Marcoule . . . . .	235
P. Cohen et C. Gailledreau (France)	
A solution for the storage of radioactive sludge in the ground at Marcoule (Translation of foregoing paper) . . . . .	251
P. Cohen and C. Gailledreau (France)	
Stockage et évacuation des déchets radioactifs solides . . . . .	264
J. Pomarola (France)	
Storage and disposal of solid radioactive waste (Translation of foregoing paper) . .	274
J. Pomarola (France)	
Problèmes de manipulation et de transport . . . . .	284
J. Pomarola et J. Savouyaud (France)	
Handling and transport problems (Translation of foregoing paper) . . . . .	290
J. Pomarola and J. Savouyaud (France)	

## C. TREATMENT AND PROCESSING OF RADIOACTIVE WASTES

### Treatment of liquid effluent

Elimination par dilution d'effluents liquides faiblement radioactifs . . . . .	304
P. Bovard et C. Candillon (France)	
Disposal of low-activity liquid effluents by dilution: their effects (Translation of foregoing paper) . . . . .	319
P. Bovard and C. Candillon (France)	
Traitement des effluents radioactifs au C.E.N. de Saclay . . . . .	333
G. Wormser (France)	
Treatment of radioactive effluents at the Saclay Nuclear Research Centre (Translation of foregoing paper) . . . . .	345
G. Wormser (France)	
The decontamination of low-level radioactive waste water at Research Establishment Risö . . . . .	351
I. Larsen (Denmark)	
Facilities for waste disposal at Chalk River, Canada . . . . .	361
C. A. Mawson and A. E. Russell (Canada)	
The permanent disposal of highly radioactive wastes by incorporation into glass . . . . .	373
L. C. Watson, A. M. Aikin and A. R. Bancroft (Canada)	

## D. PRESENT METHODS OF WASTE DISPOSAL

The practice of waste disposal in the United Kingdom Atomic Energy Authority . . . . .	403
H. J. Dunster and L. F. U. Wix (United Kingdom)	
Radioactive waste control at the United Kingdom Atomic Energy Research Establishment, Harwell . . . . .	411
R. H. Burns (United Kingdom)	
Некоторые вопросы локализации радиоактивных изотопов в связи с проблемой их безопасного захоронения . . . . .	431
П. В. Зимаков и В. В. Куличенко (СССР)	
Some questions on the fixation of radioisotopes in connexion with the problem of their safe burial (Translation of foregoing paper) . . . . .	441
P. V. Zimakov and V. V. Kulichenko (USSR)	
Treatment and processing of radioactive wastes . . . . .	449
W. A. Rodger (United States of America)	
Treatment, processing and future disposal of radioactive wastes at the Idaho Chemical Processing Plant . . . . .	483
J. I. Stevens (United States of America)	
Radioactive waste facilities at the Australian Atomic Energy Commission Research Establishment . . . . .	509
C. L. W. Berglin, L. H. Keher, G. L. Miles and A. R. W. Wilson (Australia)	
Treatment of radioactive waste at Japan's Atomic Energy Research Institute . . . . .	525
Y. Yamamoto, M. F. Ito, T. Ishihara, N. Mitsuishi and S. Sakata (Japan)	

## E. ADMINISTRATIVE AND GENERAL CONSIDERATIONS IN WASTE DISPOSAL

The public health problems of nuclear waste disposal . . . . .	543
H. J. Dunster (United Kingdom)	
Technical and administrative considerations in the management of radioactive wastes . . . . .	551
A. Wolman and J. A. Lieberman (United States of America)	
Legal and administrative problems of controlling the disposal of nuclear wastes in the sea . . . . .	563
L. M. Hydeman and W. H. Berman (United States of America)	
The hydrographical features of the Baltic Sea and the disposal of radioactive wastes . . . . .	573
I. Hela (Finland)	
Some international legal aspects of the enclosed seas, especially the Baltic Sea, with regard to their protection against pollutive agents . . . . .	589
E. J. Manner (Finland)	

A  
NATURE OF RADIOACTIVE WASTES





# THE ORIGIN AND NATURE OF RADIOACTIVE WASTES IN THE UNITED STATES ATOMIC ENERGY PROGRAMME

## Abstract

The composition of nuclear power wastes is determined, firstly, by the treatment employed for fuel reprocessing, and secondly, by the initial fuel composition. In solvent-extraction processing, wastes of high, intermediate and low activity are produced in approximate volumetric ratios of one, five and 100, respectively.

The wastes resulting from reprocessing of highly enriched fuels are large in volume because they contain the fuel diluent. Low-enrichment fuels usually consist of a uranium core clad with aluminium, zirconium or stainless steel, and they produce two waste streams: one, an intermediate-activity waste containing about 0.1% of the fission products, results from the chemical dissolution; the other is a high-activity waste containing the remainder of the fission products in a dilute nitric acid solution.

The volume of evaporated waste varies from 0.8 l/kg of uranium for low-enrichment fuels to 500 l/kg of uranium for highly-enriched fuels. The average volume of combined high- and intermediate-level waste from the anticipated United States nuclear power industry is about five l/kg of uranium. In the United States it is estimated that the nuclear power industry will have produced about 3,000 million curies of radioactivity in 27 million litres of solution by 1970, and 60,000 million curies in 1,100 million litres of solution by 2000.

Power reactor wastes present new storage and disposal problems because of their composition.

## ORIGINE ET NATURE DES DECHETS RADIOACTIFS DE L'EXECUTION DES PROGRAMMES D'ENERGIE ATOMIQUE AUX ETATS-UNIS

### Résumé

La composition des déchets nucléaires est déterminée, d'abord, par le procédé employé pour le traitement du combustible et, ensuite, par la composition du combustible lui-même. Le procédé de l'extraction par solvants produit des déchets ayant une activité élevée, moyenne ou faible dans des proportions volumétriques d'environ 1,5 et 100 respectivement.

Les déchets provenant du traitement des combustibles fortement enrichis, représentent un volume important parce qu'ils contiennent le diluant du combustible. Les cartouches de combustible faiblement enrichi consistent ordinairement en un élément d'uranium gainé d'aluminium, de zirconium ou d'acier inoxydable et donnent lieu à deux sortes de déchets. Les uns sont des déchets à activité moyenne contenant environ 0,1% de produits de fission et proviennent de la dissolution chimique. Les autres sont des déchets à activité élevée contenant tous les autres produits de fission dans une solution d'acide nitrique étendue.

Le volume des déchets condensés varie de 0,8 litre par kilogramme d'uranium pour les combustibles peu enrichis à 500 litres par kilogramme d'uranium pour les combustibles fortement enrichis, le volume de l'ensemble des déchets d'intensité élevée et intermédiaire qui, à ce que l'on prévoit, proviendront des centrales nucléaires aux Etats-Unis est d'environ 5 litres par kilogramme d'uranium en moyenne. Aux Etats-Unis, on estime qu'en 1970, les centrales

nucléaires auront produit environ 3 milliards de curies contenues dans 27 millions de litres de solution, et en l'an 2000, 60 milliards de curies contenues dans 1,1 milliard de litres de solution.

En raison de leur composition, les déchets provenant des réacteurs de puissance posent des problèmes nouveaux d'entreposage et d'élimination.

## ПРОИСХОЖДЕНИЕ И ХАРАКТЕР РАДИОАКТИВНЫХ ОТХОДОВ В АЭК ПРОГРАММЕ

### Резюме

Состав отходов, образуемых при эксплуатации реакторов, зависит, во-первых, от обработки, которая применяется при воспроизводстве топливных элементов, и, во-вторых, от первоначального состава топливных элементов. При обработки с применением экстракции водных растворов высокая, средняя и низкая активность отходов возникает в приблизительном объемном соотношении, равном одному, пяти и 100 соответственно.

Отходы, возникающие в результате воспроизводства высокообогащенных топливных элементов, бывают большими по объему, так как они содержат топливный разбавитель. Низкообогащенные топливные элементы обычно состоят из урановой сердцевины, покрытой алюминием, цирконием или нержавеющей сталью; эти элементы дают два вида отходов. Первые отходы средней степени активности, содержащие около 0,1% продуктов распада, возникают в результате химического растворения. Второй вид отходов с высокой степенью активности, содержит остатки продуктов распада в разведенном растворе азотной кислоты.

Объем испаряющихся отходов колеблется от 0,8 литров на килограмм урана для низкообогащенных топливных элементов до 500 литров на килограмм урана для высокообогащенных топливных элементов, средний объем соединений вместе отходов высокой и средней степени активности от ядерной энергетической промышленности Соединенных Штатов составляет около пяти литров на килограмм урана. По существующим оценкам ядерная энергетическая промышленность в Соединенных Штатах будет производить около 3 млрд. кюри радиоактивности в 27 млн. литрах раствора к 1970 году и 60 млрд. кюри в 1,1 млрд. литрах раствора к 2000 году.

## ORIGEN Y NATURALEZA DE LOS DESECHOS RADIACTIVOS EN EL PROGRAMA DE ENERGIA ATOMICA DE LOS ESTADOS UNIDOS

### Resumen

La composición de los desechos que se forman como consecuencia de la producción de energía nucleoelectrica depende en primer lugar del tratamiento empleado en la regeneración del combustible y, en segundo, de la composición inicial de éste. Por cada unidad de volumen de desechos de actividad elevada, obtenidos en la extracción mediante disolventes, se forman 5 y 100 volúmenes, respectivamente, de desechos de actividad media y baja.

Los desechos que se producen en la regeneración de los combustibles muy enriquecidos son muy voluminosos por contener el diluyente del combustible. Los combustibles poco enriquecidos consisten habitualmente en un núcleo

de uranio revestido de aluminio, zirconio o acero inoxidable, y producen dos clases de desechos: desechos de actividad media, que resultan de la disolución química, y contienen el 0,1 por ciento, aproximadamente, del total de los productos de fisión, y desechos de actividad elevada, que contienen el resto de los productos de fisión en una solución diluida de ácido nítrico.

El volumen de los desechos evaporados varía entre 0,8 l por kilogramo de uranio, para los combustibles poco enriquecidos y 500 l por kilogramo de uranio, para los muy enriquecidos, se prevé que la industria nucleoelectrica de los Estados Unidos producirá, como término medio unos 5 l de desechos de actividad elevada y media por kilogramo de uranio utilizado. En los Estados Unidos se calcula que para 1970 esta industria habrá producido unos  $3 \times 10^9$  curies disueltos en un volumen de  $2,7 \times 10^7$  l y  $6 \times 10^{10}$  curies en  $1,1 \times 10^9$  l de solución para el año 2000.

Los desechos procedentes de los reactores generadores plantean, por razón de su composición, nuevos problemas de almacenamiento y evacuación.

## THE ORIGIN AND NATURE OF RADIOACTIVE WASTES IN THE UNITED STATES ATOMIC ENERGY PROGRAMME

F. R. BRUCE

OAK RIDGE NATIONAL LABORATORY, OAK RIDGE, TENN.  
UNITED STATES OF AMERICA

### 1.0 Introduction

Man's efforts to achieve the benefits offered by nuclear energy are beset throughout by the hazards of the materials with which he must work. Uranium ores emit toxic radon gas, which makes necessary special provisions for ventilation before ores may be safely mined. The winning of uranium from ores frees radium and other radioactive elements, which contaminate water supplies if released to the environment. The preparation of reactor fuel gives rise to wastes that require special handling, while the obtaining of energy from fissionable atoms in nuclear reactors results in radioactivity which remains a hazard to man for thousands of years. This radioactivity stems both from the capturing of neutrons by structural materials in the reactor and from the fission products formed in the fission process. Successful utilization of nuclear energy requires the recovery of unspent fissionable and fertile material from used reactor fuels. The quantity of radioactive waste originating from this source is at least a thousand times greater than that arising from all other sources combined. In irradiated-fuel recovery processes, fission products are converted to aqueous solutions which must be handled in such a way as to safeguard man and his environment.

In approaching the problem of radioactive waste treatment and disposal, it is most important to have knowledge of the origin and nature of radioactive wastes. It is the purpose of this paper to provide this background by presenting available information on the sources from which radioactive wastes arise; their quantities, their compositions, and properties that are pertinent in considering disposal methods. The methods described for reprocessing uranium and aluminium-uranium alloys clad with aluminium are well established. However, there is great uncertainty in defining the nature of radioactive wastes from power-reactor fuel reprocessing since it is not yet possible to say which

processing methods will be used. For this reason several approaches to the problem and the wastes associated with each are described here.

## 2.0 Raw materials processing

The problem of radioactive wastes is encountered in the very first operations that are carried out in the establishment of a nuclear power enterprise. Uranium ores contain members of the  $U^{238}$  and  $U^{235}$  radioactive decay series, a total of 26 radioactive isotopes. The most important hazards are  $Ra^{226}$ ,  $Rn^{222}$  and uranium itself (Table 2.1). In the case of the United States uranium milling industry, which processes about 65,000 lb of uranium per day, about 9.8 c of  $U^{238}$  and a total of about 100 c of radioactive elements per day are delivered to the mills (1).

### 2.1 Uranium Mining

In the past, about 50% of all miners in the Schneeberg region of Germany and the Joachimsthal region of Czechoslovakia died of carcinoma of the lung (2). Radon, which is given off at a rate between  $5 \times 10^{-7}$  and  $2 \times 10^{-5}$  c/min per thousand cu ft of mine volume (3), is probably the principal cause of this. The average radon concentration in European mines is about  $2.9 \times 10^{-9}$  c/l of air, and may have been as high as  $2.5 \times 10^{-8}$  c/l for at least ten years before the hazard was recognized. The 1955 Recommendation of the International Committee on Radiological Protection is a limit of  $10^{-10}$  c/l for continuous exposure to radon in equilibrium with its short-lived daughters. Radiation exposure during the mining of uranium is minimized by ventilation, and it is recommended that about 3 cu.m of air per minute per ton of ore per day be provided.

### 2.2 Uranium Milling

Uranium ores, containing an average of 0.25%  $U_3O_8$  in the United States, are shipped to mills where uranium is separated from most of the impurities. The ore is crushed and the uranium is leached from the gangue. Two leaching methods are in common use, one using sulphuric acid, and the other a mixture of sodium carbonate and sodium bicarbonate. In acid-leach processes the uranium is recovered from the leach liquor either by anion exchange or by solvent extraction with a dialkyl phosphate or an amine (4). The uranium is removed from the ion-exchange resin or organic solvent with a suitable aqueous solution and is precipitated with caustic, lime, or magnesium oxide. Uranium is

TABLE 2.1  
RADIOACTIVITY\* ASSOCIATED WITH NATURAL URANIUM (1)

Isotope	Half-life	Radiation	Permissible Concentration in Unrestricted Area, $\mu\text{c/ml}$ of solution
U, natural		Alpha	$7 \times 10^{-6}$
$Th^{234}$	24.5 days	Beta	$3 \times 10^{-1}$
$Th^{230}$	$8.0 \times 10^4$ years	Alpha	Undefined
$Ra^{226}$	$1.6 \times 10^3$ years	Alpha	$4 \times 10^{-9}$
$Rn^{222}$	3.825 days	Alpha	$2 \times 10^{-7}$
$Po^{210}$	138.4 days	Alpha	$3 \times 10^{-6}$

\* This table gives only the most significant of the 26 radioisotopes in the  $U^{238}$  and  $U^{235}$  decay series.

TABLE 2.2  
ANALYSES OF MILL EFFLUENTS (5)

Process	pH	U <sub>3</sub> O <sub>8</sub> , g/l	Mill Effluent Analyses, µc/ml		
			Ra <sup>226</sup>	Th <sup>230</sup>	Th <sup>234</sup> +Pa <sup>234</sup>
Acid leach-solvent extraction	1.5	0.10	$4.0 \times 10^{-6}$	$3.9 \times 10^{-5}$	$1.2 \times 10^{-4}$
Acid leach-RIP	3.3	0.0034	$5.4 \times 10^{-6}$	$1.2 \times 10^{-6}$	$4.8 \times 10^{-6}$
	7.7	0.0003	$3.3 \times 10^{-7}$	$2.8 \times 10^{-8}$	$1.8 \times 10^{-7}$
Combined acid leach-RIP and alkaline leach-precipitation	6.9	0.02	$8.1 \times 10^{-8}$	$9.4 \times 10^{-8}$	$5.1 \times 10^{-6}$
Alkaline leach	9.9	0.009	$2.0 \times 10^{-6}$		
Acid leach-countercurrent decantation-solvent extraction	2.0	0.002	$2.7 \times 10^{-6}$	$1.1 \times 10^{-5}$	

TABLE 2.3  
CALCULATED COMPOSITION OF NEUTRALIZED MILL-EFFLUENT OVERFLOW (6)

Constituent	Acid RIP	Amount, ppm		
		Alkaline Filtration	Acid-Counter current Decantation-D2EHPA* Extraction	Acid Counter current Decantation-Amine Extraction
Cl	—	—	—	—
SO <sub>4</sub>	4,556	3,870	96	—
Mg	430	1.1	3,706	4,569
NO <sub>3</sub>	1,470	0	0	140
Cu	—	2.7	<0.6	4
F	—	—	<5.6	—
B	—	—	—	—
Fe	0.1	0.1	0.1	0.1
Mn	332	0	395	760
Pb	—	—	—	—
As	—	—	—	—
Cr (VI)	0.05	—	<0.6	<1.1
U <sub>3</sub> O <sub>8</sub>	12.5	33.2	17.0	22.0
Na	250	3,610	665	28
Ca	735	56.5	735	735
HCO <sub>3</sub>	0	2,560	0	0
CO <sub>3</sub>	—	—	0	0
Mo	2.5	67	—	5.0
NH <sub>4</sub>	430	0	68	175
Si	—	46	285	275
V <sub>2</sub> O <sub>5</sub>	4.5	650	100	100
Dissolved solids	8,221	10,903	6,122	7,059
Suspended solids	—	—	—	—
SAR**	1.8	131	6.4	0.2
Organic	0	0	283	160
pH	7.0	—	—	7.0

\* Di-2-ethyl-hexyl phosphoric acid.

\*\* In meq/litre,  $[\text{Na}]/\sqrt{([\text{Ca}] + [\text{Mg}])/2}$ .

recovered from alkaline-process leach liquor by precipitation. In both processes, the final uranium product is filtered from the solution and dried for shipment.

In general, about 4 tons of waste per ton of dry ore are obtained from plants using the acid-leach process and about 1 ton of waste per ton of dry ore from plants using the alkaline-leach process. The volume of mill tailings in the United States averages about 865 gal per ton of ore processed. The wastes from the acid-leach process are neutralized with lime to a pH of 7 and, like the wastes from the alkaline-leach process, are sent to settling ponds. A typical radiochemical analysis of mill effluents from several processes is shown in Table 2.2. The calculated composition of wastes from typical processes in use in the United States is shown in Table 2.3.

Most of the radioactivity in the ore, exclusive of uranium, is associated with the finely divided solids, which are sent to waste after the leaching operation. In one study (7) of a mill using an anion-exchange process, the combined liquid and solid wastes contained radium equivalent to about 0.6 mg/ton of original ore. Of this, 99.8% was insoluble and passed to the settling pond with the sands and slimes, while only 0.2% was either in solution or went with the uranium product. Of the radium in solution in the waste, 85% was removed by neutralizing the waste before sending it to the settling pond. In order to minimize environmental contamination, the sands and slimes, which contain the bulk of the radioactivity, must be retained. In some cases, uranium in solution is also an important contaminant in mill wastes, being present in concentrations of 0.5–50 mg/l (8).

### 3.0 Uranium refining

The final product from ore-milling is a solid compound containing approximately 70 % uranium. Uranium of sufficient purity for reactor use is prepared by dissolving the concentrate in nitric acid and further separating the uranium from impurities by tributyl phosphate extraction.

For each ton of uranium processed, approximately 1,000 gal of waste solution are produced. The radioactivity in this waste is due mainly to residual radium, associated with the product from the milling operation, and the decay products of  $U^{238}$ , which are primarily  $T^{234}$  with a half-life of 24.5 days and  $Pa^{234}$  with a half-life of 6.7 hr. Since this waste is low in dissolved salts, it is usually volume-reduced and stored. The large volume of such waste produced in the United States is indicated by the 95,000,000 gal produced by the Feed Materials Production Center alone in 1955.

Uranium refineries also process high-grade uranium ores such as pitchblende. The waste from this processing contains appreciable quantities of radium, which is removed from the waste before discharge by co-precipitation on barium sulphate, followed by filtration of solids. The radium-containing solids are stored (9).

Uranium itself is one of the most important contaminants in wastes from uranium-refining operations. In general, wastes containing uranium in concentrations in excess of those which may be discharged to the environment are reprocessed for economic reasons.

Solvent-extraction purification yields a pure solution of uranyl nitrate. This is thermally decomposed to uranium trioxide, which is next reduced with hydrogen and treated with hydrogen fluoride to produce uranium tetrafluoride. Uranium tetrafluoride is reduced to uranium metal with magnesium

or calcium (10). The resulting uranium metal is remelted for additional purification and rolled into forms suitable for fuel element manufacture. In all these operations gases slightly contaminated with particulate matter, especially uranium, are obtained. Gaseous waste may be discharged after purification by cyclone separators and filtration.

#### 4.0 Fuel fabrication

Fuel fabrication consists in cladding uranium metal, or uranium oxide, with a material that is highly resistant to corrosion under reactor conditions, alloying the uranium with other metals, cladding the resulting alloy, and fabricating the uranium, or its alloys, into fuel elements. Wastes from fuel manufacturing operations (11) may be either liquid or solid.

Liquid wastes result from the cleaning of various metals. In 1957, a typical fuel fabrication plant produced 5,000 gal of dilute nitric acid-hydrofluoric acid waste containing a small amount of uranium. The contamination level of such waste is sufficiently low that it may be discarded to the environment after dilution with clean solution.

Solid wastes from fuel element fabrication consist of sheaths used in hot-metal-working operations, asbestos gloves, refractory crucibles, stirring rods, etc. In a typical year, 7,000 lb of copper and steel sheathing, contaminated with about 3 ppm of uranium, were discarded after being cleaned with acids. An additional 5,000 lb of solid waste contaminated with less than 200 g of uranium were disposed of in drums. Fifty drums, 55 gal each, containing combustible material such as paper, were burned and the uranium reclaimed.

#### 5.0 Nuclear reactor operation

##### 5.1 *Fission and Radioactivity*

Heat for the production of power and neutrons for research purposes are derived from the nuclear fission process, in which fissionable atoms— $U^{235}$ ,  $U^{233}$ , and  $Pu^{239}$ —split into two lighter nuclei and yield approximately 2.5 neutrons. The total masses of the new nuclei are less than the mass of the parent nucleus, the difference being released as 200 MeV of energy. For each watt of reactor power approximately  $3.2 \times 10^{10}$  fissions per second occur. Isotopes of 37 elements, ranging from zinc to dysprosium in the periodic table, have been experimentally detected in fission product mixtures. They occur in yields from  $\sim 10^{-7}\%$  to about 7% (Fig. 1) and include isotopes of the rare gases krypton and xenon.

The fission of 1 g of fissionable material per day gives rise to approximately 1 MW-day of heat and 1 g of fission products. Accordingly, if a reactor with a fuel loading of 100 tons of uranium operates at a power of 100 MW for 10 days, about 1,000 g of fission products will be produced. Each ton of fuel—and the waste resulting from its processing—will then contain about 10 g of fission products.

Most fission products are unstable and decay, by beta emission, to the element having the next higher atomic number. The half-lives of the fission products range from less than a second to thousands of millions of years. On an average, radioactive fission products decay to stable nuclides through three stages of decay, and after a short time, there are about 250 radioactive species in a fission mixture. The most important fission products encountered in waste treatment and disposal operations are shown in Table 5.1.



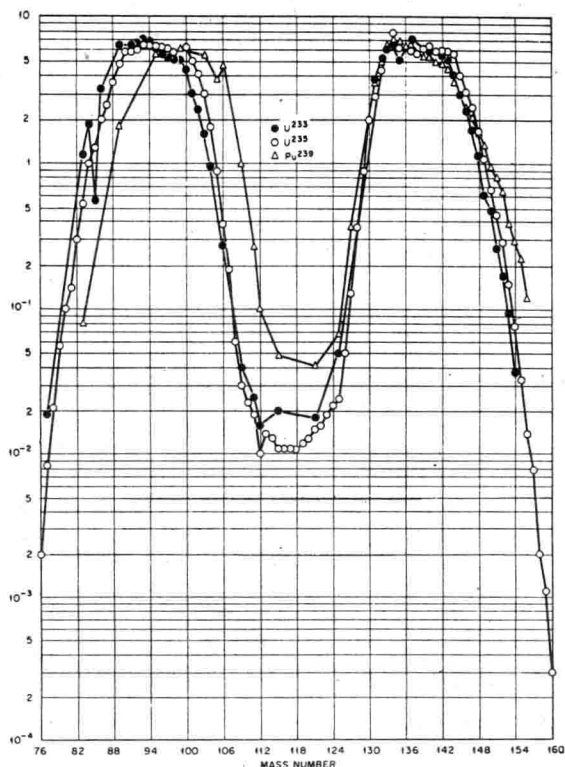


Fig. 1  
Fission yield curve

Although the quantity of a given fission product present in spent fuel waste solution may be calculated (13), it is usually more convenient to determine their quantities graphically (14) from previously calculated machine results. Amounts of fission products are usually expressed in curies rather than mass units, 1 curie being that amount which gives  $3.7 \times 10^{10}$  disintegrations per second. Similarly, 1 mc of an element is the quantity which gives  $3.7 \times 10^7$  disintegrations per second and  $1 \mu\text{c}$  is that quantity giving  $3.7 \times 10^4$  disintegrations per second. Table 5.2 indicates the quantity of the fission products present in 100 gal of waste resulting from the reprocessing of 1 ton of uranium irradiated to 2,500 MW-days per ton and decayed 90 days.

## 5.2 Fission-Product Decay Heat

The heat released from fission-product mixtures as a result of the emission of beta-particles and gamma-rays must be removed from stored wastes. A useful expression for estimating the rate of heat release from a fission-product mixture is

$$H = 5.9 \times 10^{-3} P [(\tau - T_0)^{-0.2} - \tau^{-0.2}] \quad (1)$$

where  $H$  is the rate of beta and gamma-energy emission, in watts,  $\tau$  days after reactor start-up,  $P$  is the reactor power in watts, and  $T_0$  is the reactor operating time in days. The fission product activity,  $C$ , in curies is

$$C = 1.4 P [(\tau - T_0)^{-0.2} - \tau^{-0.2}] \quad (2)$$



TABLE 5.1  
IMPORTANT FISSION PRODUCTS

Nuclide	Half-life	$\beta$ Energy, MeV	$\gamma$ Energy, MeV
Kr <sup>85</sup>	10.27 years	0.695 (99 + %)	none
Sr <sup>89</sup>	54 days	1.463 (100 %)	none
Sr <sup>90</sup>	28 years	0.61 (100 %)	none
Y <sup>90</sup>	64.5 hours	2.18 (100 %)	1.4 (0.4 %)
Y <sup>91</sup>	58 days	1.537 (~100 %)	1.22 (0.3 %)
Zr <sup>95</sup>	63 days	0.364 (57 %), 0.396 (42 %), 0.883 (1 %)	0.717 (98 %)
Nb <sup>95</sup>	35 days	0.160 (100 %)	0.745 (100 %)
Mo <sup>99</sup>	67 hours	0.45 (13 %), 1.23 (87 %)	0.780, 0.740, 0.181, 0.140
Ru <sup>103</sup>	41 days	0.217 (95 %), 0.698 (5 %)	0.498 (95 %)
Ru <sup>106</sup>	1.0 year	0.0392 (100 %)	none
Rh <sup>106</sup>	30 sec	2.44 (12 %), 3.1 (11 %), 3.53 (68 %)	2.41 (0.25 %), 1.55 (0.5 %), 1.045 (2 %)
Te <sup>127m</sup>	90 days	none	0.0885 (100 %)
Te <sup>127</sup>	9.3 hours	0.7 (100 %)	none
Te <sup>129m</sup>	33 days	none	0.106 (100 %)
Te <sup>129</sup>	72 min	1.8 (100 %)	0.8 (100 %), 0.3 (100 %)
I <sup>131</sup>	8.05 days	0.250 (2.8 %), 0.335 (9.3 %), 0.608 (87.2 %), 0.815 (0.7 %)	0.722 (3 %), 0.637 (9 %), 0.364 (80 %), 0.284 (5.3 %)
Xe <sup>133</sup>	5.27 days	0.345 (100 %)	0.081 (100 %)
I <sup>135</sup>	6.68 hours	0.5 (35 %), 1.0 (40 %), 1.4 (25 %)	1.8 (50 %), 1.27 (50 %)
Xe <sup>135</sup>	9.13 hours	0.548 (5 %), 0.910 (95 %)	0.6 (4 %), 0.25 (96 %), 0.37 (1 %)
Cs <sup>137</sup>	26.6 years	0.523 (92 %), 1.17 (8 %)	none
Ba <sup>140</sup>	12.8 days	0.480 (40 %), 1.022 (60 %)	0.54 (30 %), 0.30 (10 %), 0.16 (10 %), 0.03 (100 %)
La <sup>140</sup>	40.2 hours	1.32 (70 %), 1.67 (20 %), 2.26 (10 %)	3.0 (1 %), 2.5 (5.4 %), 1.596 (94 %), 0.8151 (29 %)
Ce <sup>141</sup>	32 days	0.442 (67 %), 0.585 (33 %)	0.145 (67 %)
Pr <sup>143</sup>	13.7 days	0.932 (100 %)	none
Nd <sup>143</sup>	stable	none	none
Ce <sup>144</sup>	290 days	0.170 (30 %), 0.300 (70 %)	0.134 (10 %), 0.100 (10 %), 0.0087 (10 %), 0.054 (10 %), 0.0337 (10 %)
Pr <sup>144</sup>	17.5 min	0.8 (3 %), 2.3 (2 %), 2.98 (95 %)	2.185 (1 %), 1.480 (2 %), 0.695 (4 %)
Nd <sup>147</sup>	11.3 days	0.38 (25 %), 0.60 (15 %), 0.83 (10 %)	0.532 (25 %), 0.318 (15 %), 0.092 (60 %)
Pm <sup>147</sup>	2.6 years	0.223 (100 %)	none
Sm <sup>149</sup>	stable	none	none
Sm <sup>151</sup>	93 years	0.076 (100 %)	0.019 (100 %)
Eu <sup>155</sup>	1.7 years	0.152 (84 %), 0.252 (16 %)	0.1368 (—), 0.1309 (—), 0.1045 (—), 0.0858 (—)

or, more simply,  $C = 240 H$ . Equation's (1) and (2) are applicable for values of  $\tau$  greater than 10 sec but less than 100 days. An example of heat generation in wastes from the processing of slightly-enriched uranium irradiated to a level of 10,000 MWd/ton computed as a function of decay time is shown in Fig. 2.

### 5.3 Transuranic Elements

Recycling of power reactor fuel will result in the production of transuranic elements which are hardly less important as hazards in long-decayed waste than are strontium-90 and caesium-137. The most important of these heavy