Disposal of Radioactive Wastes into Fresh Water

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA 1963
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DISPOSAL OF RADIOACTIVE WASTES INTO FRESH WATER
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The main objective of the Agency is "to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world".

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DISPOSAL OF RADIOACTIVE WASTES INTO FRESH WATER

REPORT OF AN AD HOC PANEL OF EXPERTS

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA 1963
FOREWORD

One of the important tasks of the International Atomic Energy Agency is to encourage the development of safe and practical methods for managing radioactive wastes. In carrying out this task the Agency has used the experience and talents of highly qualified experts from many disciplines and from widely distributed parts of the world.

This report is the work of an ad hoc panel of experts convened by the Agency to study the disposal of radioactive wastes into fresh water. It met on three occasions, the final meeting being held in October 1961. In view of the fact that a previous ad hoc panel of experts studied radioactive waste disposal into the sea and reported its findings in February 1960, this report completes the initial phase of the Agency's work relative to the discharge of radioactive wastes into the hydrosphere. It presents the opinions of the panel members and not necessarily those of their Governments or of the International Atomic Energy Agency.

March 1963

SIGVARD EKLUND
Director General
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PART I

I. INTRODUCTION

A. IMPORTANCE OF FRESH WATERS

Fresh water is a basic necessity of man. It is essential for most forms of life and for the maintenance and growth of civilization.

Fresh water has manifold uses which may be broadly classified as domestic, industrial and recreational uses. Domestic use includes drinking, cooking and cleansing; industrial requirements cover applications in many processes such as those in the chemical, photographic, textile and other manufacturing industries, in power production and transport, as well as for uses in agriculture and horticulture. Also included here should be water bodies which support commercial fisheries. Recreational uses involve such activities as swimming, sailing and sport fishing.

The amount of water required for these purposes is already great and is growing rapidly as the world population increases and as its agricultural and industrial needs expand. In many areas of the world fresh water is already in short supply and the demand for it is increasing. One example should suffice to illustrate the heavy use of water in developed countries. At present the average urban use of water for domestic purposes in the United States is greater than 500 l/d per person; its use increases to over 4000 l/d per person if the water required for industrial purposes is included. This already heavy consumption is predicted to double by 1975.

Because of its important role in man’s present and future economy the protection of this natural resource is an important task.

B. CHARACTER OF FRESH WATERS

The world’s supplies of fresh water are maintained predominantly by rain and snow. The continuous cycle of evaporation and pre-
cipitation which supports this has many variations, as illustrated in Fig. 1. Nevertheless, almost all natural moisture, in whatever form - in the atmosphere, on the surface of the earth or underground - forms part of a world-wide circulatory system.

![Principal aspects of the cycle of terrestrial water](image)

Fig. 1

Principal aspects of the cycle of terrestrial water

(KUENEN, P.H., Realms of water, Cleaver-Hume Press, London (1955))

The distinction between surface water and ground water is based merely on their temporary positions relative to the land surface. The two are closely interrelated parts of a larger system of circulating waters. In humid climates, ground waters generally flow into and support the discharge of streams and springs or may flow directly into the sea; in arid climates surface streams generally seep down into the ground and support the flow of ground water. The movement of ground water may be divided into two interrelated zones: the non-saturated zone (also called the vadose zone of aeration) and the saturated zone. The non-saturated zone exists between the land surface and the upper surface of the saturated zone (i.e., the water-table). In this zone pore spaces are filled partly with water and water vapour and partly with air; the movement of water is essentially vertical, usually downward, but under certain conditions may be temporarily upward. The thickness of the non-saturated zone may range from
nearly zero in humid climates where the terrain is flat, to several hundreds of meters in arid or semi-arid climates. In the saturated zone, on the other hand, all pore spaces are completely filled with water and the flow is essentially horizontal except near areas of discharge into streams or wells and near ground-water divides.

In addition to supplying domestic and industrial water, fresh-water bodies also constitute storage, transport and dispersion systems which civilized man has utilized to flush away the wastes of cities and industries. This latter use of water systems has considerable economic importance and is a significant, if not always obvious, factor in the development of industries requiring convenient and economical disposal of wastes.

The protection of the multiple use of bodies of water becomes increasingly difficult as the volumes and complexity of domestic and industrial wastes increase and it cannot be effective in the absence of detailed knowledge of the effects of these wastes.

C. NATURE AND SOURCES OF RADIOACTIVE WASTES

Some of the newest and most complex wastes are produced by the nuclear energy industry. When an atom undergoes fission and releases energy, or when certain atoms are bombarded by neutrons, the resulting materials emit ionizing radiations which can damage living organisms. The radioactivity of these materials decreases only with time and their radioactive properties cannot be altered or destroyed by any physical, chemical or biological treatment. In this respect, radioactive wastes differ from all other types of wastes.

It must be noted, of course, that even under natural conditions most bodies of water contain trace amounts of radioisotopes that are derived primarily from radioactive rocks and minerals with which the water has been in contact and which contribute to the natural or background radiation to which man is universally exposed.

Artificially produced radioactive materials are used in many ways and in many places, but, depending on the degree of containment necessary for safe handling, the wastes which result from their use may be divided into three general categories; high-, intermediate- and low-activity level, as shown diagrammatically by Fig. 2. High-activity waste, representing the preponderance of waste produced by the nuclear energy industry, is not purposely released to the environment and must be immobilized and stored for indefinite
periods. Therefore this type of waste will accordingly not be considered further in this report, except to point out that the processing of such waste usually involves the production of quantities of wastes having much lower levels of radioactivity, in the range of a few to several thousand picocuries per liter, as a kind of by-product. Thus the percentage of the total radioactive waste which is being released to the environment is very small indeed.

Proper management of radioactive waste requires the consideration of two basic features: safety and economy. It is generally agreed that the release of radioactive waste should be restricted as far as possible in order to keep the radiation exposures at a minimum.

The handling and disposal of radioactive waste from a plant requires many difficult administrative and engineering decisions. Absolute decontamination of a plant effluent may not be possible. Suitable decontamination of some higher-activity-level wastes may be attained by the use of existing methods only at a cost so great that, in some cases, the benefits of the plant may be nullified. On the other hand, a minimal purification system which adds little or no
economic burden to the project may not be in the best long-range interest of the public. Between these two extremes lies a rational compromise whereby some small exposure increment is acceptable so that the benefits of the project may be obtained without a substantial penalty in cost.

D. PURPOSE AND SCOPE OF REPORT

The purpose of this report is to present the principles and practices of radioactive waste management which will ensure that the use of fresh-water systems will in no way be jeopardized. Major attention has been devoted to disposal in streams, lakes and subsurface waters. The report does not include specific studies of other environments, such as estuaries, glaciers and icecaps, snow, etc. Much of the information concerning fresh waters applies to estuaries as well, even though it should be recognized that estuarine and freshwater environments differ in many important respects.

The main body of the report is a general treatise on the fate of radioactive wastes disposed into fresh-water systems; radiation exposure estimation and control; site and monitoring requirements; and the legal, administrative and organizational principles of radioactive-waste pollution control at national and international levels. Conclusions and recommendations are presented. Factors of particular significance are treated in Part II in more detail than is possible in the main body of the report. Subjects on which considerable research is currently being undertaken, such as radioecology, are treated more fully than those, such as hydrology, for which a number of excellent textbooks already exist.

II. FATE OF RADIOACTIVE WASTES INTRODUCED INTO FRESH WATER

If a radioactive waste is introduced into a body of water, various natural processes begin to dilute, disperse, deposit, remove, transform and reconcentrate all or part of the radioactive materials involved. An understanding of these processes is required for an accurate estimation of the movement and fate of the radionuclides in a water system and, similarly, for an accurate estimation of the degree of hazard involved.
The important factors are the movement of the water, the dispersal of the dissolved and suspended matter in the water, the adsorption of substances by natural materials in contact with the water and incorporation into the biota.

A. STREAMING, TURBULENCE AND ISOTOPIC DILUTION

When a waste is introduced into flowing water, such as a river, it is dispersed by molecular diffusion, by dispersion resulting from the method of discharge, e.g. a submerged jet, and by turbulent diffusion. In almost all cases turbulent dispersion is by far the most important dilution mechanism. In addition, when a radioisotope is first introduced into a body of water it will be isotopically diluted with the stable form of the same element which is already present in the water in a dissolved state. It will also become isotopically diluted by exchange with the stable form of the element which is not in solution. In fast-flowing turbulent streams dispersion takes place much more rapidly than in turgid streams and in lakes and ground waters dilution can be a slow process.

Dilution must be considered as a function of both time and distance. It is possible, for example, that in a given stream the dilution that obtains a fixed distance downstream from a point of waste injection will be greater when the stream is flowing at a low velocity than when flowing at a high velocity. Generally speaking, dilution as a function of distance from a point of waste injection is more important than dilution as a function of time.

The minimum dilution for different flow conditions can be estimated from a study of velocity profiles downstream from the place of discharge. Useful information can be obtained from models which simulate the actual flow conditions, and from studies (in situ) with fluorescein and other tracers. Dispersion of wastes introduced into a fresh-water system may be reduced by stratification or laminar flow.

Where wastes are discharged into a section of a stream affected by tides some field investigations are usually necessary. If the discharge is fairly continuous, the mean dilution and dispersion characteristics are of primary interest. If intermittent discharge is possible, a knowledge of the stages of the tide which are most favourable for dispersion of the waste is desired. In preparation for coping with accidental releases of large quantities of radioactive materials, the
probable dilution and dispersion patterns at various stages of the tide should be determined.

B. SEDIMENTATION, ION-EXCHANGE AND ADSORPTION

Any solid particles discharged into fresh water or formed later in the water (see section C) will be affected by water movements. Particles which are more dense than the water will tend to settle out, deposition increasing as stream turbulence decreases. The largest and heaviest particles will of course settle out first.

Some fraction of the radionuclides released into fresh-water environments becomes associated with suspended materials and bottom sediments through ion-exchange and adsorption processes. The natural materials responsible for much of the retention capacity of water-borne sediments are the clay minerals and humic substances. The tenacity with which the radionuclides are retained varies according to the chemical properties of the radionuclides, the materials with which they are associated and the nature of the receiving waters. A change in the chemical composition or physical state of a water body may result in the liberation of radioactive materials from suspended particles and sediments.

The removal of radioactive materials from the water by precipitation and sedimentation will reduce exposure which would otherwise result from the use of the water. It has to be remembered, however, that sediments may be redistributed at times of high river flow, or exposed during low-water conditions. In addition, many forms of aquatic life are closely associated with bottom sediments.

As a result of differential adsorption, certain radionuclides are concentrated in sediments much more easily than others.

C. CO-PRECIPITATION AND FLOCCULATION

Radioactive wastes are chemical mixtures. Upon discharge the wastes can enter into chemical reactions with the receiving water. As a result of such reactions, precipitates containing radionuclides may form and settle out. Where receiving waters come into contact with other waters of a different chemical composition, particularly in estuaries and at river junctions, new chemical reactions leading to precipitation or re-solution of already precipitated materials may occur.
The likelihood of precipitation reactions between different kinds of wastes and various receiving waters can, to a large extent, be investigated in the laboratory.

D. UPTAKE BY AQUATIC ORGANISMS

The tissues of plants and animals contain many of the elements present in their external environment and when certain radionuclides are introduced into aquatic environments they may be taken up by the organisms living in that environment. The amount of accumulation will vary over many orders of magnitude depending upon the element involved and various physical, chemical and biological factors. In the case of elements essential to life, such as nitrogen, phosphorus, iodine, potassium, calcium, carbon, magnesium, manganese, iron, copper, zinc, boron and sulphur, they are present in much higher concentrations in the organisms than in the environment and are assimilated by the organisms much more readily than other elements not commonly found in the biota. The radioisotopes of a few uncommon elements, such as caesium and strontium, may also spread out in the biota, however, because they follow metabolic pathways similar to those of essential elements of the same chemical group to which they belong.

The radionuclides will be taken up by the organism through the processes of:

(a) Adsorption (concentration on the exposed surfaces); and
(b) Incorporation (uptake by the organism directly from the water or from food).

The relative importance of these processes will depend upon the kind and quantity of organisms involved, the nature and concentration of the radioisotope available and its physical and chemical state and the presence of other substances dissolved in the water. Particularly important to the total uptake of an element is the presence of other elements of similar metabolic behaviour.

Radioisotopes can be very usefully applied in determining the relative concentration of elements taken up by aquatic organisms. The "concentration factor" [μg/μl of water/μg/ml of water] for any radionuclide cannot exceed the ratio which exists between the concentrations of the stable form of the element in the organism and in the water. Maximum concentration factors for various radionuclides can thus be predicted from conventional quantitative chemical measurements. Such predictions will usually be high, however, because the quantity of the
element available for exchange from the solids of the ecosystem is not taken into account. For short-lived radionuclides accumulated by fish, significant radioactive decay can occur if the introduced nuclide must reach the fish via a long food chain. In addition, several days or weeks may be required for the nuclide to become uniformly mixed with stable atoms of the same element present in the organs of deposition. Seasonal changes can also be of significance since the rate at which elements are replaced in the organisms is dependent upon metabolic rates.

E. SPECIAL FEATURES OF LAKE ENVIRONMENTS

The foregoing considerations apply, in general, to all water environments, whether flowing or still. There are, however, some features peculiar to lake environments which are of importance. During warm seasons heat is radiated into lakes from above and is absorbed into the uppermost layers. These layers are ploughed under by the wind and distributed by turbulent currents within an upper layer which is usually only a few meters deep (Fig. 3(a)). This upper layer (epilimnion) is, through being warmed, lighter than the deeper cold water and does not mix freely with it. Sufficient light in the upper warm layer allows production of organic substances by photosynthesis in plants. Therefore radionuclides are incorporated into aquatic organisms mainly in this layer.

Dead organisms and other suspended organic materials sink out of the upper layer into the zone lying below it (the transition zone or metalimnion) and are here partly decomposed. The oxygen content of this layer is thereby diminished (Fig. 3(a)). Under these conditions, radionuclides which have been bound in the organisms may be released.

Organic substances which do not decompose in the transition zone sink deeper and, according to the depth of the lake, decompose mainly in deeper water layers (hypolimnion) or on the lake bed. The radionuclides contained in the dead organisms are liberated to the water by autolysis and microbial action and may be adsorbed by sediment or become incorporated into other organisms. The decomposition of the organic substances depletes the available oxygen. If the oxygen is completely consumed during stagnation, the solubility and adsorption characteristics of many substances in the sediment may change. This occurs chiefly in fairly shallow lakes or in those burdened with organic wastes.
If the temperature of the upper layer of a lake falls to a temperature below that of the deep layers, then complete mixing can occur by winds or convection (Fig. 3(b)). This smooths out the differences in oxygen-stratification and distributes substances from deep water throughout the lake. Some radionuclides are once more taken up by organisms. Others may be oxidized, flocculated or adsorbed and slowly return to the lake bottom.
The sequence of stagnation and circulation differs in the various climatic zones or areas. For example, there are lakes with nearly permanent circulation in tropical-mountainous and in windy-temperate regions; lakes with permanent stagnation which are under ice during the whole year; lakes with one or more stagnation and circulation periods yearly; and lakes in tropical regions which have no periodicity of circulation pattern. In some lakes circulation may involve only the upper layers because of chemical density differences and lack of wind. Even in the same climatic area, lakes can be quite different, showing strong differences in patterns of circulation and stagnation, especially of the oxygen curve, chemical compositions and flora and fauna.

F. SPECIAL FEATURES OF UNDERGROUND WATERS

Radionuclides disposed of into artificial ponds, pits and trenches may eventually reach other fresh waters. The radionuclides will be principally transported and dispersed by the movement of ground water, whereas the sorptive properties of the earth materials through which the water flows will act as a delaying mechanism. The rate of transport in underground waters is dependent upon the permeability of the soil and rocks and is, in general, considerably slower than in surface waters. Because of the relatively slow rate of movement of wastes through the ground considerable decay can occur between the point of waste injection and the point or points of eventual water use. Cracks and discontinuities in underground structures can affect the overall flow pattern of underground waters. Also, if large volumes of liquid wastes are injected into the ground the original velocity and direction of movement of the ground water may be changed.

Soil materials retain, to varying degrees, the radionuclides contained in liquid wastes and therefore delay their rate of transport. Opportunity for contact between radionuclide ions and adsorbing surfaces is less in limestone, lava flows or in beds of gravel than in sand sediments. Retention of radionuclides by adsorption and ion-exchange can delay the movement of radionuclides through the ground until considerable radioactive decay has occurred. In order to make reasonably accurate predictions of the behaviour of wastes in the ground both the hydrology and the physico-chemistry of the soil-waste system should be investigated. In order to improve the sorptive capacity, in some cases it may be desirable to modify the chemical characteristics of the waste before discharge.
III. RADIATION EXPOSURE FROM THE DISPOSAL OF RADIOACTIVE WASTES INTO FRESH WATER

A. RADIATION-EXPOSURE CONTROL CRITERIA

The release of radioactive materials into water systems means that some radionuclides may find their way into human and other forms of life. The principal environmental pathways appear in Part II Chapter II (See also Fig.4).

In evaluating the hazards likely to result from discharging radioactive wastes into fresh water, the effects on both the human and other forms of life are of first-order importance. Although the concentration factors of certain radionuclides in aquatic organisms can be quite high, the need to keep human exposure below acceptable limits probably precludes the possibility of damage to other organisms. This view is considered plausible because man is one of the most radio-sensitive of all organisms and because the limits which are applicable to human exposure from environmental sources are substantially below exposures which are believed to have any measurable effect.

The primary standard in the assessment of exposure to man (and other organisms) is the dose to the whole organism or to a particular organ, usually referred to as the critical organ. The International Commission for Radiological Protection (ICRP) has given extremely valuable scientific guidance by recommending maximum permissible doses to man resulting from radiation exposure from all sources (except natural background radiation and medical radiation) from which no "unacceptable" effects are likely. These recommendations are used in most, if not all, countries as a working guide, bearing in mind the overall recommendation that exposures should be kept as low as possible.

As presented by the ICRP the recommended maxima apply to the following categories of exposure:

(A) Occupational exposure;

(B) Exposure of special groups:

(a) Adults who work in the vicinity of controlled areas; but who are not themselves employed on work causing exposure to radiation;

(b) Adults who enter controlled areas occasionally in the course of their duties, but are not regarded as radiation workers;
Exposure pathways for radioactive wastes in surface waterways

(c) Members of the public living in the neighbourhood of controlled areas.

(C) Exposure of the population at large.

The exposure of persons whose duties involve the disposal of wastes is considered as part of their occupational exposure, and the normal radiation protection practices of the nuclear energy industry are applied. The exposure of special groups and the exposure of the
populations at large are important in relation to the discharge of radioactive waste to the environment. It should be noted, for example, that the discharge of radioactive wastes to fresh water from any one controlled area may result in exposure of only a small fraction of the population, but this has to be considered as part of the total exposure of the population which may include contributions from several other sources.

B. RADIATION EXPOSURE ESTIMATION

The radioactive wastes discharged to fresh water frequently involve a mixture of radionuclides. The exposure which limits the discharge may be the genetic dose to the population or may be that of individuals or the contribution to the average somatic dose to the population. In the last two cases the critical organ will have to be determined. An assessment of the exposure by qualified experts is required, since there are many factors to be considered and individual circumstances may vary widely.

It is possible to calculate the size of the special group affected by disposal of waste into fresh waters and, relative to the size of the whole population, the size of a group above which the genetic dose is more limiting than the exposure of the individual. This will depend on the apportionment of the genetic dose which is allotted to the special group and upon the variance of individual exposures. A calculation of the permissible size of a special group can be made if the average exposure of the group is known. Part II Chapter III contains relevant examples of dose calculations.

Given sufficient information about the behaviour of waste radionuclides discharged to the environment, it would be possible to calculate with considerable accuracy the concentrations in which they appear in positions hazardous to man and other forms of life. On the basis of this it would then be possible to calculate with a fair degree of accuracy the resulting radiation doses to different organs. This degree of refinement in calculation is not possible at the moment and is unlikely to be possible in the foreseeable future. However, in practical problems of providing scientific guidance concerning waste disposal, it may only be necessary to demonstrate the "acceptability" of the worst conceivable case. For example, if it could be demonstrated that no unacceptable radiation exposure would arise even if all of a particular radionuclide released to a fresh-water environment found
its way into any species of edible fish, then it would not be necessary to conduct a detailed investigation of the accumulation of the radionuclide in fish. This sort of approach can be carried out in stages, using what is known about the environment, the behaviour of radionuclides and dietary habits, etc. to provide refinement of calculation wherever necessary.

While the ICRP has not recommended maximum permissible concentrations of radionuclides in food, the maximum permissible concentrations in water (MPC\textsubscript{w}) given by the ICRP can be used with care and judgement for estimates of radiation dose resulting from the ingestion of foodstuffs. The MPC\textsubscript{w} values tabulated are based on occupational exposure and care must be taken that appropriate factors are used when conditions are different from those of occupational exposure. Exposure at a constant or near constant level over a period of 50 years was assumed by the ICRP and should be taken into account in situations where the discharge of wastes is not constant.

The MPC\textsubscript{w} values of the ICRP, which are based upon intake of 2200 ml/d of water, cannot be used directly for determining the maximum permissible concentrations of radionuclides in foodstuffs. However, an application of the MPC\textsubscript{w} to foodstuffs can be conveniently made as illustrated in the following example. If, for example, 100 g of butter are eaten daily, then the maximum permissible concentration for a particular radionuclide in butter can be obtained by multiplying MPC\textsubscript{w} by 2200/100. (This example assumes that butter is the only contaminated food.) It may be necessary to make the illustrated calculation for each radionuclide of interest.

If dietary habits are known, the rate of ingestion of individual radionuclides can be estimated. The radiation exposure resulting from dietary intake, expressed as a fraction of permissible dose, can be assumed to be the same as the fraction of the MPC\textsubscript{w} which is actually ingested.

C. OTHER CONSIDERATIONS IN EXPOSURE ESTIMATION

In addition to the several uncertainties in exposure estimates there are others which warrant consideration at the present stage of knowledge and technology. These can be categorized broadly as measurement uncertainties, variations amongst individuals and applicability of limits.
The necessity for measuring particular radionuclides in various media and the degree of accuracy needed are functions of the biological importance of individual radionuclides. During the last few years there have been substantial advances in analytical techniques for the assessment of individual radionuclides. These analyses are usually necessary in addition to the analyses for gross beta, gamma and gross alpha-emitters. Because of the use of the measurements in dose estimates it is important to report the results in such a manner that they can be treated statistically.

Variations in the diets of individuals are quite pronounced. It is generally desirable to obtain quantitative knowledge of the dietary habits of persons residing in the vicinity of radioactive-waste disposal sites. Statistical treatment can be used and the habits of individuals (NB of all ages) as well as the averages for special groups and whole population considered. Variation in other habits, such as place of residence, recreation and occupation, can be determined and used in estimating exposures. Consideration should also be given to the possibility of local vegetation or food being consumed at other, perhaps distant, places.

Some of the apparent deviations from the conditions assumed by the ICRP in deriving the MPC\textsubscript{w} values have been discussed previously. One further departure warrants discussion. The special groups (B) (c) and (C) include children, pregnant women, the aged and infirm. The ICRP used "standard man" characteristics in developing MPC\textsubscript{w} values which were intended to represent an average adult occupationally engaged in radiation work. Many of the characteristics included in the "standard man" assumption do not apply for individuals. Some groups of qualified experts have been giving particular attention recently to the determination of limits for children.

If exposure standards based on "standard man" are to be applied to a population as a whole, the age distribution of that population would have to be considered in view of the variations in age structure of populations in different parts of the world.

The information needed for dose evaluation forms the basis for designing the monitoring programme and planning control measures and these have bearing on site evaluation and selection.
IV. WASTE-DISPOSAL SITE CONSIDERATIONS: DETERMINATION OF DISCHARGE PROCEDURES AND MONITORING AND CONTROL REQUIREMENTS

A. ENVIRONMENTAL EVALUATION AS A FACTOR IN SITE SELECTION

As described in the previous sections, the behaviour of radionuclides after discharge depends upon the chemical and physical properties of the wastes and upon many complex and variable processes in the environment. Because of these variables the fate of the radionuclides will differ considerably for discharges into different types of fresh-water systems.

In order to keep the radiation exposure resulting from waste disposal well below acceptable limits, it is essential to consider each individual potential discharge in relation to the uses of the environment throughout the region likely to be affected. A knowledge of the kinds and amounts of radionuclides which already exist in the region is also needed.

For small amounts of wastes, particularly those containing radionuclides with short half-lives, the problems of minimizing exposure at the points of water use are relatively simple. These problems become progressively greater as the amounts and half-lives increase. Therefore the potential consequences of fresh-water contamination both under normal and abnormal operating conditions are a very important consideration when deciding where to locate large nuclear installations such as power reactors and chemical plants for processing irradiated fuel elements.

Initially, when weighing the advantages and disadvantages of several possible sites, the likely effects of controlled and uncontrolled discharges of radioactive materials have to be calculated using readily available information such as hydrological and geological maps, water-flow records and the knowledge of authorities acquainted with local circumstances. Clearly, too, when classing a site the possible release of radioactive material in an accident will have a bearing on the requirements of the fresh-water system which is to be used.

B. ACTION BEFORE DISCHARGE

Once a site has been chosen, however, estimates must be made of the possible concentrations of radionuclides at the important points
of water usage (i.e. drinking and consumption of products influenced by the water) and the resulting radiation exposure during both normal and emergency situations. To do this, the factors which are likely to limit the amount and rate of discharge of particular radionuclides must be identified. (Although this report is concerned solely with hazards due to the radioactive characteristics of discharged wastes, it is also important to consider the possibility that radioactive-waste disposal might be limited by the chemical toxicity of the materials discharged.)

These estimates can then be applied to:
(a) Plan the waste treatment necessary;
(b) Establish control procedures; and
(c) Determine the need for, and extent of, monitoring.

The accuracy of predicting human exposure will, in general, be limited by lack of knowledge of release rates and hydrological, limnological and other environmental factors which may contribute to the exposure to man through food chains or other mechanisms, and a number of assumptions must be made.

In many instances, assumptions will have to be based on the results of experimental investigations if they are to be sufficiently reliable. Fundamental research may be needed to establish the validity of various hypotheses underlying the schemes of calculation. These in turn may need to be verified on a step-by-step basis by experiments under field conditions approximating those expected to be met in practice. Extensive environmental and ecological studies may be necessary in order to identify all factors of interest. Statistical and economic studies may be necessary to establish the pattern of man's use of the relevant aquatic resources. Experimental releases of simulants, or of actual wastes, on a limited scale, must be followed to verify the overall pattern of dispersal when such a dispersal mechanism is an essential factor in determining the safe limit.

Experience at a number of large nuclear installations has shown that the observed environmental concentrations may be somewhat different from predicted values. Therefore, disposals must be started very cautiously by releasing only small amounts of radionuclides, and measuring the concentrations of radionuclides in different environmental samples. The measured values can then be used to improve the accuracy of the calculations. In order to do this effectively the background concentrations of relevant radionuclides in the environment should be determined before disposal. The extent and nature of these background measurements will depend upon the radio-
nuclides which are likely to prove limiting and the size of the region likely to be affected during both normal and abnormal conditions.

It is advisable to take into account the level of the existing radioactive materials in a water system. If the level of activity of these materials is at the limit calculated according to one of the previously mentioned means for the determination of exposure of human beings, some special requirements in relation to the content of the effluent will be necessary.

The discharge of radionuclides into a water system in which these radionuclides, or chemically similar radionuclides, are already present to a significant degree, should be discouraged. It must be borne in mind that release of radionuclides at concentrations lower than those already existing in the receiving water will not be objectionable since the release actually causes some dilution.

Where the receiving water can be used in the near vicinity of the plant outfall for a purpose which contributes significantly to human exposure, the waste must be diluted adequately and rapidly. The method of discharge will be determined by local conditions at the disposal site and investigation may be necessary in order to design for maximum dispersion throughout the width and depth of the water system. Should there be any uncertainty as to whether the effluent will be sufficiently diluted, the concentration of the radionuclides should not exceed that specified by the ICRP as the maximum permissible for drinking water for occupational workers. It should be borne in mind, however, that, in any case, special restrictions on the rate of release of some radionuclides will usually be required to limit the dose to the population from all exposure pathways. These restrictions may differ at different seasons of the year or at different volumes of river flow.

Control and measurement of radioactive effluents are greatly facilitated by the provision in the system of holding tanks for retention of the fluid before discharge. Such a system is particularly valuable where the quantities of wastes may vary considerably with time.

Holding tanks may not be practical, however, where the water is used in the cooling circuits of reactors; where very large volumes of liquid are produced continuously at constant but low levels of activity; or for small installations, such as hospitals, where the quantity of radioactive material has been assessed as extremely small in relation to the volume of liquid discharged.

The size of a tank system will vary according to circumstances and should be capable of holding any amount of effluent which may
have to be held for measurement purposes before disposal and for
treatment in the case of an off-standard effluent. The amounts of
wastes which might be produced during an emergency situation and
require retention must also be considered.

Following an assessment of radiation exposure which may be
contributed by an individual establishment, the permissible releases
from that establishment can be expressed as a simple formula. Such
a formula allows the operator of the facility a degree of flexibility
in relation to the composition of his effluent. The formula would be
of the form:

\[ x \text{ (curies of the most limiting alpha- or beta-emitting isotope)}
+ y \text{ (curies of the second most limiting alpha- or beta-emitting iso­}
\text{tope)} + \ldots p \text{ (total curies of remaining alpha-emitting isotopes)}
+ q \text{ (total curies of remaining beta-emitting isotopes)} \leq R \text{ (curies}
in a given period).\]

The isotopes selected for a specific assessment in the formula will
depend on the circumstances of the uses of the fresh water and of
the biological conditions connected with these uses. These consider­
ations will indicate which isotopes may be limiting. The values of
the constants x, y, p and q are derived from the same considerations,
together with the appropriate maximum permissible exposure levels.

An example of this type of formula is the one used for discharges
from Harwell into the Thames:

\[ 2500 \text{ (radium)} + 420 \text{ (other alpha-emitters)} + 50 \text{(radiostrontium}
\text{ and radiocalcium)} + \text{other beta-emitters} \leq 20 \text{ c in one month.}\]

Here (radium) etc. stands for number of curies of the radioactive
material discharged. In this case, use of the water as drinking water
is the most limiting use and there are no concentration processes.
Therefore, the values of the constants are inversely proportional to
their MPC\text{w}.

C. ACTION AFTER DISCHARGE

A system of effluent monitoring based on continuous or frequent
sampling and analysis of the discharged materials is necessary for
controlling the effluent release and for taking proper action in the
event of abnormal circumstances arising. For installations releasing small amounts of radioactive material, the amount of information obtained from such monitoring may be a sufficient basis for forming a judgement about the possibility of any harmful effects.

Because of lack of knowledge of the distribution of radioelements in the environment it is necessary for larger installations to carry out a programme of environmental monitoring. The environmental monitoring will be modified on the basis of the measurements made during the initial discharge period.

If human radiation exposure is the overall limiting factor, concentrations of radionuclides in, for example, selected foodstuffs and drinking water, may have to be determined. The sampling and measuring programme which will be required for this purpose should be determined on the basis of information about food chains and dietary habits, etc. In some cases, the use of water by industry may limit disposal, in which case special monitoring systems may be set up to safeguard, for example, plants producing photographic materials.

For large nuclear installations the possibility of large discharges of radionuclides during abnormal situations may result in a need for special equipment for the detection of accidental releases. In considering the effects of abnormal situations, experience has shown that it is advisable to consider each operation of an installation and not only those where the likelihood of abnormal situations developing seems to be highest. Plans should be made for the rapid expansion of the normal monitoring programme to meet abnormal situations.

D. MEASUREMENT TECHNIQUES

The ability to determine quantities of radionuclides in various media is being constantly improved, but simple determinations of gross beta, gamma and alpha-activities are still extremely useful, provided that these are supported by other more specific measurements. One difficulty inherent in the simple gross beta-analysis is that all activity observed must be assumed to be that of the most limiting isotope, which in most cases is Sr$^{90}$. If the total beta-activity then exceeds the maximum permissible concentration for Sr$^{90}$ a specific analysis for that radionuclide becomes desirable.

If the limiting isotope of a mixture of isotopes is known, it may be sufficient to determine if the gross activity of the entire mixture is lower than the maximum permissible for that particular isotope.
When there are appreciable concentrations of radionuclides, it is usually necessary to determine the quantities of individual radionuclides to see whether or not these are below the individual limits and to estimate doses to man.

Separate determination of individual radionuclides need be used only when detailed knowledge of these radionuclides is required. This can be done by chemical or radiochemical analysis or gamma-ray spectrometry, as appropriate.

Gamma-ray spectrometry, which is a widely used method for identifying and measuring gamma-emitters, has the advantage of requiring little or no sample preparation.

For the measurement of pure beta-emitters such as Sr$^{90}$ and P$^{32}$, techniques for the measurement of beta-emitters must be used. This method can be used also for determining beta-gamma-emitters in a mixture of radionuclides when gamma-ray spectrometry is difficult or impracticable.

Whole-body measurement is now being used to determine in situ the body-burden of gamma and beta-emitting radionuclides. The measurements can be made directly by measuring the radiation which originates from internally deposited radioactive contaminants and penetrates the surrounding tissue. Indirect assessment of body burdens can also be made by measuring the quantity of radionuclides excreted per unit time.

Measurement results may be used to decide acceptable waste release rates, particularly for radionuclides which tend to concentrate in food chains leading to man. Comparison of results with those of similar measurements made on samples collected at a carefully chosen reference site unaffected by waste disposal, can be very useful in determining increases due to waste disposal. Increases which occur in both areas can then be attributed to fall-out or some cause other than waste disposal.

V. LEGAL, ADMINISTRATIVE AND ORGANIZATIONAL PRINCIPLES OF RADIOACTIVE-WASTE POLLUTION CONTROL

Authorities responsible for the abatement of water pollution are in a favourable position in respect to the nuclear energy industry, since no radioactive pollution of fresh-water bodies which requires correction is known to exist. This currently favourable situation,
coupled with the great public interest in the injurious effects of radioactive materials, is furthering pioneer action in the field of pollution control. The prevention of pollution obviously requires a definition of pollution, and such a definition has been approached in several ways.

A. DEFINITION OF POLLUTION BY RADIOACTIVE MATERIAL

In view of the fact that most available evidence does not favour the concept of a threshold level below which the effects of exposure are nil, a definition of pollution by radioactive material must recognize the importance of minimizing exposure of persons and populations. As with other agents which are both beneficial and potentially harmful, society must weigh risk against value. One definition of pollution could be that condition which restricts the use of the water body for any purpose, the restriction being based on an unallowable exposure which would result if the water were used. Another definition could be for an authority to establish maximum permissible exposures with pollution being defined as that amount of radioactive material in the environment which produces above-permissible exposure. This second definition can then be extended to estimating the concentration and amount of material which, if released into a water system, will not result in above-permissible exposures. Such an estimation must consider the many factors described in Chapters II-IV above.

B. LEGISLATIVE TECHNIQUES

Control of waste disposal should be undertaken at a national level through the enactment of a statute dealing specifically with such disposal, or by inclusion of provisions relating to disposal in a statute which deals with the utilization of nuclear power generally or prohibition of pollution of the environment. Such a statute would incorporate by reference detailed technical regulations regarding disposal into fresh water, into the atmosphere, the soil or the sea. Such regulations would also contain one or more annexes with scientific data relevant to the text.

In the case of states with a federal structure, fundamental regulatory norms are embodied in federal legislation while it is left to state authorities, which are in general responsible for control of
waste-disposal activities, to enact state legislation applicable in con­formity with the federal statute. The degree to which a federal authority may enter into state waste-disposal activities which might have international effects should be clearly established.

While a statute would normally refer to radioactive waste in general terms, regulations formulated under it would specify the particular prohibitions relating to the disposal of solid, liquid or gaseous wastes.

Statutes and regulations generally designate the authority competent to regulate matters relating to nuclear energy, including the disposal of radioactive waste. The necessary powers may be invested in a central state administration, such as a ministry of public health, in local authorities or in special autonomous bodies charged with the duty of preventing pollution of the environment.

The use of radioactive materials above exempt levels* should be governed by regulations. Licence conditions can be attached to this to protect the environment. These conditions can contain the following obligations of the holder of the licence:

(a) To supply registration of the amounts of radioactive materials used and discharged with particulars about the concentration and composition of the waste;
(b) To allow inspection of the premises and to give all particulars of importance concerning the waste;
(c) To obtain prior approval of the methods of disposal of radioactive wastes - this may include submission of data on environmental studies;
(d) To meet the requirements about the quantity and composition of the waste discharged as prescribed in the licence;
(e) To provide methods of handling radioactive wastes, approved by the competent authority;
(f) To allow sampling and monitoring of the radioactive wastes.

The competent authority would usually administer regulations relating to disposal and take action to secure abatement of pollution and application of the sanctions prescribed thereunder.

Where local authorities are not principally responsible for the control of waste disposal, provision is generally made for the maintenance of close co-operation in particular cases between the competent authority and such local authorities as would in general be responsible for the public health administration of the area concerned.

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* Many authorities exclude the use of small amounts of radioactive substances from their regulatory procedures.
Regulations regarding the disposal of radioactive wastes should include the following points:

(a) Methods of disposal;
(b) Disposal sites, inspection and monitoring;
(c) Character and quantity of wastes; frequency of disposal;
(d) Sampling;
(e) Records;
(f) Health and safety at disposal facility;
(g) Procedures to be followed in the event of an abnormal situation.

National regulations should consider the question of sanctions and liability insurance.

VI. RADIOACTIVE-POLLUTION CONTROL OF INTERNATIONAL FRESH WATER

A national government disposing of radioactive wastes into fresh waters within its own territory can completely determine the details of such disposal, provided the following conditions are complied with:

(a) The disposal should be carried out in such a manner that detrimental effects will not be felt in marine waters;
(b) Such disposal should not give rise to any deleterious effects outside the borders of that country.

Planning for the disposal of radioactive wastes into international fresh waters can be more difficult than planning for disposal into a purely national water, because the resulting conditions, which are acceptable to one country, may not be acceptable to another. It is a general rule of international law that a state must not abuse its rights under international law by allowing alteration of the natural conditions of its own territory to the disadvantage of the natural conditions of the territory of a neighbouring state.

Expert opinion in support of this viewpoint has been expressed in the following terms: "... in accordance with established principles of customary international law no state should pass on its waters to its neighbouring states in such a polluted condition that this water would seriously damage the interest of its neighbouring states."*

Application of the permissible exposure (as mentioned in chapter V) to international fresh-water pollution control becomes complex.

for the reason previously stated, i.e. what may be permissible for
the country disposing of wastes may not be permissible for the ri-
parian states.

The problem of disposal of radioactive wastes into international
fresh waters could be solved by the countries affected by the dis-
posal in one of the following ways;

(a) The establishment of a bilateral or multilateral convention
based on the principle that the water passing from one country into
another should meet certain specific quality criteria. The task of
implementing the convention could be approached either by the states
themselves directly, or through a commission set up for the purpose.

(b) The establishment of special bodies for particular drainage
basins by bilateral or multilateral conventions possibly consisting of
representatives of all the countries concerned which would meet from
time to time to lay down conditions governing the discharge of wastes
into the international fresh waters by each of these countries. This
method would be different from the one proposed under (a) in the
sense that whereas (a) envisages a static convention, (b) envisages
a dynamic convention which would enable the countries concerned
to consider each fresh proposal for waste disposal in the light of the
conditions already existing and suggest suitable measures for en-
suring radiation safety to the population concerned. The problem of
implementation of the decisions thus reached could be approached in
two ways. In the first, the body or bodies constituted to formulate
the bilateral or multilateral agreements from time to time could be
purely advisory, the actual ratification and implementation of the
formulas being left to the governments concerned. Alternatively,
these bodies could be empowered not only to arrive at mutually
agreed upon disposal criteria, but also to take such control, ad-
ministrative and enforcement measures as may be necessary to
implement the agreement. The creation of such bodies with full execu-
tive powers to make and implement decisions which bind participating
states and their nationals would involve the assignment of some
degree of state authority. It is, however, the opinion of this panel
that in view of the rapidly expanding atomic energy programmes in
various countries of the world, such bodies for particular water
systems would provide a satisfactory solution to the problem of
radioactive-waste disposal into international fresh waters.

Since the chemical balance of a fresh-water system will affect
its capacity to accept radioactive wastes, the terms of reference
of any such bodies must be broadened to include the general problem
of pollution of international fresh waters by the discharge of inorganic and organic material in addition to radioactive wastes.

For international negotiations of the type mentioned above, the recommendations of the ICRP could be used conveniently as a starting point. In addition, general suggestions for the establishment of such international river basin pollution control bodies are to be found in the document "Tentative Suggestions for the Establishment of International River Pollution Control Bodies in Europe"; Annex III of Economic Commission for Europe Document E/ECE/340 (6 March 1959).

VII. CONCLUSIONS AND RECOMMENDATIONS

The fate of radionuclides introduced into fresh water will be influenced not only by the volume of the water available for dilution, but also by a number of other physical, chemical and biological factors, viz:

- Replacement of the water mass;
- Turbulence and location of the currents;
- Chemical composition of effluent and receiving water;
- Suspended materials;
- Thermal characteristics and density differences;
- Precipitation and sedimentation;
- Ion-exchange and adsorption;
- Incorporation by living organisms;
- Special features characteristic of the type of water body involved, i.e. whether stream, lake, estuary or sub-surface.

Existing technology for the management of radioactive waste is adequate to ensure that releases to fresh water can be held to acceptable amounts under normal operating conditions. However, the release of radioactive waste to fresh-water systems should be kept to a minimum so that radiation exposures can be kept to as low a level as possible.

Broad estimates of the concentrations of radioactive materials which may ultimately exist in the environment as a consequence of normal and abnormal operating conditions can be made from a general knowledge of release rates and physical, chemical and biological factors, and are useful in order to:

(a) Determine the suitability of a site under consideration;
(b) Plan the waste treatment necessary;
(c) Establish control procedures; and
(d) Predict the need for, and extent of, monitoring.

Except in special cases, maximum dilution in the minimum time is desirable when radioactive wastes are disposed into fresh waters. Disposals should, therefore, be carried out in large water systems and, whenever possible, under conditions favouring turbulent diffusion.

Since the accuracy with which the resulting concentrations of the radionuclides in the environment may be predicted is limited by available knowledge of their probable behaviour and distribution in the water, biota and sediments, disposal of wastes should be started in a very conservative manner.

Usually the potential exposure to man will limit the release rate of the radioactive waste, but in some cases other circumstances, such as industrial requirements for high-purity water, may be more limiting.

The primary radiation exposure standards of the ICRP should be used as the basis for evaluating the effects of radioactive waste disposals.

Practical standards derived from the maximum permissible concentration tables of the ICRP Sub-committee II should be based on the total intake of radionuclides. Due allowance must be made for the contribution from external radiation.

The size and nature of the population group affected by waste-disposal operations should be determined with reasonable accuracy so that the appropriate exposure standard can be selected.

The control of population exposure should be ensured by the following procedures:

(a) Preliminary assessment of the results of waste disposal using the most accurate relevant data that are available. This may lead to research necessary to establish the validity of assumptions made in the assessment.

(b) In some cases experimental discharges will be required to confirm the results of the above calculations;

(c) Environmental monitoring after discharges begin will be required in all cases where radionuclides will reach man in significant amounts.

The dose which may be contributed from each individual site must be added to that from other sources and multiple pathways of exposure must be evaluated.
Measurement of the radionuclides present in the vicinity of a plant before start-up, or in a remote but similar environment, will provide valuable data for comparison with results subsequent to the disposal of radioactive wastes.

The information obtained from the environmental surveillance programmes can be used as a basis for estimating the dose to persons living in the vicinity of a proposed plant, for identifying the radionuclides which would contribute most significantly to the total dose, for relating dose to the release rate of the effluent and for the establishment of acceptable rates.

Measurement of the concentrations of radionuclides in water, aquatic forms of life and foodstuffs following release by an installation provides the best data from which to estimate the extent of potential exposure from that installation and to relate release rates to maximum permissible limits.

Tanks or other holding facilities must be provided for retention of effluent which can possibly exceed predetermined limits.

Where only small quantities of radioactive materials are discharged, infrequent monitoring of the effluent may be sufficient to demonstrate the impossibility of significant exposure resulting from the discharge.

In many cases it may be practical to consider very low-level wastes as "not significantly radioactive" and thus to dispose of them without special precautions. Until criteria are established, the propriety of such release should be determined by appropriate licensing authorities.

Large-scale release of radioactive wastes requires frequent sampling of the effluent and environmental monitoring. The detection of off-standard effluent should initiate immediate action to stop the release. Provision should be made for the rapid expansion of the environmental monitoring programme to evaluate abnormal situations.

Legal, administrative and organizational measures, both at national and international levels are necessary to ensure that radioactive waste disposal procedures do not result in unacceptable radiation exposure to the public, nor interfere with further use of the waters concerned.

The use of radioactive materials above exempt levels is governed by legislation and similar measures should also apply to the handling and disposal of radioactive waste. The conditions that should govern
procedures relating to the disposal of radioactive waste are given on pages 32 and 33.

The method of administration of these regulations by a competent authority should include periodic measurements to ensure that the regulatory criteria are fully met and that where necessary, adequate control measures are undertaken.

When radioactive waste is discharged into fresh waters within a country, care should be taken to ensure that detrimental effects do not accrue in waters outside the borders of that country or in marine waters.

In view of the rapid expansion of atomic energy programmes throughout the world and in view of the fact that in many cases international waters will be involved in waste-disposal procedures, the interest of all countries concerned must be taken into account. Therefore it is the view of this panel that the procedure outlined in Chapter VI, page 34, (b) would be the best means of effective control over waste-disposal procedures involving international fresh waters.
PART II

I. RADIOACTIVE WASTES - SOURCES, AMOUNTS AND MANAGEMENT PRACTICES [1, 2, 3]

A. SOURCES OF RADIOISOTOPES

Radioisotopes in general originate from one of three principal sources: naturally-occurring uranium-238, uranium-235, uranium-234, thorium-232, and their decay series; fission of uranium-235, uranium-233 or plutonium-239, and neutron activation in nuclear reactors.


Uranium-238, uranium-235, uranium-234, thorium-232 and their decay products are among the radioactive substances occurring in nature which might appear in the wastes from nuclear energy installations. The more significant of these are shown in Table I. The decay products of uranium-235 and uranium-233 are not significant because of the low concentration of the parent elements. The decay daughters of thorium contain isotopes which emit much stronger radiation than the uranium daughters; thorium, therefore, is more hazardous than uranium. In the uranium decay chain the most hazardous isotopes are radium-226, radon-222 and uranium-238.

2. Fission products

When the nucleus of a fissionable atom such as uranium-235, uranium-233 or plutonium-239 captures a thermal neutron, the resulting nucleus in an excited state undergoes fission, releasing an amount of energy equivalent to the mass defect of the reaction. The immediate products of the fission process generally are two lighter elements called "fission fragments", and 2.5 neutrons, on the average, which are emitted instantly by the fission fragments. The mass
numbers of the fission fragments are distributed over a wide range in the periodic table, from zinc-72 to dysprosium-161. The mass distribution of fission fragments resulting from fission or uranium-233, uranium-235 and plutonium-239 is shown in Fig. 5[5]. The Figure shows the "double-peak" mass distribution which is characteristic of low-energy fission. The primary fission fragments are "neutron-rich" and, therefore, are unstable [6]. On the average radioactive fission products decay through three stages of decay and for a short time there are about 200 radioactive species in a fission mixture. The most important fission products encountered in waste treatment and disposal operations are illustrated in Table II [6].
3. Radioisotopes produced by neutron activation

During nuclear reactor operation, materials placed in the field of neutron flux may interact with neutrons in non-fission capture and become radioisotopes. Neutron activation serves some useful purposes such as breeding fissionable material from fertile elements and manufacturing isotopes for application in medicine, industry and research laboratories. On the other hand, undesirable waste
<table>
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<th>Half-life</th>
<th>Nuclide</th>
<th>Half-life</th>
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<tr>
<td>Kr(^{85})</td>
<td>10.27 yr</td>
<td>Cs(^{137})</td>
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<td>Sr(^{89})</td>
<td>54 d</td>
<td>Ba(^{144})</td>
<td>12.8 d</td>
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<td>28 yr</td>
<td>La(^{140})</td>
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<td>Y(^{90})</td>
<td>64.5 h</td>
<td>Ce(^{141})</td>
<td>32 d</td>
</tr>
<tr>
<td>Y(^{91})</td>
<td>58 d</td>
<td>Pr(^{143})</td>
<td>13.7 d</td>
</tr>
<tr>
<td>Zr(^{95})</td>
<td>63 d</td>
<td>Nd(^{142})</td>
<td>stable</td>
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<tr>
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<td>67 h</td>
<td>Pr(^{144})</td>
<td>17.5 min</td>
</tr>
<tr>
<td>Ru(^{103})</td>
<td>41 d</td>
<td>Nd(^{147})</td>
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</tr>
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<td>93 yr</td>
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<td>9.3 h</td>
<td>Eu(^{155})</td>
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<td>I(^{131})</td>
<td>8.05 d</td>
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<td>Xe(^{133})</td>
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<td></td>
</tr>
<tr>
<td>Xe(^{135})</td>
<td>9.13 h</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Materials are also produced during the reactor operation by neutron activation of the coolant, as well as impurities originally present in the coolant, corrosion products circulating in the coolant, the fuel element and the structural materials of the reactor. Within the fuel, a fissionable element such as uranium-235 may, in some cases, capture a neutron without undergoing fission and produce a transuranic element, which may be a small part of wastes discharged from the chemical processing plant of spent elements.
B. RADIOACTIVE WASTES FROM REACTOR-FUEL CYCLE

Practically all of the radioactive wastes which are produced today originate from the reactor-fuel cycle. A schematic flow-sheet of the cycle is shown in Fig. 6[4]. Naturally-occurring radioisotopes are present primarily in the early stage of the cycle which includes such operations as:

Mining;
Milling of uranium ores;
Feed materials preparation and isotope separation by gaseous diffusion method;
Fuel-element fabrication.

In reactor operations most of the wastes discharged are due to the isotopes produced by neutron activation. In fuel-reprocessing plants the waste streams contain practically all of the fission products produced during the reactor operations and the fission products in the wastes are of primary concern in the handling and disposal operations.

1. Mining

Mining for uranium and thorium and, to a lesser extent, for other minerals in areas where uranium and thorium occur releases dust, soluble compounds and gases. The isotopes in the radioactive wastes associated with the mining of uranium and thorium ores are the members of the uranium decay series.

2. Milling

The primary function of milling operations is to reduce the weight and bulk of the ores by separating uranium from the secondary minerals in the main bulk of the ores. In Canada and the United States of America the concentrated ores contain about 70% U₃Ο₈ when they are discharged from the mill. With uranium ores conventional ore-separation methods used for other elements, such as specific gravity concentration and flotation, are rarely used. Very few uranium ores contain uranium-rich particles large enough in size for selective concentration by the gravity method and no satisfactory flotation reagents have been developed for uranium ores. Since these physical separation methods are not applicable, extraction of uranium is effected almost exclusively by leaching with chemical reagents.
Schematic flowsheet for origin of wastes

Fig. 6

(1) HIGH-ACTIVITY WASTE: ACTIVITY IS ORDER OF MAGNITUDE ESTIMATE, c/gal (US)
(2) LOW-ACTIVITY WASTE.

This publication is not longer valid
Please see http://www-ns.iaea.org/standards/
The milling process can be divided into three general stages of operations. In the first stage of the process the raw ores are crushed and wet-ground in a ball mill. Airborne dust particles of radium and uranium are the major radioactive wastes in this operation. In the chemical processing stage which follows, the ground ores are fed into a digester and either acidic or alkaline leaching solution is added to dissolve the ores. Ores containing limestone or other acid soluble materials are treated by an alkaline reagent and acid reagent is used for ores rich in silica and similar materials insoluble in acid in order to reduce reagent consumption. The leach solution from the digester is then further treated for selective recovery of uranium by one of three methods: precipitation, ion-exchange or solvent extraction.

Most of the radioactive daughters of uranium delivered to the mill are released with the liquid-waste stream from the chemical processing unit, and the volume of liquid waste produced is about 865 gal(US)/t of ore processed, of which about 60 gal remain with the solid waste and the rest is usually allowed to flow into a large retention pond [4]. Analyses of samples of clarified tailings-pond water from various types of uranium mills, which are shown in Table III, indicate that radium and uranium are the major contaminants in the liquid waste [7]. The final stage of the mill process handles uranium compounds which are relatively pure except for traces of other isotopes.

In the United States of America the major portion of the uranium milling industry is located in the Colorado River Basin where the four States of Colorado, New Mexico, Utah and Arizona have a common boundary. Generally the wastes from uranium ore processing in this area, including wash waters, sands and slimes, and various process liquors are discharged to tailings-ponds or lagoons [8]. Liquid volume is reduced by seepage and evaporation. Many of the tailings-ponds are located on the banks of streams and in some cases seepage and even direct discharges occur.

In 1955 the United States Public Health Service made a physical, chemical and biological survey of eight streams in the vicinity of uranium mills. Samples were taken above and below the mills and were filtered. Gross alpha and beta counts were run on the filtrate and the filter solids and these were designated dissolved and suspended solids. Soluble radium was also determined by co-precipitation with barium sulphate. The results of these analyses are presented in Table IV.
TABLE III
ANALYSES OF TAILINGS-POND EFFLUENTS [7]

<table>
<thead>
<tr>
<th>Mill process</th>
<th>pH</th>
<th>U₃O₈ (g/l)</th>
<th>Ra²²⁶ (µc/cm³)</th>
<th>Th²³⁰ (µc/cm³)</th>
<th>Th²³⁴ + Pa²³⁴ (µc/cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Acid leach - solvent extraction</td>
<td>1.5</td>
<td>0.10</td>
<td>4.0 × 10⁻⁶</td>
<td>3.9 × 10⁻⁵</td>
<td>1.2 × 10⁻⁴</td>
</tr>
<tr>
<td>Acid leach - resin-in-pulp</td>
<td>3.3</td>
<td>0.0034</td>
<td>5.4 × 10⁻⁶</td>
<td>1.2 × 10⁻⁶</td>
<td>4.8 × 10⁻⁶</td>
</tr>
<tr>
<td>Acid leach - resin-in-pulp</td>
<td>7.7</td>
<td>0.0003</td>
<td>3.3 × 10⁻⁷</td>
<td>2.8 × 10⁻⁸</td>
<td>1.8 × 10⁻⁷</td>
</tr>
<tr>
<td>Combined acid leach - resin in pulp and alkaline-leach-precipitation</td>
<td>6.9</td>
<td>0.02</td>
<td>8.1 × 10⁻⁸</td>
<td>9.4 × 10⁻⁸</td>
<td>5.1 × 10⁻⁶</td>
</tr>
<tr>
<td>Alkaline leach</td>
<td>9.9</td>
<td>0.009</td>
<td>2.0 × 10⁻⁶</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Alkaline leach - counter-current decantation - solvent extraction</td>
<td>2.0</td>
<td>0.002</td>
<td>2.7 × 10⁻⁶</td>
<td>1.1 × 10⁻⁵</td>
<td></td>
</tr>
</tbody>
</table>
**TABLE IV**

QUALITY* OF SURFACE STREAMS, 1955 [8]

<table>
<thead>
<tr>
<th>Mill</th>
<th>Gross alpha</th>
<th></th>
<th>Gross beta</th>
<th></th>
<th>Soluble radium**</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Susp.</td>
<td>Diss.**</td>
<td>Total</td>
<td>Susp.</td>
<td>Diss.**</td>
</tr>
<tr>
<td>(a) Above mills</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>2</td>
<td>0</td>
<td>2</td>
<td>4</td>
<td>28</td>
</tr>
<tr>
<td>B</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>7</td>
</tr>
<tr>
<td>C</td>
<td>10</td>
<td>3</td>
<td>13</td>
<td>16</td>
<td>6</td>
</tr>
<tr>
<td>D</td>
<td>2</td>
<td>8</td>
<td>10</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>E</td>
<td><em>T</em>*</td>
<td><em>T</em>*</td>
<td><em>T</em>*</td>
<td><em>T</em>*</td>
<td><em>T</em>*</td>
</tr>
<tr>
<td>F</td>
<td>7</td>
<td>16</td>
<td>23</td>
<td>10</td>
<td>41</td>
</tr>
<tr>
<td>G</td>
<td>2</td>
<td>31</td>
<td>33</td>
<td>32</td>
<td>35</td>
</tr>
<tr>
<td>H</td>
<td>210</td>
<td>10</td>
<td>220</td>
<td>330</td>
<td>39</td>
</tr>
<tr>
<td>(b) Below mills</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>7</td>
<td>27</td>
<td>34</td>
<td>22</td>
<td>4</td>
</tr>
<tr>
<td>B</td>
<td>8</td>
<td>41</td>
<td>49</td>
<td>15</td>
<td>0</td>
</tr>
<tr>
<td>C</td>
<td>43</td>
<td>140</td>
<td>183</td>
<td>115</td>
<td>180</td>
</tr>
<tr>
<td>D</td>
<td>24</td>
<td>124</td>
<td>148</td>
<td>23</td>
<td>38</td>
</tr>
<tr>
<td>E</td>
<td>2540</td>
<td>2160</td>
<td>4700</td>
<td>4800</td>
<td>690</td>
</tr>
<tr>
<td>F</td>
<td>5</td>
<td>0</td>
<td>5</td>
<td>0</td>
<td>28</td>
</tr>
<tr>
<td>G</td>
<td>-§</td>
<td>-§</td>
<td>-§</td>
<td>-§</td>
<td>-§</td>
</tr>
<tr>
<td>H</td>
<td>170</td>
<td>0</td>
<td>170</td>
<td>178</td>
<td>26</td>
</tr>
</tbody>
</table>

* Single grab samples

** Average natural background radioactivity: gross alpha (dissolved), 10 pc/l; gross beta (dissolved), 24 pc/l, and soluble radium, 0.3 pc/l

*** Inaccessible

§ Listed under quality of mill effluents

During 1956 samples of stream biota were taken and analysed for gross radioactivity [9]. The results are shown in Table V. Station 1 was above Mill D and represented a 15-mile reach of stream. Station 11
### TABLE V
RADIOACTIVITY OF STREAM BIOTA [8]

<table>
<thead>
<tr>
<th>Station Number</th>
<th>Type of biota</th>
<th>Gross radioactivity (pc/g)*</th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Alpha</td>
<td>Beta</td>
<td>Radium</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>Algae</td>
<td>14</td>
<td>125</td>
<td>2.2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Insects</td>
<td>3</td>
<td>260</td>
<td>1.7</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Chara</td>
<td>19</td>
<td>26</td>
<td>8.1</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Juvenile suckers</td>
<td>6</td>
<td>0</td>
<td>0.6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Dace</td>
<td>3</td>
<td>260</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Ribs of 1 sucker</td>
<td>4</td>
<td>32</td>
<td>0.2</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>Algae</td>
<td>10500</td>
<td>15600</td>
<td>3560</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>Algae</td>
<td>410</td>
<td>1130</td>
<td>200</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Insects</td>
<td>760</td>
<td>790</td>
<td>185</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Juvenile suckers</td>
<td>9</td>
<td>0</td>
<td>14.0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Bonytails</td>
<td>11</td>
<td>110</td>
<td>5.7</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Dace</td>
<td>12</td>
<td>120</td>
<td>10.6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Ribs of 1 sucker</td>
<td>12</td>
<td>0</td>
<td>1.7</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>Chara</td>
<td>85</td>
<td>45</td>
<td>26</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Insects</td>
<td>730</td>
<td>570</td>
<td>270</td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>Algae</td>
<td>1080</td>
<td>3280</td>
<td>750</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Insects</td>
<td>65</td>
<td>560</td>
<td>44</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Juvenile suckers</td>
<td>52</td>
<td>20</td>
<td>47</td>
<td></td>
</tr>
</tbody>
</table>

* Ashed weight

was about 40 miles below Mill E. It is apparent that algae concentrated alpha and beta-activity to a higher degree than the other biota and that the effects were evident far downstream. Accumulation of activity in river muds was also noted, the highest values being observed at Station 2, with gross alpha of 9900, gross beta of 10600 and total radium of 2100 pc/g. In recent years improved waste hand-
ling by the mills has substantially reduced the quantities of radioactive materials entering the streams.

3. Feed materials preparation

The ore concentrates produced at the uranium mills contain about 70% U_3O_8 and must be further processed to obtain pure uranium compounds. Certain impurities, if present even in a trace amount, have high neutron-absorbing characteristics and therefore the purity of uranium compounds to be used as reactor fuel is extremely important. Two general methods of ore purification are solvent extraction and precipitation. In the solvent extraction method ore concentrates are first dissolved in nitric acid and the uranyl nitrate in the acid solution is extracted selectively by certain oxygenated organic solvents such as tributyl phosphate (TBP), diethyl ether, or methyl isobutyl ketone. In the precipitation method uranyl peroxide (UO_4 \cdot 2H_2O) is precipitated from weakly acidic solutions of uranyl salts by hydrogen peroxide. About 1000 gal (US) of solvent extraction wastes are produced per ton of uranium processed [4]. The radioisotopes present in the liquid wastes are uranium and its daughters, which are similar to those present in the mill wastes. The final products from the purification step are uranium compounds in the form of uranium peroxide, UO_4, uranyl nitrate, UO_2(NO_3)_2 \cdot 6H_2O, ammonium diuranate, (NH_4)_2U_2O_7, and uranium trioxide, UO_3. In the subsequent conversion step the uranium compounds are calcined first to uranium trioxide and reduced to uranium dioxide which can be directly used as a reactor fuel.

As a feed material to the gaseous diffusion plant where uranium-235 is separated from uranium-238, uranium hexafluoride is produced by first converting uranium dioxide into uranium tetrafluoride with hydrogen fluoride and then to the hexafluoride by reaction with fluorine. Uranium metal is produced by reducing uranium tetrafluoride with calcium or magnesium. Gaseous wastes are produced during the reduction and hydrofluorination operations and are scrubbed to remove chemical fumes and entrained uranium [4]. The hydrofluorination step also produces liquid wastes containing a minor quantity of activity. Waste materials from uranium metal production are leached to remove contained uranium [4]. Gaseous diffusion plant operations produce a large volume of liquid wastes containing small amounts of activity [9]. Solid wastes such as uranium-contaminated ore drums, used equipment and piping, filter cakes,
iron scrap, magnesium fluoride and dolomite liners from metal reduction, and other solid residues from the feed material preparation are only slightly contaminated and can be easily handled [9].

Practice in the United States of America is typified by operations at the Feed Materials Production Center, Fernald, Ohio [11]. After simple treatment of the liquid wastes from each part of the process, they are held briefly in settling tanks and discharged to the Miami river, which is used for industrial purposes. Drinking water in the area is supplied from wells. After recovery of the uranium, solid wastes are transferred to pits dug into clay. These wastes include slurries and the excess water is drained to the river.

Uranium refining in the United Kingdom is carried out at Springfields Works, Salwick, Lancashire. Liquid wastes are treated for uranium recovery, mixed with general waste waters from the whole plant and pumped to the tidal estuary of the River Ribble. Solid wastes, including insoluble slags, discarded drums and equipment, and incinerator ash, are buried. Process residues not suitable for burial are stored on concrete rafts [12]. Gaseous wastes are cleaned by conventional equipment and discharged through low stacks [13].

4. Isotopic enrichment

The principal process for the enrichment of the isotope uranium-235 is the gaseous diffusion of uranium hexafluoride through membranes. The process takes place in vacuum-tight plant under reduced pressure and the main source of waste is the decontamination and maintenance of equipment. The principal contaminants are uranium isotopes, but plutonium and fission products also occur if recycled reactor fuel is processed. At Oak Ridge liquid waste, from which any economically significant uranium has been recovered, is passed through a settling basin to a local stream. At Paducah the discharge is direct to a stream. Solid wastes containing uranium are sent from the Oak Ridge plant to the disposal ground of the Oak Ridge National Laboratory, while those at Paducah are stored in an open pit. Both plants release traces of uranium in gaseous wastes from the vents of refrigerated traps. The depleted uranium has no market value and is stored as the hexafluoride [14].

The United Kingdom diffusion plant is at Capenhurst, Cheshire where the liquid wastes, diluted by the purge water from the cooling towers, are discharged by open brook to the tidal estuary of the River Dee [12]. Solid wastes are buried either in a disused quarry,
or in the disposal ground at Drigg, Cumberland. As in the United States, the depleted uranium is stored as the hexafluoride.

5. Fuel-element fabrication

Almost all fuel elements at present in use are either natural or slightly-enriched uranium metal or oxide in a metal sheath or enriched uranium alloyed with aluminium or zirconium. Those containing plutonium are being used on an experimental scale. Wastes from the production of natural-uranium fuel elements are similar in character to some of the wastes from uranium refining and in particular include slags and crucibles from casting operations. In general, three types of wastes are produced: liquids, combustible solids and non-combustible solids [10]. The liquid wastes are dilute nitric-hydrofluoric acid solutions used for metal cleaning and etching operations. The specific activity in these solutions is extremely low and the solutions are readily diluted and discharged to streams. Combustible wastes consist of paper or plastics used for covering work surfaces when handling uranium dioxide. The contamination level of these wastes is low and the wastes can be directly stored in drums or can be burned, with proper precautionary measures, to recover uranium. Steel and copper jackets used for enclosing the uranium-bearing ingot become slightly contaminated during the forging and hot-rolling operations. The surface contamination of such metals can be sufficiently removed by cleaning with an acidic solution and the cleaned metals can be used as non-radioactive scrap. Other solid wastes, such as asbestos gloves, refractory crucibles and stirring rods contain very little activity and are stored in drums and disposed.

The operations of the Nuclear Facilities Plant at Lynchburg, Virginia, is typical of both United States and United Kingdom practice [10]. Liquid wastes, which include pickling liquors and decontamination wastes, are treated for the recovery of uranium if necessary, neutralized, diluted by other plant wastes and discharged locally. Solid wastes are incinerated or buried in selected areas. At Lynchburg copper and steel wastes are decontaminated and sold as scrap. Some of the solid wastes contain metallic zirconium and special care has to be taken to prevent explosions during the disposal of these wastes. Among other places fuel elements are produced at both Hanford and Savannah River in the United States and at both Springfields and Dounreay in the United Kingdom. In all cases the
wastes produced are trivial in comparison with those produced from other operations on the same sites.

6. Reactor operations

During reactor operations radioactive materials are produced by fission of the fuel element and by neutron activation of the surrounding atoms which are exposed to the neutron flux in the reactor. Except for the case when the fuel elements fail and the cladding is ruptured, allowing the coolant to come in direct contact with the fuel element, the fission products are rarely released from the reactor itself. The primary radioactive wastes from reactor operations are, therefore, due to isotopes produced by neutron activation. The waste products of activation include coolant and the impurities originally present in coolant, corrosion products circulating in the coolant and the structural materials of the reactor. For coolants to have maximum effectiveness, they circulate as near as possible to the source of the heat which is the fissioning fuel nuclei. Thus they are exposed to intense fields of radiation. The nature of the resulting activation products depends upon the type of coolant used and the impurities contained therein.

(a) Water-cooled reactors

Water is a commonly used coolant material. During operation of a water-cooled reactor, the very short-lived isotopes, nitrogen-16 (7 sec), nitrogen-17 (4 sec), oxygen-19 (30 sec) resulting from nuclear reactions with the water, build-up activities, depending upon the power level, to levels approaching a few microcuries per milliliter. Because of their short half-lives they have no waste disposal significance.

The products resulting from activation of impurities in the water are, however, of prime concern. It is not always necessary for a parent element to be present in a relatively large quantity for a significant amount of an isotope be present in the effluent. In the case of phosphorus-32, for example, a considerable amount can be formed from phosphate ion in the cooling water in such a minute amount that its concentration cannot be accurately measured by conventional chemical methods. Cobalt-58 and cobalt-60 are formed from the nickel and cobalt in stainless steel used for construction.

Table VI shows the probable reactions which produce radioisotopes in the coolant [15] for a typical research reactor, the Low Intensity Training Reactor in Oak Ridge. As a coolant clean-up system,
several types of ion-exchange beds are used. The radioisotopes found on the ion-exchange beds are shown in Table VII [15].

At the Hanford Plant, Richland, Washington, the plutonium-production reactors are cooled by a single pass of purified river water. The residual impurities in the water become activated by neutron capture, and the resultant large volume of waste is discharged to the Columbia River after a delay of one to three hours in retention basins. In the case of abnormal activity in the coolant, it can be diverted to seepage trenches adjacent to the river. These trenches are also used for higher activity wastes from operations associated with the reactors. Gaseous wastes are released from high stacks [16].

The Canadian NRX reactor at Chalk River is cooled by river water, which is returned to the Ottawa River through delay tanks. Irradiated fuel rods are stored in water before processing and the water contains fission products from damaged rods. The water is circulated through filters and a continuous bleed is run into a pit in a sandy area. The water percolates away leaving most of the radio-nuclides adsorbed on the sand. Gaseous wastes from the reactor are discharged, originally without filtration, but now through filters, from a stack which provides dilution for the argon-41 [17].

Where heavy water is used as moderator and/or coolant, tritium is produced by activation of deuterium by the reaction, \[ H^2 (n, p)H^3. \] Since tritium has a relatively long half-life of 12 yr, careful handling of the coolant is necessary, though fortunately the MPC value is high.

The reactors at the Savannah River Plant near Augusta, Georgia, are moderated and cooled by heavy water. This is circulated through filters and ion-exchangers to prevent accumulation of fission and corrosion products. The capture of neutrons in the deuterium of the heavy water produces tritium. During discharge of the fuel any leakage of tritium is controlled by ventilation, use being made of a 200-ft stack to provide adequate dilution.

(b) Pressurized-water reactors

When increased temperature in the water coolant is desired and the system is pressurized to prevent boiling, the corrosion rate of the structural members increases and is reflected in an increase in the activated corrosion products. Table VIII lists typical corrosion products of such an installation. The amount and activity of wastes produced by the Pressurized Water Reactor (PWR) are shown in Table IX [15]. In this reactor the primary coolant contains lithium hydroxide to reduce corrosion and the purge water therefore contains tritium as well as dissolved and particulate activity. This water...
### TABLE VI

**PROBABLE MODES OF FORMATION OF RADIOACTIVE NUCLIDES IDENTIFIED IN THE LOW-INTENSITY TESTING REACTOR COOLING-WATER SYSTEM [15]**

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Primary mode of formation</th>
<th>Secondary mode of formation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ba&lt;sup&gt;140&lt;/sup&gt;</td>
<td>Fission product</td>
<td></td>
</tr>
<tr>
<td>Br&lt;sup&gt;80&lt;/sup&gt;</td>
<td>Br&lt;sup&gt;79&lt;/sup&gt; (n,γ) Br&lt;sup&gt;80&lt;/sup&gt;</td>
<td>Br&lt;sup&gt;80m&lt;/sup&gt; ¹⁰⁺γ Br&lt;sup&gt;80&lt;/sup&gt;</td>
</tr>
<tr>
<td>Br&lt;sup&gt;80m&lt;/sup&gt;</td>
<td>Br&lt;sup&gt;79&lt;/sup&gt; (n,γ) Br&lt;sup&gt;80m&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>Br&lt;sup&gt;82&lt;/sup&gt;</td>
<td>Br&lt;sup&gt;81&lt;/sup&gt; (n,γ) Br&lt;sup&gt;82&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>Ce&lt;sup&gt;141&lt;/sup&gt;</td>
<td>Fission product</td>
<td>Ce&lt;sup&gt;140&lt;/sup&gt; (n,γ) Ce&lt;sup&gt;141&lt;/sup&gt;</td>
</tr>
<tr>
<td>Ce&lt;sup&gt;143&lt;/sup&gt;</td>
<td>Fission product</td>
<td>Ce&lt;sup&gt;142&lt;/sup&gt; (n,γ) Ce&lt;sup&gt;143&lt;/sup&gt;</td>
</tr>
<tr>
<td>Co&lt;sup&gt;60&lt;/sup&gt;</td>
<td>Co&lt;sup&gt;59&lt;/sup&gt; (n,γ) Co&lt;sup&gt;60&lt;/sup&gt;</td>
<td>Co&lt;sup&gt;63&lt;/sup&gt; (n,α) Co&lt;sup&gt;60&lt;/sup&gt;</td>
</tr>
<tr>
<td>Cr&lt;sup&gt;51&lt;/sup&gt;</td>
<td>Cr&lt;sup&gt;50&lt;/sup&gt; (n,γ) Cr&lt;sup&gt;51&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>Cu&lt;sup&gt;64&lt;/sup&gt;</td>
<td>Cu&lt;sup&gt;63&lt;/sup&gt; (n,γ) Cu&lt;sup&gt;64&lt;/sup&gt;</td>
<td>Zn&lt;sup&gt;64&lt;/sup&gt; (n,p) Cu&lt;sup&gt;64&lt;/sup&gt;</td>
</tr>
<tr>
<td>F&lt;sup&gt;18&lt;/sup&gt;</td>
<td>O&lt;sup&gt;18&lt;/sup&gt; (p,γ) F&lt;sup&gt;18&lt;/sup&gt;</td>
<td>Co&lt;sup&gt;59&lt;/sup&gt; (n,p) Fe&lt;sup&gt;59&lt;/sup&gt;</td>
</tr>
<tr>
<td>Fe&lt;sup&gt;59&lt;/sup&gt;</td>
<td>Fe&lt;sup&gt;58&lt;/sup&gt; (n,γ) Fe&lt;sup&gt;59&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>I&lt;sup&gt;131&lt;/sup&gt;</td>
<td>Fission product</td>
<td></td>
</tr>
<tr>
<td>I&lt;sup&gt;132&lt;/sup&gt;</td>
<td>Fission product</td>
<td></td>
</tr>
<tr>
<td>I&lt;sup&gt;133&lt;/sup&gt;</td>
<td>Fission product</td>
<td></td>
</tr>
<tr>
<td>I&lt;sup&gt;135&lt;/sup&gt;</td>
<td>Fission product</td>
<td></td>
</tr>
<tr>
<td>La&lt;sup&gt;140&lt;/sup&gt;</td>
<td>Fission product</td>
<td>La&lt;sup&gt;139&lt;/sup&gt; (n,γ) La&lt;sup&gt;140&lt;/sup&gt;</td>
</tr>
<tr>
<td>Mn&lt;sup&gt;56&lt;/sup&gt;</td>
<td>Mn&lt;sup&gt;55&lt;/sup&gt; (n,γ) Mn&lt;sup&gt;56&lt;/sup&gt;</td>
<td>Fe&lt;sup&gt;56&lt;/sup&gt; (n,p) Mn&lt;sup&gt;56&lt;/sup&gt;</td>
</tr>
<tr>
<td>Mo&lt;sup&gt;99&lt;/sup&gt;</td>
<td>Mo&lt;sup&gt;98&lt;/sup&gt; (n,γ) Mo&lt;sup&gt;99&lt;/sup&gt;</td>
<td>Fission product</td>
</tr>
<tr>
<td>Na&lt;sup&gt;24&lt;/sup&gt;</td>
<td>Al&lt;sup&gt;27&lt;/sup&gt; (n,α) Na&lt;sup&gt;24&lt;/sup&gt;</td>
<td>Na&lt;sup&gt;23&lt;/sup&gt; (n,γ) Na&lt;sup&gt;24&lt;/sup&gt;</td>
</tr>
<tr>
<td>Mg&lt;sup&gt;24&lt;/sup&gt;</td>
<td>Na&lt;sup&gt;24&lt;/sup&gt;</td>
<td>Mg&lt;sup&gt;24&lt;/sup&gt; (n,p) Na&lt;sup&gt;24&lt;/sup&gt;</td>
</tr>
<tr>
<td>Np&lt;sup&gt;239&lt;/sup&gt;</td>
<td>U&lt;sup&gt;238&lt;/sup&gt; (n,γ) U&lt;sup&gt;239&lt;/sup&gt;</td>
<td>U&lt;sup&gt;236&lt;/sup&gt; (n,γ) U&lt;sup&gt;236&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>U&lt;sup&gt;239&lt;/sup&gt; ¹⁰⁻→ Np&lt;sup&gt;239&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Th&lt;sup&gt;222&lt;/sup&gt; (n,γ) Th&lt;sup&gt;223&lt;/sup&gt;</td>
<td>U&lt;sup&gt;236&lt;/sup&gt; (n,γ) U&lt;sup&gt;237&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>Th&lt;sup&gt;223&lt;/sup&gt; ¹⁰⁻→ Pa&lt;sup&gt;233&lt;/sup&gt;</td>
<td>U&lt;sup&gt;237&lt;/sup&gt; ¹⁰⁻→ Np&lt;sup&gt;237&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>Pa&lt;sup&gt;233&lt;/sup&gt;</td>
<td>Np&lt;sup&gt;237&lt;/sup&gt; ¹⁰⁺→ Pa&lt;sup&gt;233&lt;/sup&gt;</td>
</tr>
<tr>
<td>Sb&lt;sup&gt;122&lt;/sup&gt;</td>
<td>Sb&lt;sup&gt;121&lt;/sup&gt; (n,γ) Sb&lt;sup&gt;122&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>Sr&lt;sup&gt;91&lt;/sup&gt;</td>
<td>Fission product</td>
<td></td>
</tr>
</tbody>
</table>
TABLE VI (cont’d)

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Primary mode of formation</th>
<th>Secondary mode of formation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sr$^{90}$</td>
<td>Fission product</td>
<td></td>
</tr>
<tr>
<td>Tc$^{99m}$</td>
<td>Mo$^{99}$ (n,γ) Mo$^{99}$</td>
<td>Fission product</td>
</tr>
<tr>
<td></td>
<td>Mo$^{99} \Rightarrow$ Tc$^{99m}$</td>
<td></td>
</tr>
<tr>
<td>Te$^{132}$</td>
<td>Fission product</td>
<td></td>
</tr>
<tr>
<td>Xe$^{131m}$</td>
<td>Fission product</td>
<td></td>
</tr>
<tr>
<td>Xe$^{133m}$</td>
<td>Fission product</td>
<td></td>
</tr>
<tr>
<td>Xe$^{135m}$</td>
<td>Xe$^{132}$ (n,γ) Xe$^{133}$</td>
<td></td>
</tr>
<tr>
<td>Y$^{91m}$</td>
<td>Fission product</td>
<td></td>
</tr>
<tr>
<td>Zn$^{65}$</td>
<td>Zn$^{64}$ (n,γ) Zn$^{65}$</td>
<td>Cu$^{65}$ (p, n) Zn$^{65}$</td>
</tr>
</tbody>
</table>

TABLE VII

RADIOACTIVE NUCLIDES IDENTIFIED ON ION-EXCHANGE RESINS OF THE LOW-INTENSITY TESTING REACTOR COOLING-WATER PURIFICATION SYSTEM [15]

<table>
<thead>
<tr>
<th>Cation-exchange resin</th>
<th>Anion-exchange resin</th>
<th>Mixed-bed resin</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nuclide</td>
<td>Half-life</td>
<td>Nuclide</td>
</tr>
<tr>
<td>Na$^{24}$</td>
<td>15.0 h</td>
<td>I$^{131}$</td>
</tr>
<tr>
<td>Np$^{239}$</td>
<td>2.33 d</td>
<td>I$^{133}$</td>
</tr>
<tr>
<td>Zn$^{65}$</td>
<td>245 d</td>
<td>Mo$^{99}$</td>
</tr>
<tr>
<td>Co$^{60}$</td>
<td>5.2 yr</td>
<td>+Tc$^{99m}$</td>
</tr>
<tr>
<td>Ba$^{140}$</td>
<td>12.8 d</td>
<td>Te$^{128}$*</td>
</tr>
<tr>
<td>Al$^{140}$</td>
<td>40.2 h</td>
<td>+I$^{129}$*</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Cr$^{51}$*</td>
</tr>
</tbody>
</table>

* Presence not confirmed

is stored, passed through ion-exchangers if necessary, then through a gas-stripping column and finally diluted by inactive cooling water before discharge to the Ohio River. Other liquid wastes, mainly from
## TYPICAL RADIOCHEMISTRY OF COOLANT IN NAVAL REACTOR TEST FACILITY

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>Specific activity $\mu$c/ml</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>N$^{16}$</td>
<td>7.3 sec</td>
<td>100</td>
<td>$\text{O}^{16}$ in $\text{H}_2\text{O}$</td>
</tr>
<tr>
<td>N$^{17}$</td>
<td>4.1 sec</td>
<td>800$^b$</td>
<td>$\text{O}^{17}$ in $\text{H}_2\text{O}$</td>
</tr>
<tr>
<td>K$^{38}$</td>
<td>7.7 min</td>
<td>$5 \times 10^{-2}$</td>
<td>?</td>
</tr>
<tr>
<td>A$^{41}$</td>
<td>1.8 h</td>
<td>$4 \times 10^{-2}$</td>
<td>$\text{Air}$ in $\text{H}_2\text{O}$</td>
</tr>
<tr>
<td>F$^{18}$</td>
<td>1.9 h</td>
<td>$4 \times 10^{-2}$</td>
<td>$\text{Air}$ in $\text{H}_2\text{O}$</td>
</tr>
<tr>
<td>Mn$^{55}$</td>
<td>2.6 h</td>
<td>$0.5 \times 10^{-2}$</td>
<td>$\text{Steel}$</td>
</tr>
<tr>
<td>Cu$^{64}$</td>
<td>12.8 h</td>
<td>$3 \times 10^{-4}$</td>
<td>$17-4$ pH steel</td>
</tr>
<tr>
<td>Na$^{24}$</td>
<td>15 h</td>
<td>$1 \times 10^{-3}$</td>
<td>Na in $\text{H}_2\text{O}$</td>
</tr>
<tr>
<td>W$^{187}$</td>
<td>24 h</td>
<td>$3 \times 10^{-3}$</td>
<td>$\text{Stellite}$</td>
</tr>
<tr>
<td>Fe$^{59}$</td>
<td>45 d</td>
<td>$1.1 \times 10^{-5}$</td>
<td>$\text{Steel}$</td>
</tr>
<tr>
<td>Ta$^{182}$</td>
<td>111 d</td>
<td>$0.6 \times 10^{-5}$</td>
<td>$\text{Steel}$</td>
</tr>
<tr>
<td>Co$^{60}$</td>
<td>5.3 yr</td>
<td>$2.5 \times 10^{-5}$</td>
<td>$\text{Steel}$</td>
</tr>
</tbody>
</table>

---


$^b$ neutrons/(sec)(ml)

---

decontamination of equipment, are concentrated by evaporation. The condensate joins the primary coolant purge and the concentrate is made into concrete for ocean disposal with non-combustible solid waste and incinerator ash. Spent ion-exchange resin and large non-combustible solids are stored. Gases from the primary coolant stripping column are delayed by storage tanks and then diluted with ventilating air before being discharged from a stack [18].

The wastes from pressurized-water reactors in nuclear-powered naval ships are similar in character to those at Shippingport. The primary coolant is not purged, but part of it is discharged as the water expands while the reactor is being brought up to operating temperature. The activity in the coolant is kept low by ion-exchangers.
Boiling-water reactors

Boiling-water reactors produce wastes similar to the pressurized-water reactors, the difference being a considerable increase in the gaseous wastes. The Experimental Boiling-Water Reactor (EBWR) produces about 30,000 gal(US)/yr of liquid waste with a specific activity of $5 \times 10^{-5}$ μc/ml and 1000 ft³ of solid wastes containing about 0.1 c of activity, of which 10 to 20% is contained in the spent ion-exchange resin beds [20]. The major isotopes found in the coolant are listed in Table X.

(d) Air-cooled reactors

A number of reactors utilize air as a primary coolant and air is also quite often used for cooling of shielding, instrument tubes or experimental facilities. The chief radioisotopes resulting from the irradiation of clean air are oxygen-19 (half-life 29 sec), nitrogen-16 (half-life 7.3 sec), and argon-41 (half-life 1.83 h). The argon-41 is the most significant of the three because of its longer half-life. These gaseous wastes usually can be dispersed by mixing with large quantities of atmospheric air through discharge from tall stacks. However, if the cooling air contains suspended particulate matter, such as dust, some of the material will become radioactive in passing through
the neutron field and can possibly contaminate the area near the discharge. For that reason, it is good practice to clean the air before and/or after using it as a coolant. If the suspended solids are removed after irradiation they can be handled as any other solid waste would be. While not strictly an air-cooled reactor, the Argonne National Laboratory Experimental Boiling Water Reactor, when operating at 20 MW, discharges approximately 40 c/d of gaseous radioactivity, mainly argon-41, from secondary coolant systems [20].

Air-cooled reactors are in operation for research purposes at Oak Ridge National Laboratory and at Brookhaven National Laboratory. In both reactors, the air is filtered, drawn through the reactor, filtered again, this time by high-efficiency filters, and discharged from a stack. The principal contaminant is argon-41 and the radiation dose from this at ground level is controlled by a suitably designed height of stack [21, 22].

The air-cooled research reactor at Harwell discharges the cooling air from a stack after filtration on both entering and leaving the reactor. The stack is sufficient to provide adequate dilution for the argon-41, the principal activity in the discharge. (STEWART et al. (1954)). Similar arrangements were made for the cooling air from the production reactors at Windscale Works, Sellafield, Cumberland. Experience here showed that particulate uranium oxide had been able to pass the exhaust filters and, before the operation of the reactors was discontinued, had caused some local deposition of strontium [23].

(e) Gas-cooled reactors

The main liquid waste from the gas-cooled power-reactors is the purge water from the ponds in which irradiated fuel is stored before transfer to the chemical plants. At Chapelcross Works, Annan,
Dumfriesshire, fuel elements are transferred from the reactor to the storage pond. If they are known to be damaged they are segregated, but there are traces of uranium on the outside of the cans and some contamination of the water by fission products occurs. Radioactive corrosion products from the magnesium alloy sheaths also enter the water. The pond water is bled intermittently through filters and, if necessary, centrifuged, mixed with other minor wastes and diluted by the purge water from the condenser-water cooling towers before being pumped to a tidal estuary, the Solway Firth. Gaseous wastes arise from the shield-cooling air, which contains principally argon-41 and is discharged from stacks on the reactors. Some of the carbon dioxide coolant leaks from the primary circuit and controlled discharges of coolant are periodically made through the stacks when the reactors are brought to atmospheric pressure for refuelling. The larger reactors now under construction as part of the United Kingdom power programme will be refuelled under pressure and on load. The main problems set by leakage and discharge of the coolant are those of dispersal of the carbon dioxide and monoxide. The radioactive constituents present no problems. The pond-water filters and by-pass gas filters on the primary coolant circuits form the main solid wastes and these are transferred to permanent storage or burial facilities at Windscale Works, Cumberland.

(f) Liquid-metal-cooled reactors [20]

Sodium and NaK are commonly used as liquid-metal coolants in reactors. Principal isotopes found in the coolants are shown in Table XI. Unlike in the water or heavy-water-cooled reactors, the liquid-metal coolants are not processed for removal of radioactivity but the activity level is kept below the threshold limit by withdrawing

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Half-Life</th>
<th>Nuclear reaction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na$^{24}$</td>
<td>15 h</td>
<td>Na$^{23}$ ($n,\gamma$) Na$^{24}$</td>
</tr>
<tr>
<td>Na$^{32}$</td>
<td>2.6 yr</td>
<td>Na$^{31}$ ($n,2n$) Na$^{32}$</td>
</tr>
<tr>
<td>Rb$^{86}$</td>
<td>19.5 d</td>
<td>Rb$^{85}$ ($n,\gamma$) Rb$^{86}$</td>
</tr>
<tr>
<td>Sb$^{124}$</td>
<td>60 d</td>
<td>T$^{127}$ ($n,\alpha$) Sb$^{124}$</td>
</tr>
</tbody>
</table>
a sidestream of the coolant. The disposal problem of sodium and NaK metal is complicated by the violent chemical reaction of the metal with water, oxygen or carbon dioxide.

(g) Organic-moderated reactors [20]

The wastes produced from the organic-moderated reactors (OMRE) are decomposition products of polyphenyls which are used as the moderator and coolant. The specific activity of the waste is low, and the radioactive contaminants can be readily disposed of by burning.

C. IRRADIATED FUEL PROCESSING

Separation of the fission products from intimate association with the remaining fuel and the consequent handling, treatment and storage of the resulting intensely radioactive solutions create the most demanding waste-management problems in the nuclear industry. Fission products in an operating reactor parasitically capture neutrons, thus decreasing the neutron efficiency of the reactor, and, therefore, must be removed periodically. They are always intimately associated with the valuable fuel material. Present-day reactors only fission, or "burn-up", a few per cent of the fissionable atoms before fresh fuel is required for satisfactory operation of the reactor.

A natural-uranium-fuelled reactor, such as the Brookhaven Reactor, operating at 28 MW power for a year will form approximately 10220 g of fission products. (This can be roughly calculated by multiplying the one gram of fission products per megawatt reactor power x 28 MW x 365 d). Using similar approximations, the composition of the fuel at the end of one year's operation can be shown to be:

<table>
<thead>
<tr>
<th>Component</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium (all isotopes)</td>
<td>99.954%</td>
</tr>
<tr>
<td>Plutonium-239</td>
<td>0.023</td>
</tr>
<tr>
<td>Fission Products</td>
<td>0.023</td>
</tr>
</tbody>
</table>

This means that, if the total fuel loading of the reactor is 50 t, each ton of fuel will contain about 0.46 lb of fission products and the same amount of plutonium which must be separated from 1999 lb of uranium. The radioactivity of the fission products after 100 d cooling time will be $4.8 \times 10^6$ c [24]. This can be calculated from the equation:

$$\text{Activity of fission products at time } T_1 \text{ measured from start-up of the reactor} = 1.4 P \left[ (T_1 - T_0)^{-0.2} - (T_0)^{-0.2} \right] c,$$

where $T_0$ is the time in days during which the reactor operated and
Total beta particle and gamma-ray power from fission products

$P$ is the reactor power level in watts. This equation is expressed graphically in Fig. 7.

This means that the separations processing must be done in heavily shielded, remotely operated facilities. Also, the processing
must be done in amounts that will not produce a "criticality" configuration. The process wastes will contain the fission products and will be extremely radioactive. The volume of wastes from the processing varies with the particular process used.

Four main processing systems have been proposed to date:

(1) Precipitation;
(2) Adsorption (or sorption);
(3) Solvent extraction; and
(4) Volatilization.

At the present time, the most widely used reprocessing method is solvent extraction, which is the primary source of the radioactive wastes produced in the nuclear fuel cycle. A typical solvent extraction unit consists of three counter-current liquid-liquid contactors (Fig. 8).

Aqueous feed of dissolved fuel elements is introduced into the extraction column where an organic solvent extracts selectively uranium and plutonium complexes, leaving fission products behind in the aqueous phase. The organic phase is then fed into the partition column.
where an aqueous scrub solution back-extracts plutonium from the organic phase and leaves the bottom of the column. The organic solvent, containing only uranium, is fed into the strip column and is stripped free of uranium by an aqueous solution. The stripped organic solution leaves the top of the column and is recovered for recycling to the unit. The general steps described above constitute the solvent-extraction cycle. In order to obtain products of sufficient purity the cycle can be repeated any number of times; the partitioning contactor is generally included in only one cycle in a plant, the other cycles consisting of an extraction and a stripping cycle only. The aqueous solution leaving the bottom of the first-cycle extraction column is a high-level liquid waste, containing about 99.9% of the total fission products originally present in the spent fuel-elements. The specific activity of the aqueous waste depends on such factors as the irradiation history of the fuel elements in the reactor, the type of fuels used and the length of decay cooling before reprocessing. The magnitude of the activity present in typical high-level liquid wastes is shown in Table XII[9, 25]. Such wastes are usually further concentrated before storage. The potential difficulty and hazard associated with handling and disposal of such wastes is evident.

The irradiated fuel elements which are discharged from reactors go through the following three general operations at the reprocessing plant:

1. Decay cooling

   The purpose of decay cooling is primarily to allow the short-lived fission products to decay, thus reducing the amount of shielding required at the processing plant and minimizing the loss of processing reagents due to radiation damage.

   The decay characteristics of the principal fission products, which are shown in Table XIII [4], indicate the relative importance of each radioisotope encountered in waste-handling and disposal problems.

   Decay cooling is carried out in a canal, in which the irradiated fuel elements are submerged below several feet of water. The canal water may be contaminated by fission products which are released through defective cladding. In the Oak Ridge National Laboratory, two canals, each with 144,000 gal capacity, were the source of about half the activity released to the uncontrolled environment [6]. The canal water contains a gross gamma activity of 500 cpm/ml under stagnant conditions and as much as 10,000 cpm/ml when sediment on
### TABLE XII
CHARACTERIZATION OF FIRST CYCLE HIGH-LEVEL AQUEOUS WASTES FROM SELECTED SOLVENT EXTRACTION PROCESSES [9, 25]

<table>
<thead>
<tr>
<th>Process</th>
<th>Waste volume gal/g U\textsuperscript{235} consumed</th>
<th>Approximate concentration of U in feed (gm/l)</th>
<th>Waste activity (c/gal(US))</th>
<th>Total (W/gal(US))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Redox</td>
<td>0.27</td>
<td>450</td>
<td>1720</td>
<td>8.6</td>
</tr>
<tr>
<td>Purex</td>
<td>0.34</td>
<td>300</td>
<td>1320</td>
<td>6.7</td>
</tr>
<tr>
<td>Thorex</td>
<td>0.34</td>
<td>350 (Th)</td>
<td>81</td>
<td>0.4</td>
</tr>
<tr>
<td>Hexone-25</td>
<td>0.31 - 0.13</td>
<td>2-5</td>
<td>1620-3940</td>
<td>8.1-20.2</td>
</tr>
<tr>
<td>TBP-25</td>
<td>0.22 - 0.11</td>
<td>3-6</td>
<td>2580-5160</td>
<td>12.9-26.0</td>
</tr>
<tr>
<td>Zirconium-HF for enriched U</td>
<td>~ 1.50</td>
<td>0.3</td>
<td>350</td>
<td>1.75</td>
</tr>
<tr>
<td>Stainless steel-H\textsubscript{2}SO\textsubscript{4} for enriched U</td>
<td>~ 0.35</td>
<td>2-3</td>
<td>1500</td>
<td>7.5</td>
</tr>
</tbody>
</table>

Notes:

1. Wastes are untreated; they remain essentially as they leave the solvent-extraction plant and are subject to further treatment by evaporation, neutralization, chemical treatment for fission-product removal, etc.

2. Basis for activity numbers: Irradiation period 4000 MWd/t for natural uranium, $5 \times 10^{-3} n/(cm)^2$(sec); 4000 g U\textsuperscript{233} chain per ton for thorium; 53% burn-up for U\textsuperscript{235} in enriched fuel elements; 100 d decay cooling from time of reactor discharge.

3. After 100 d decay, the distribution of energy is approximately 50% gamma and 50% beta.

4. Waste volume per gram of U\textsuperscript{235} consumed is an inverse function of burn-up; i.e. for Hexone-25 at 20% burn-up, the gal/g U\textsuperscript{235} = (53/20)(0.14).

5. Waste activity varies approximately as (irradiation level)$^{0.2}$.

the canal bottom is disturbed. Mixed-bed ion-exchange is used to remove the fission products from the canal water.

2. Decladding of fuel elements

Except for the alloyed fuel elements, claddings around fuel cores can be removed without losing any appreciable amount of fissionable...
### TABLE XIII

**FISSION PRODUCTS ACTIVITY AFTER DECAY [4]**

<table>
<thead>
<tr>
<th>Fission product</th>
<th>Half-life</th>
<th>Approximate % of total activity after decay</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>100 d</td>
</tr>
<tr>
<td>Cs$^{137}$</td>
<td>26.6 yr</td>
<td>&lt; 2</td>
</tr>
<tr>
<td>Sr$^{90}$</td>
<td>28.0 yr</td>
<td>&lt; 2</td>
</tr>
<tr>
<td>Pm$^{147}$</td>
<td>2.6 yr</td>
<td>3</td>
</tr>
<tr>
<td>Ce$^{144}$ + Pr$^{144}$</td>
<td>230 d</td>
<td>45</td>
</tr>
<tr>
<td>Kr$^{85}$ (gas)</td>
<td>10.3 yr</td>
<td>&lt; 1</td>
</tr>
<tr>
<td>I$^{131}$ (gas)</td>
<td>8.1 d</td>
<td>&lt; 1</td>
</tr>
<tr>
<td>Zr$^{95}$ + Nb$^{95}$</td>
<td>63 d</td>
<td>33</td>
</tr>
<tr>
<td>Ba$^{140}$ + La$^{140}$</td>
<td>12.8 d</td>
<td>&lt; 1</td>
</tr>
<tr>
<td>Ru$^{103}$ + Rh$^{103}$</td>
<td>41.0 d</td>
<td>5</td>
</tr>
<tr>
<td>Ru$^{106}$ + Rh$^{106}$</td>
<td>1.0 yr</td>
<td>2</td>
</tr>
<tr>
<td>Sr$^{89}$</td>
<td>54 d</td>
<td>7</td>
</tr>
<tr>
<td>Xe$^{133}$ (gas)</td>
<td>5.27 d</td>
<td>&lt; 1</td>
</tr>
</tbody>
</table>

Note: Irradiation basis is exposure for one year to $3 \times 10^{13}$ thermal neutron flux.

and fertile materials. Mechanical de-jacketing requires complex machines with high operating costs [26], although the method may have the advantage of producing solid waste of low activity. The more conventional methods of de-cladding are based on the following chemical dissolution procedures:

(a) Aluminium-clad fuel [27-29]

Dissolution of aluminium cladding is effected by boiling with either an aqueous solution of sodium hydroxide or with a mixture of sodium hydroxide and sodium nitrate for 8 h. The waste solution contains 0.04\% of the total uranium and plutonium in the fuel element and the specific activity of the waste solution is in the range of tens of microcuries per milliliter. The chemical and the radiochemical nature of the de-cladding waste solution is shown in Table XIV.
TABLE XIV
COMPOSITION OF WASTE FROM
ALUMINIUM DECLADDING STEP
Decladding reagent: 18.8M NaOH + 3.6M NaNO₃

<table>
<thead>
<tr>
<th>Chemical composition</th>
<th>Radiochemical composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaAlO₂</td>
<td>Gross beta 9.1 μc/ml</td>
</tr>
<tr>
<td>NaOH</td>
<td>Gross beta 27.0 μc/ml</td>
</tr>
<tr>
<td>NaNO₃</td>
<td>Cs beta 1.2 μc/ml</td>
</tr>
<tr>
<td>NaNO₂</td>
<td>Sr beta 0.5 μc/ml</td>
</tr>
<tr>
<td>Na₂SiO₂</td>
<td>Rare earths 0.2 μc/ml</td>
</tr>
<tr>
<td>Specific gravity</td>
<td>Zr gamma 8.0 μc/ml</td>
</tr>
<tr>
<td>U and Pu (SRE I)</td>
<td>Nb gamma 6.2 μc/ml</td>
</tr>
<tr>
<td>or U and Th (SRE II)</td>
<td>Ru beta 5.0 μc/ml</td>
</tr>
<tr>
<td></td>
<td>Pu 1500 dpm/ml</td>
</tr>
</tbody>
</table>

Volume: 353 gal(US)/t of uranium

(b) Zirconium-clad fuel [26, 30, 31]
Zircaloy-2 cladding around a core of uranium dioxide may be dissolved in aqueous ammonium fluoride (Zirflex process) and ammonium nitrate in 3 h at 100° C. The ammonium nitrate oxidizes the 2% tin in the Zircaloy-2 into a water-soluble compound. The fission product content of the decladding waste solution is about 0.02% of the total, which is estimated from the equivalent amount of fuel core dissolved. Compositions of the waste solution are shown in Table XV.

(c) Stainless-steel-clad fuels [26]
Stainless-steel cladding (Type 304L stainless steel) can be removed from uranium dioxide and uranium dioxide-thorium fuel cores by dissolution in sulphuric acid (Sulflex process). The dissolution reaction runs for 6 h at 100° C. The relatively low dissolution rate of UO₂ in sulphuric acid prevents any appreciable loss of fuel. The waste characteristics are shown in Table XVI. The excess acid in
TABLE XV
COMPOSITION OF WASTE FROM ZIRFLEX DECLADDING PROCESS

<table>
<thead>
<tr>
<th>Component</th>
<th>Original waste</th>
<th>Neutralized waste</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zr$^{4+}$</td>
<td>0.2 M</td>
<td>0.18 M</td>
</tr>
<tr>
<td>Na$^+$</td>
<td>-</td>
<td>1.6 M</td>
</tr>
<tr>
<td>F$^-$</td>
<td>1.5 M</td>
<td>1.4 M</td>
</tr>
<tr>
<td>OH$^-$</td>
<td>-</td>
<td>0.05 M</td>
</tr>
<tr>
<td>NO$_3^-$</td>
<td>0.02 M</td>
<td>0.05 M</td>
</tr>
<tr>
<td>NH$_4^+$</td>
<td>0.75 M</td>
<td>-</td>
</tr>
<tr>
<td>Sn$^{4+}$</td>
<td>0.003 M</td>
<td>0.003 M</td>
</tr>
<tr>
<td>Al$^{3+}$</td>
<td>0.0075 M</td>
<td>0.006 M</td>
</tr>
<tr>
<td>U, Pu, fission products</td>
<td>~0.02% of total</td>
<td>~0.02% of total</td>
</tr>
</tbody>
</table>

Volume, gal (US)/t U | 4200 | 4600 |

Reagent: 6M NH$_4$F + 1.0M NH$_4$NO$_3$
Fuel: PWR blanket; clad contains 6 kg of Zircaloy.

TABLE XVI
COMPOSITION OF WASTE FROM SULFEX DECLADDING PROCESS

<table>
<thead>
<tr>
<th>Component</th>
<th>Original waste</th>
<th>Neutralized waste</th>
</tr>
</thead>
<tbody>
<tr>
<td>H$^+$</td>
<td>4.4 M</td>
<td>-</td>
</tr>
<tr>
<td>OH$^-$</td>
<td>-</td>
<td>0.5 M</td>
</tr>
<tr>
<td>Fe$^{3+}$, Cr$^{3+}$, Ni$^{2+}$</td>
<td>1 M</td>
<td>0.72 M</td>
</tr>
<tr>
<td>SO$_4^{2-}$</td>
<td>3.3 M</td>
<td>2.4 M</td>
</tr>
<tr>
<td>Na$^+$</td>
<td>-</td>
<td>5.3 M</td>
</tr>
<tr>
<td>U$^{6+}$</td>
<td>0.04% of total</td>
<td>0.04% of total</td>
</tr>
<tr>
<td>Th$^{4+}$</td>
<td>0.01% of total</td>
<td>0.01% of total</td>
</tr>
</tbody>
</table>

Volume, gal(US)/t U+Th | 2140 | 2950 |

Reagent: 6M H$_2$SO$_4$
Fuel: Consolidated Edison fuel assembly; clad containing 9.9 kg of 304 stainless steel.
Dilute aqua regia may be used for dissolving the cladding around thorium dioxide cores (Darex process). The waste solution contains chloride ion which may be removed before storage. The compositions of the original and treated waste solution are shown in Table XVII.

3. Fuel dissolution and chemical processing

Preparation of the feed solution for the solvent extraction unit involves contacting declad fuel cores or the entire alloyed fuel elements with a strong acid. The concentration is adjusted to meet the requirement for the particular solvent extraction process to be used. During the dissolution operation most of the gaseous fission products will be released and leave the dissolver with other gases produced in the dissolution reactions. Among the gaseous isotopes listed in

---

**TABLE XVII**

**COMPOSITIONS OF WASTE FROM DAREX DECLADDING PROCESS**

<table>
<thead>
<tr>
<th>Component</th>
<th>Original waste</th>
<th>Neutralized waste</th>
<th>Waste from which chloride has been removed</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Evaporated</td>
</tr>
<tr>
<td>H⁺</td>
<td>2.7 M</td>
<td>-</td>
<td>1 M</td>
</tr>
<tr>
<td>OH⁻</td>
<td>-</td>
<td>0.4 M</td>
<td>-</td>
</tr>
<tr>
<td>Fe³⁺, Cr³⁺, Ni²⁺</td>
<td>0.83 M</td>
<td>0.64 M</td>
<td>2 M</td>
</tr>
<tr>
<td>Cl⁻</td>
<td>1.5 M</td>
<td>1.15 M</td>
<td>50 ppm</td>
</tr>
<tr>
<td>NO₃⁻</td>
<td>~3.6 M</td>
<td>~2.8 M</td>
<td>7 M</td>
</tr>
<tr>
<td>Na⁺</td>
<td>~</td>
<td>4.3 M</td>
<td>-</td>
</tr>
<tr>
<td>U, Th, fission products</td>
<td>0.05% of total</td>
<td>0.05% of total</td>
<td>0.05% of total</td>
</tr>
<tr>
<td>Volume, gal(US)/t U+Th</td>
<td>2560</td>
<td>3330</td>
<td>1060</td>
</tr>
</tbody>
</table>

Reagent: 5M HNO₃ + 2M HCl

Fuel: Consolidated Edison fuel assembly; clad containing 9.9 kg of 304 stainless steel

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Table XVIII [32], the major contaminants are $^{131}\text{I}$ and inert gases, $^{85}\text{Kr}$ and $^{133}\text{Xe}$. At the Hanford Plant radioactive iodine has been removed by reacting the gas with silver nitrate which is deposited on a porous packing material [16]. The high-level liquid waste from the first-cycle extraction column contains about 99.9% of the total fission products. The radiochemical nature of the liquid waste is difficult to determine, but the chemical characteristics of the waste solution are not affected by the small amount of fission products present and can be specified generally for a given process. Information on the chemical compositions, as well as the activity level, of the liquid waste is important for proper treatment and disposal of the waste solution.

(a) Uranium and uranium-dioxide fuels

In the widely used Purex process [33, 34, 35], the declad fuel cores are dissolved in nitric acid and uranium and plutonium complexes are extracted selectively with tributyl phosphate (TBP), using nitric acid as the salting agent. The volume of high-level liquid waste from the extraction column is reduced by evaporation and neutralized with 50% aqueous sodium hydroxide. The Purex waste characteristics are shown in Table XIX. The high-level wastes stored in the Hanford waste tanks show corrosion rates of $10^{-5}$ to $10^{-4}$ in of mild-steel liner per month at 220°F.

(b) Stainless-steel fuels

In the Darex process dilute aqua regia is used in total dissolution of fuels consisting of $\text{UO}_2$ dispersion in stainless steel or of oxide fuels clad with stainless steel. The composition of the waste solution is shown in Table XVII. Before the solvent extraction process, the chloride ion in the aqueous solution is removed by stripping with 20 to 38% nitric acid vapour in steam.[6].

(c) Zirconium-uranium alloys [36]

Hydrofluoric acid is used for dissolving alloy fuels containing about 98% zirconium and 2% uranium. For each mole of zirconium processed, at least four moles of fluoride are required. Fluoride ion may contribute to an accelerated corrosion of any stainless-steel liner used in storage.

(d) Uranium-molybdenum alloy fuels [6]

The first-cycle waste solution is similar to the Purex waste solution except that the iron content is higher and the volume of the final waste is greater.
### TABLE XVIII

**GASEOUS FISSION PRODUCTS FROM U$^{235}$ THERMAL FISSION**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Mass No.</th>
<th>Half-life</th>
<th>Yield (%)</th>
<th>Total beta and gamma decay energy (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Br</td>
<td>82</td>
<td>35.8 h</td>
<td>3.8 x 10^-5</td>
<td>3.8</td>
</tr>
<tr>
<td></td>
<td>83</td>
<td>2.4 h</td>
<td>0.48</td>
<td>0.364</td>
</tr>
<tr>
<td></td>
<td>84</td>
<td>30 min</td>
<td>1.1</td>
<td>3.5</td>
</tr>
<tr>
<td></td>
<td>85</td>
<td>3.0 min</td>
<td>1.5</td>
<td>0.83</td>
</tr>
<tr>
<td>other very short half-life</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Kr</td>
<td>85</td>
<td>10.27 yr</td>
<td>0.3</td>
<td>0.232</td>
</tr>
<tr>
<td></td>
<td>87</td>
<td>72 min</td>
<td>2.7</td>
<td>1.57</td>
</tr>
<tr>
<td></td>
<td>88</td>
<td>2.77 h</td>
<td>3.7</td>
<td>0.36</td>
</tr>
<tr>
<td></td>
<td>89</td>
<td>1.18 min</td>
<td>4.6</td>
<td>1.3</td>
</tr>
<tr>
<td>other very short half-life</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>I</td>
<td>129</td>
<td>1.72 x 10^7 yr</td>
<td>1.0</td>
<td>0.09</td>
</tr>
<tr>
<td></td>
<td>131</td>
<td>8.05 d</td>
<td>2.9</td>
<td>0.58</td>
</tr>
<tr>
<td></td>
<td>132</td>
<td>2.4 d</td>
<td>4.4</td>
<td>2.43</td>
</tr>
<tr>
<td></td>
<td>133</td>
<td>20.8 h</td>
<td>6.5</td>
<td>1.01</td>
</tr>
<tr>
<td></td>
<td>134</td>
<td>52.5 min</td>
<td>7.6</td>
<td>1.92</td>
</tr>
<tr>
<td></td>
<td>135</td>
<td>6.88 h</td>
<td>6.9</td>
<td>1.85</td>
</tr>
<tr>
<td>other very short half-life</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Xe</td>
<td>131m</td>
<td>12.0 d</td>
<td>0.03</td>
<td>0.163</td>
</tr>
<tr>
<td></td>
<td>133m</td>
<td>2.3 d</td>
<td>0.16</td>
<td>0.233</td>
</tr>
<tr>
<td></td>
<td>133</td>
<td>5.27 d</td>
<td>6.5</td>
<td>0.196</td>
</tr>
<tr>
<td></td>
<td>135m</td>
<td>15.6 min</td>
<td>1.8</td>
<td>0.52</td>
</tr>
<tr>
<td></td>
<td>135</td>
<td>9.13 h</td>
<td>6.5</td>
<td>0.570</td>
</tr>
<tr>
<td></td>
<td>137</td>
<td>3.9 min</td>
<td>5.9</td>
<td>1.33</td>
</tr>
<tr>
<td></td>
<td>138</td>
<td>17.0 min</td>
<td>(7.6)</td>
<td>1.0</td>
</tr>
<tr>
<td>other very short half-life</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

---

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### TABLE XIX

COMPOSITION OF PUREX WASTE ORIGINATING FROM THE REPROCESSING OF URANIUM OR URANIUM DIOXIDE

<table>
<thead>
<tr>
<th>Component</th>
<th>Original waste</th>
<th>Evaporated waste</th>
<th>Evaporated and neutralized waste</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na⁺, M</td>
<td>0.02</td>
<td>0.5</td>
<td>5.1</td>
</tr>
<tr>
<td>Al³⁺, M</td>
<td>0.004</td>
<td>0.1</td>
<td>0.08</td>
</tr>
<tr>
<td>Fe³⁺, M</td>
<td>0.01</td>
<td>0.3</td>
<td>0.2</td>
</tr>
<tr>
<td>Cr³⁺, M</td>
<td>0.0004</td>
<td>0.01</td>
<td>0.008</td>
</tr>
<tr>
<td>Ni²⁺, M</td>
<td>0.0002</td>
<td>0.005</td>
<td>0.004</td>
</tr>
<tr>
<td>H⁺, M</td>
<td>2.36</td>
<td>5.6</td>
<td>-</td>
</tr>
<tr>
<td>OH⁻, M</td>
<td>-</td>
<td>-</td>
<td>0.1</td>
</tr>
<tr>
<td>NO₃⁻, M</td>
<td>2.40</td>
<td>5.8</td>
<td>5.7</td>
</tr>
<tr>
<td>PO₄³⁻, M</td>
<td>0.0004</td>
<td>0.01</td>
<td>0.008</td>
</tr>
<tr>
<td>SO₄²⁻, M</td>
<td>0.025</td>
<td>0.75</td>
<td>0.56</td>
</tr>
<tr>
<td>Si, M</td>
<td>0.0007-0.007</td>
<td>0.02-0.2</td>
<td>0.015-0.15</td>
</tr>
<tr>
<td>U and P% of total</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>Fission products % of total</td>
<td>&gt;99.9</td>
<td>&gt;99.9</td>
<td>&gt;99.9</td>
</tr>
<tr>
<td>Volume, gal(US)/t U</td>
<td>1225</td>
<td>41</td>
<td>55</td>
</tr>
</tbody>
</table>

(e) Enriched uranium–aluminium alloy

In the TBP-25 process [28], uranium–aluminium alloy fuels are dissolved in nitric acid, using mercuric nitrate as a catalyst, and uranium is extracted with tributyl phosphate in hydrocarbon diluent. The first-cycle waste solution (Table XX) contains about 99.9% of the total fission products and may be calcined to a solid with a volume of about 5400 gal(US)/t of uranium.

(f) Thorium and thorium–oxide fuels and blankets [6]

Nitric acid, with fluoride as catalyst and aluminium nitrate as corrosion inhibitor, is used for the dissolution of fuels containing thorium metal or thorium dioxide. The first-cycle extracts the
### TABLE XX

**COMPOSITION OF WASTE FROM TBP-25 PROCESSING OF 20% URANIUM - 80% ALUMINIUM ALLOY**

<table>
<thead>
<tr>
<th>Component</th>
<th>Original waste</th>
<th>Evaporated waste</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al(^{3+})</td>
<td>1.2 M</td>
<td>2.0 M</td>
</tr>
<tr>
<td>H(^+)</td>
<td>0.4 M</td>
<td>~0.6 M</td>
</tr>
<tr>
<td>NO(_3^-)</td>
<td>4.0 M</td>
<td>~6.0 M</td>
</tr>
<tr>
<td>U, Pu</td>
<td>~0.1% of total</td>
<td>~0.1% of total</td>
</tr>
<tr>
<td>Fission products</td>
<td>~99.9% of total</td>
<td>~99.9% of total</td>
</tr>
<tr>
<td>Volume, gal(US)/t U</td>
<td>44 000</td>
<td>26 400</td>
</tr>
</tbody>
</table>

Thorium and uranium with TBP and discharges high-level liquid waste (Table XXI), which can be evaporated and neutralized for storage.

### TABLE XXI

**COMPOSITION OF THOREX-PROCESS WASTE PROCESSING OF DECLAD FUEL**

<table>
<thead>
<tr>
<th>Component</th>
<th>Original waste</th>
<th>Evaporated waste</th>
<th>Evaporated and neutralized waste</th>
</tr>
</thead>
<tbody>
<tr>
<td>H(^+), M</td>
<td>3.0</td>
<td>~3.0</td>
<td>-</td>
</tr>
<tr>
<td>OH(^-), M</td>
<td>--</td>
<td>--</td>
<td>0.5</td>
</tr>
<tr>
<td>NO(_3^-), M</td>
<td>3.02</td>
<td>~6.0</td>
<td>4.3</td>
</tr>
<tr>
<td>Al(^{3+}), M</td>
<td>0.05</td>
<td>1.0</td>
<td>0.7</td>
</tr>
<tr>
<td>Na(^+), M</td>
<td>0.02</td>
<td>0.4</td>
<td>5.8</td>
</tr>
<tr>
<td>Fe(^{3+}), M</td>
<td>0.01</td>
<td>0.2</td>
<td>0.15</td>
</tr>
<tr>
<td>F(^-), M</td>
<td>0.02</td>
<td>~0.4</td>
<td>0.3</td>
</tr>
<tr>
<td>Th, U, % of total</td>
<td>~0.1</td>
<td>~0.1</td>
<td>~0.1</td>
</tr>
<tr>
<td>Fission products, % of total</td>
<td>~99.9</td>
<td>~99.9</td>
<td>~99.9</td>
</tr>
<tr>
<td>Volume, gal(US)/t Th + U</td>
<td>1440</td>
<td>72</td>
<td>101</td>
</tr>
</tbody>
</table>

72
The liquid waste can be calcined and produces about 10 gal of solid per ton of thorium plus uranium. Intermediate-level liquid wastes are also produced from the subsequent solvent extraction cycles and the solvent recovery system.

In the Hexone-25 process [37] uranium in the nitric-acid solution is extracted with hexene (methylisobutyl ketone). For a fuel containing about 3.5% uranium, about 700 l of first-cycle liquid waste (Table XXII) is produced per kilogram of uranium processed. The

<table>
<thead>
<tr>
<th>Component</th>
<th>Original waste</th>
</tr>
</thead>
<tbody>
<tr>
<td>H⁺</td>
<td>0.2 M⁻*</td>
</tr>
<tr>
<td>Al³⁺</td>
<td>1.6 M</td>
</tr>
<tr>
<td>NH₄⁺</td>
<td>1.4 M</td>
</tr>
<tr>
<td>Hg²⁺</td>
<td>0.01 M</td>
</tr>
<tr>
<td>NO₃⁻</td>
<td>6.0 M</td>
</tr>
<tr>
<td>Fe³⁺, Cr³⁺, Ni²⁺, Si</td>
<td>&lt; 1 g/l</td>
</tr>
</tbody>
</table>

* The minus sign indicates that the solution contains less than the stoichiometric amount of H⁺

volume can be reduced to 510 l by evaporation, and neutralization with 25 to 30% sodium hydroxide brings the volume of the final waste to 860 l/kg uranium.

Management of wastes at Hanford, Richland, Washington has received extensive research and development activity and is described by Parker [16]. After chemical removal of the aluminium sheaths, the natural-uranium fuel from the plutonium-producing reactors is dissolved in acid and the fission products, plutonium and uranium, are separated by solvent-extraction processes. Most of the fission products appear in an acid solution in the primary stage of the process and this solution is evaporated, with acid recovery, then neutralized and stored as a slurry in underground tanks. The volume is reduced by allowing the slurry to boil as a result of the heat released by the radioactive decay. Condensates from the concen-
tration of process solutions give rise to larger volumes of liquid containing much less radioactivity. These wastes are known as intermediate-level wastes and are allowed to percolate into the ground through underground soak-aways or "cribs". Condensate from the high-activity tanks is dealt with in the same way. The cribs and the high-activity tanks are located in dry sedimentary soil. Very large amounts of activity can be adsorbed onto this sedimentary soil before there is any penetration to the ground water some 60 m below. As soon as this occurs to a significant extent, the use of the crib is discontinued and a new one started [38]. Other intermediate wastes which are not suitable for continuous discharge because of chemical characteristics are released in batches to trenches which are then filled in to prevent dispersal of the surface activity. Process-cooling water forms a low-activity waste stream and this is disposed of to a swamp.

Solid wastes arise from many of the ancillary operations at a chemical processing plant. At Hanford, they are buried in shallow trenches in marked areas. The low rainfall, about 7 1/2 in (20 cm)/yr, and the depth of the water table below the ground level remove the need for further containment.

The dissolution of the uranium releases gaseous and volatile fission products such as krypton and iodine. The krypton is discharged to air through a stack which provides adequate dilution. The discharge of iodine is controlled partly by decay through cooling the irradiated uranium and partly by treating the off-gases from the dissolvers by chemical absorbers [39]. Radioactive particles are also produced in parts of the process and these are controlled by large sand or glass-fibre filter beds. Special attention has been paid to the removal of particles from process gases from the plutonium purification process. High-efficiency paper filters were originally used but have now been replaced by fire-resistant units.

At the Savannah River Plant, near Augusta, Georgia, irradiated fuel from the reactors is chemically declad and then dissolved and processed by solvent extraction methods broadly similar to those at Hanford [40]. High and intermediate-activity wastes are concentrated, where practicable, by evaporation, neutralized and transferred to underground tanks. Large-volume, low-activity wastes are run into seepage basins. Adsorption and dilution reduces the concentration of radioactivity in the ground water to permissible limits within 100 ft (30 m) of the basins. Solid wastes, including fuel-element components, are packaged and buried, if necessary with the aid of a
shielded crane. Gaseous wastes are treated in much the same way as at Hanford and discharged to the atmosphere.

At the National Reactor Testing Station, Idaho Falls, Idaho, the chemical processing plant recovers unconsumed fuel from enriched-uranium fuel elements. The processes are basically similar to those required for natural uranium, except that the volumes are smaller and there is no plutonium stream. The high-activity wastes from the early stages of the solvent extraction process are evaporated to reduce their volume and stored in water-cooled stainless-steel tanks. Intermediate-activity wastes are also evaporated and stored. The intermediate evaporator condensate and other low-activity streams are returned to the water table by a disposal well [41]. Solid wastes are packaged and buried in a disposal ground. The amounts are minor in comparison with the solid wastes from the reactors on the Station. During normal processing, the dissolver gases are filtered and discharged to the atmosphere. On about one day per month recently irradiated fuel is processed for the recovery of radioactive barium. The volatile iodine isotopes are scrubbed with caustic solution, passed through activated charcoal, delayed in a gas holder if the meteorological conditions are unsuitable and finally released to the atmosphere.

Irradiated natural uranium fuels from the United Kingdom power reactors are processed for the recovery of the uranium and the production of plutonium at Windscale Works, Sellafield, Cumberland [12, 13].

The fuel can be removed mechanically under water and the rods broken into manageable lengths. They are then transferred to the processing plant and dissolved in acid. The fission products are removed, and the uranium and plutonium separated and purified, by solvent extraction processes. The main liquid waste is the acid solution of fission products from the primary extraction stage. This solution is concentrated by evaporation and stored in shielded stainless-steel tanks. The concentrated solution contains no sludge and is prevented from boiling by water cooling. Medium-activity wastes arise from the later separation and purification stages. The activity is reduced by temporary storage and by a flocculation treatment before the waste is neutralized and discharged to sea from a pipeline extending about 3 km beyond the high-water mark. This pipeline is also used for the direct discharge of water from the decanning pond and of other low-activity wastes from ancillary processes such as control laboratories and the laundry [42].
Solid wastes of high activity, such as discarded fuel cans, are stored in a concrete storage block, while those of lower activity are transferred to the nearby burial ground at Drigg, Cumberland. As both Drigg and Windscale are small sites, direct burial is permitted only for low-activity wastes, there being concrete storage bays for material too active to bury.

The iodine in the gaseous wastes from the dissolvers is controlled partly by delaying the dissolving and partly by scrubbing the dissolver gases. It has not been found necessary to filter these gases or the ventilating air from the rest of the primary separation plant.

The arrangements for waste disposal from the processing of enriched uranium fuels at Dounreay Experimental Reactor Establishment, Caithness, are similar to those at Windscale. The high-activity liquids are stored and the remainder neutralized and discharged to sea. High-activity solids are buried in a shaft in the bed rock and lower-activity wastes are buried in a shallower pit in the rock. Gaseous wastes are scrubbed, filtered if necessary, and discharged from a stack [12, 42].

D. ISOTOPE UTILIZATION

The term "isotopes" is commonly used to denote those specific radioactive materials that are either separated from fission product mixtures or produced by bombarding a special material with neutrons, as in a reactor. These radioactive materials are used for medical, industrial and research purposes and, while the total amount of radioactivity involved may be exceedingly small as compared with the total of radioactivity from nuclear fission operations, the amounts are quite often sufficient to be hazardous and to require special waste handling consideration.

1. Medical applications

The diagnostic and therapeutic medical uses for radioisotopes are for the shorter-lived isotopes, iodine-131, phosphorus-32 and gold-198. The diagnostic uses are essentially tracer applications and only microcurie amounts of material are used. The therapeutic applications usually involve relatively low millicurie amounts of material per treatment, 100 to 200 mc.
The waste from therapeutic doses of iodine-131 and phosphorus-32 consists chiefly of the urine of the patients and may represent as much as 60 to 70% of the total amount given to the patient. Gold-198 is introduced into the body cavities or injected into glands or organs for the treatment of cancer. Most of the gold remains in the patient where it undergoes radioactive decay. However, there may occasionally be contamination of bed linen and gowns from drainage. The other wastes produced are associated with the administration of the radioisotopes and the care of the patient. They include microcurie quantities of left-over solutions and solid wastes, such as contaminated paper cups and wipes, glassware, syringes and rubber gloves.

Medical research programmes involve the use of microcurie to low millicurie quantities of such isotopes as calcium-45, tritium, iron-55 to 59, carbon-14, sulphur-35, sodium-24 and potassium-42 which have both short and long half-lives. In addition to the routine liquid and solid wastes indicated above, animal carcasses, which may contain 25 to 500μc of activity each, may require disposal.

Perhaps the greatest amount of radioactivity used medically is in the form of sealed sources for the treatment of cancer and other therapeutic irradiations. Cobalt-60 and caesium-137 in metallic form are almost always used for these purposes. These sealed sources have useful lives from 5 to 50 yr, being used successively for applications requiring lesser amounts of radioactivity as the activity decays.

2. Industrial applications

Sealed sources and tracer applications account for almost all the industrial uses of radioisotopes. The sealed sources are used for such applications as gauging, luminous markers and oil-well logging. Typical industrial tracer applications include interface marking in oil pipelines with antimony-124, wear studies with iron-59, catalyst circulation with zirconium-95, oil-well acidization with iodine-131 or phosphorus-32 and industrial process control with tritium. There is little or no waste associated with tracer applications because of the small quantities used and the short half-life of the radioisotope and there is almost no waste disposal required in the use of sealed sources because they are only subject to disposal when their radioactivity has decayed to levels which can no longer be used or if they are damaged.

Commercial processors and suppliers of radioactive material who prepare special types, forms and compounds of radioisotopes
and those firms which fabricate sealed sources are likely to accumulate relatively large quantities of radioactive waste because they may be using bulk curie amounts of long half-life alpha, beta and gamma-emitting radioisotopes. The waste material produced as a result of these operations consists chiefly of contaminated glassware, handling equipment, paper and residual solutions. The amounts of waste may be in the millicurie range and include long half-life radioisotopes.

3. Research laboratories

Research laboratory programmes can be quite diversified and utilize a great number of different isotopes. Usually, however, large quantities of material are not involved. In general, the wastes are in the form of left-over solutions and contaminated solids such as laboratory equipment, paper wastes, glassware and animal carcasses. The levels of activity of the wastes are usually low. However, the wastes may include a variety of types and forms of radioisotopes and may include some of the longer half-life isotopes such as strontium-90.

The problems of the disposal of waste from research establishments arise mainly from the diversity of operations rather than from the amounts of radioactivity. Isotope production results in some regular wastes from standard processes but a considerable amount of special processing or experimental work also occurs. Disposal arrangements have therefore to be flexible and capable of giving individual treatment to special batches of waste.

At the Centre d'Etudes Nucléaires at Saclay, near Paris, laboratory wastes which are not expected to be radioactive are monitored and diluted with sewage and storm water before being released to a small lake, the overflow from which runs into a local stream. Active laboratory wastes are collected in delay tanks and transferred by tank truck to the treatment plant, where they are treated by delay and by flocculation. The sludges are stored in drums awaiting a decision on the best method of permanent disposal, while the treated waste liquid is taken to the nearby Research Centre at Fontenay-aux-Roses, where it is prediluted and discharged to the local sewers [43, 44, 45].

At the Risø Research Establishment, Denmark, the dispersion in the Roskilde Fiord is poor and all liquid wastes are held and monitored before discharge. Sinks in the active laboratories are provided with de-ionized water and the sink wastes are concentrated by evaporation. The condensate is discharged to the fiord and the concentrate evaporated to a solid residue which is stored in drums [46].
At Atomic Energy of Canada Ltd., Chalk River, low-activity liquids can be discharged untreated to the Ottawa River. Other radioactive liquids are allowed to seep into the ground or contained in drums or bottles and buried as solid wastes in asphalt-lined pits. Other solid wastes are buried in the sandy soil or in concrete-lined trenches. High-activity solids are dropped into vertical, concrete-lined holes [17].

Liquid wastes at the Australian Research Establishment at Lucas Heights, Sydney, New South Wales, are segregated, treated and discharged to the tidal estuary of the Woronora River. Oysters from the estuary are used as food and a very high standard of control has consequently to be exercised. Treatment concentrates and solid wastes are buried or stored. Baling is used to reduce the volume [47].

Although the Japan Atomic Energy Research Institute is situated on the Pacific coast, the area is not suitable for the construction of a pipeline and the discharges are made close to the shore. Extensive treatment facilities are available, including flocculation, evaporation and ion-exchange. Concentrates, incombustible solid wastes and incinerator ash are stored in concrete trenches. Permanent burial and ocean disposal are under consideration. Gaseous wastes are controlled by conventional air-cleaning equipment and high-efficiency filters [48].

The practice at laboratories in the United Kingdom is typified by that at the Atomic Energy Research Establishment, Harwell, Berkshire. Surface water and treated sewage are both regarded as being free from appreciable radioactivity and are discharged into a small stream which eventually runs into the River Thames. Both these wastes are monitored and the sewage can be diverted into the radioactive disposal system if necessary. Trade wastes from inactive buildings and from cooling systems are collected, checked for radioactivity, chemically treated to make them fit for discharge into the river and pumped by pipeline to the River Thames. They can be diverted for removal of radioactivity if necessary. The more concentrated radioactive wastes are collected from laboratories in containers or run into local delay tanks. After sampling and analysis, the waste is pumped through special drains to a treatment plant or, if the activity is too high, it is taken by tank truck to the treatment area for special treatment. High-activity wastes are stored in shielded tanks and other wastes, after suitable stages of treatment, are discharged into the River Thames. The sludges resulting from this treatment are concentrated by filtration and put into drums. Solid wastes can
be stored in a shielded-area, covered to exclude rain and surface water, or may be transferred to concrete-lined steel drums for disposal at sea. Volume reduction is achieved by baling, melting, or incineration. Drums containing only small amounts of radioactive material in the form of sludges and incinerator ash are disposed of to an area in the English Channel. More active wastes are disposed of to the Atlantic Ocean at depths greater than 1500 fathoms. Gaseous wastes are cleaned by filtration and, in some cases, by electrostatic precipitation or scrubbing. They are then discharged through short stacks [49].

At Oak Ridge National Laboratory, Oak Ridge, Tennessee, the main liquid wastes are of such volume that even temporary storage would be very costly. Wastes likely to be of high-activity are segregated and the remaining large volume, low-activity streams are frequently sampled and monitored. If the activity is found to be too high to permit continued discharge to White Oak Creek, leading to the Clinch River, the wastes are diverted through a treatment plant. The high-activity streams are stored to allow some reduction by decay and are then pumped into open pits from which the water seeps into the Clinch River leaving most of the radioactivity adsorbed onto the soil. The treatment sludges are also transferred to the pits. Solid wastes are buried in a disposal area which serves other installations. Gaseous waste from cells and cabinets is of low volume and is cleansed by such devices as scrubbers, electrostatic precipitators and filters. These give rise to secondary liquid and solid wastes. Air from fume hoods and general ventilating air is filtered, if necessary, and discharged to the atmosphere [21].

The Argonne National Laboratory, Argonne, Illinois, is situated in a populated area in which considerable use is made of the ground water. Laboratory wastes are segregated in containers and delay tanks and transferred to the waste-processing plant, where the bulk of the radioactivity is converted to solid form and the liquid discharged with treated sewage to the Des Plaines River. Solid wastes are baled to reduce the volume and sent to Oak Ridge for burial. If the radioactivity is too high for it to be possible to comply with the transport regulations, the waste is stored on-site. Gaseous wastes are filtered locally if necessary and again before discharge just above roof level [50].

At the Brookhaven National Laboratory, Long Island, New York, the liquid wastes from the high-activity laboratories are passed to adjacent processing facilities where the wastes can be concentrated
by evaporation and converted to concrete for disposal at sea. Wastes from other laboratories are segregated in containers, if they are of high-activity and low volume, or run into delay tanks. These are monitored and the waste can be either passed to the sewage system or returned by truck to the processing area. The sewage passes through a settling tank and a large sand filter to a small stream feeding the Peconic River. Solid wastes are baled or mixed with concrete and cast in concrete in drums for sea disposal. Items requiring more shielding than can be provided by the concrete in the drum are stored in concrete-lined pits. Low-level solid wastes can be buried on-site. The drums for disposal at sea are dumped in 2100 fathoms in the Atlantic Ocean. Gaseous wastes are scrubbed if necessary, filtered and dispersed to the atmosphere [22].

E. AMOUNTS OF RADIOACTIVE WASTES

Quantitative information on the amounts of radioactivity discharged from installations of all kinds is sparse, but both the United Kingdom and the United States have published some information relating to atomic energy installations. These publications give some idea of the relative magnitudes of the problems at different types of installations. As already explained, these discharge figures cannot be used to assess the environmental consequences of the discharges.

Information on discharges from the establishments of the United Kingdom Atomic Energy Authority was given by DUNSTER and WIX [12] in 1960. This information has been brought up-to-date and extended and the revised data are shown in Table XXIII. Except where indicated, the figures are typical of the years 1958 to 1960 inclusive. All the figures, and especially those for solid wastes, are approximate. The establishments referred to in Table XXIII and their functions are as follows:

Springfields Works, Salwick, Lancashire: Uranium-refining and fabrication of natural-uranium fuel elements;
Capenhurst Works, Cheshire: Enrichment processes;
Windscale Works, Sellafield, Cumberland: Reactor operation and reprocessing of natural-uranium fuel;
Chapelcross Works, Annan, Dumfriesshire: Reactor operation;
Dounreay Experimental Reactor Establishment, Caithness: Reactor operation and fabrication and reprocessing of enriched-uranium fuel elements;
Harwell Atomic Energy Research Establishment, Berkshire: Atomic energy research;

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### TABLE XXIII

TYPICAL QUANTITIES OF WASTE FROM ESTABLISHMENTS OF THE UNITED KINGDOM ATOMIC ENERGY AUTHORITY [2]

<table>
<thead>
<tr>
<th>Site</th>
<th>Type of waste</th>
<th>Amount of water and radioactive content</th>
<th>Method of disposal</th>
</tr>
</thead>
<tbody>
<tr>
<td>Springfields</td>
<td>Liquid</td>
<td>$2.5 \times 10^6$ m$^3$/yr&lt;br&gt;50 c/yr, alpha&lt;br&gt;1500 c/yr, beta</td>
<td>Pipeline to tidal estuary</td>
</tr>
<tr>
<td></td>
<td>Solid</td>
<td>2000 t/yr&lt;br&gt;Less than 0.25% U</td>
<td>Burial in quarry and in disposal ground</td>
</tr>
<tr>
<td></td>
<td>Gaseous</td>
<td>Approx. 1 c/yr, alpha</td>
<td>Discharge from stacks</td>
</tr>
<tr>
<td>Capenhurst</td>
<td>Liquid</td>
<td>$2 \times 10^6$ m$^3$/yr&lt;br&gt;1 c/yr, alpha(U)</td>
<td>Open brook to tidal estuary</td>
</tr>
<tr>
<td></td>
<td>Solid</td>
<td>200 t/yr&lt;br&gt;Less than 0.1% U</td>
<td>Burial in quarry and in disposal ground</td>
</tr>
<tr>
<td></td>
<td>Gaseous</td>
<td>Approx. 0.1 c/yr&lt;br&gt;alpha (U)</td>
<td>Discharge from stacks</td>
</tr>
<tr>
<td>Windscale and Calder Hall</td>
<td>Liquid</td>
<td>$8 \times 10^6$ m$^3$/yr&lt;br&gt;90 000 c/yr, beta&lt;br&gt;40 000 c/yr, Ru&lt;br&gt;1500 c/yr, Sr&lt;br&gt;.70 c/yr, alpha</td>
<td>Pipeline to open sea</td>
</tr>
<tr>
<td></td>
<td>Solid (H.A.)</td>
<td>1000 m$^3$/yr</td>
<td>Storage in special buildings</td>
</tr>
<tr>
<td></td>
<td>Solid (L.A.)</td>
<td>10 000 m$^3$/yr</td>
<td>Burial in controlled areas</td>
</tr>
<tr>
<td></td>
<td>Effluent treatment sludge</td>
<td>1000 m$^3$/yr&lt;br&gt;100 000 c/yr beta&lt;br&gt;70 000 c/yr Ru&lt;br&gt;10 c/hr $\alpha^{41}$</td>
<td>Tank storage</td>
</tr>
<tr>
<td></td>
<td>Gaseous</td>
<td></td>
<td>Discharge from stacks</td>
</tr>
<tr>
<td>Chapelcross (1960)</td>
<td>Liquid</td>
<td>$3 \times 10^4$ m$^3$/yr&lt;br&gt;4.5 c/yr alpha and beta&lt;br&gt;8 mc/yr Sr$^{90}$</td>
<td>Pipeline to tidal estuary</td>
</tr>
<tr>
<td></td>
<td>Gaseous</td>
<td>10 c/hr $\alpha^{41}$</td>
<td>Discharge from stacks</td>
</tr>
</tbody>
</table>

Footnotes a, b, c see next page

This publication is not longer valid
Please see http://www-ns.iaea.org/standards/
TABLE XXIII (cont'd)

<table>
<thead>
<tr>
<th>Site a</th>
<th>Type of waste b</th>
<th>Amount of water and radioactive content</th>
<th>Method of disposal c</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dounreay (1959 - 60)</td>
<td>Liquid</td>
<td>$10^5$ m$^3$/yr 4000 c/yr beta 20 c/yr Sr$^{90}$ 5 c/yr alpha</td>
<td>Pipeline to open sea</td>
</tr>
<tr>
<td></td>
<td>Solid</td>
<td>6000 m$^3$/yr</td>
<td>Burial in rock</td>
</tr>
<tr>
<td></td>
<td>Gaseous</td>
<td>$0.5$ mc/hr A$^{41}$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Harwell</td>
<td>Liquid</td>
<td>$7 \times 10^6$ m$^3$/yr 15 c/yr beta 0.5 c/yr Sr$^{90}$ 0.02 c/yr alpha</td>
</tr>
<tr>
<td></td>
<td>Solid (L.A.)</td>
<td>800 t/yr 80 c/yr beta 4 c/yr alpha</td>
<td>Dump in light drums on bed of English Channel</td>
</tr>
<tr>
<td></td>
<td>Solid (M.A.)</td>
<td>50 t/yr 1000 c/yr beta 200 c/yr alpha</td>
<td>Dump in strong drums on bed of Atlantic Ocean</td>
</tr>
<tr>
<td></td>
<td>Gaseous</td>
<td>Approx. 30 mc/yr beta 1 mc/yr alpha 50 c/hr A$^{41}$</td>
<td>Discharge from stacks</td>
</tr>
<tr>
<td></td>
<td>Amersham</td>
<td>Liquid</td>
<td>2000 m$^3$/yr 2 c/yr beta 0.006 c/yr alpha</td>
</tr>
<tr>
<td></td>
<td>Gaseous</td>
<td>15 mc/week 1$^{31}$</td>
<td>Discharge from stacks</td>
</tr>
<tr>
<td></td>
<td>Aldermaston</td>
<td>Liquid</td>
<td>$5 \times 10^4$ m$^3$/yr 0.02 c/yr beta 0.06 c/yr alpha</td>
</tr>
<tr>
<td></td>
<td>Solid (L.A.)</td>
<td>400 t/yr 2 c/yr alpha 0.5 c/yr beta</td>
<td>Dump in light drums on bed of English Channel</td>
</tr>
<tr>
<td></td>
<td>Solid (M.A.)</td>
<td>50 t/yr 200 c/yr alpha 15 c/yr beta</td>
<td>Dump in strong drums on bed of Atlantic Ocean</td>
</tr>
<tr>
<td></td>
<td>Gaseous</td>
<td>Approx. 20 mc/yr beta 3 mc/yr alpha</td>
<td>Discharge from stacks</td>
</tr>
</tbody>
</table>

a Except where otherwise stated, the quantities are representative of the years 1958 - 60.
b M.A. - Medium activity. L.A. - Low activity. These terms are not used in a precise sense and usage varies from one establishment to another.
c All disposals are controlled by written authorizations issued to the Atomic Energy Authority by two Departments of Her Majesty's Government.
<table>
<thead>
<tr>
<th>Site</th>
<th>Type of waste</th>
<th>Amount of waste and radioactive content</th>
<th>Method of disposal</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hanford (1959)</td>
<td>Liquid</td>
<td>3000 c/d beta(^a)</td>
<td>Pipeline to river</td>
<td>[1]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1200 c/d Cu(^a)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>70 c/d Zn(^{65})</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.2 c/d Sr(^{90})</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(1944-58)</td>
<td>Liquid</td>
<td>1.5 \times 10^7 m^3</td>
<td>Seepage into ground</td>
<td>[2]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.5 \times 10^6 c</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Gaseous</td>
<td>1 c/d (\text{d}^{131})</td>
<td>Discharge from stacks</td>
<td>[3]</td>
</tr>
<tr>
<td>Idaho (1954-58)</td>
<td>Liquid</td>
<td>1700 c/yr beta</td>
<td>Discharge to ground water</td>
<td>[4]</td>
</tr>
<tr>
<td></td>
<td>Solid</td>
<td>7000 yd^3/yr</td>
<td>Burial in soil</td>
<td>[4]</td>
</tr>
<tr>
<td></td>
<td>Gaseous</td>
<td>100,000 c/yr beta, mainly very short half-life, and noble gases</td>
<td>Discharge from stacks</td>
<td>[4]</td>
</tr>
<tr>
<td>Oak Ridge National Laboratory (1954-57)</td>
<td>Liquid</td>
<td>(10^6) m^3/yr 250 c/yr beta 50 c/yr Sr(^{90})</td>
<td>Discharge to local stream</td>
<td>[5]</td>
</tr>
<tr>
<td>Oak Ridge Gaseous Diffusion Facility</td>
<td>Liquid</td>
<td>3000 m^3/yr 0.02 c/yr alpha(U) 0.25 c/yr alpha(U)</td>
<td>Discharge to stream and in settling basin</td>
<td>[6]</td>
</tr>
<tr>
<td></td>
<td>Gaseous</td>
<td></td>
<td>Discharge from stacks</td>
<td>[6]</td>
</tr>
<tr>
<td>Brookhaven (1957-58)</td>
<td>Liquid</td>
<td>(5 \times 10^5) m^3/yr 0.1 c/yr beta</td>
<td>Discharge to stream</td>
<td>[7]</td>
</tr>
<tr>
<td></td>
<td>Solid</td>
<td>1000 c/yr</td>
<td>Dump in drums on bed of Atlantic Ocean</td>
<td>[7]</td>
</tr>
<tr>
<td></td>
<td>Gaseous</td>
<td>700 c/hr A(^{41})</td>
<td>Discharge from stacks</td>
<td>[7]</td>
</tr>
</tbody>
</table>
Radiochemical Centre, Amersham, Buckinghamshire: Preparation and commercial sale of radioisotopes; Atomic Weapons Research Establishment, Aldermaston, Berkshire: Research associated with atomic weapons and their detection and effects.

Table XXIV shows similar information about some of the installations for the United States Atomic Energy Commission. The quantities of waste are approximate and are typical of the years preceding 1958. The installations concerned and their functions are as follows:

Hanford Atomic Products Operation, Richland, Washington: Reactor operation, preparation and reprocessing of natural-uranium fuel elements;
National Reactor Testing Station, Idaho: Reactor operation and testing, fuel-element reprocessing;
Oak Ridge National Laboratory, Tennessee: Atomic energy research and isotope production;
Brookhaven National Laboratory, Long Island, New-York: Atomic energy research;
Oak Ridge Gaseous Diffusion Plant, Tennessee: Enrichment processes.
II. MAIN TRANSFER MECHANISMS OF RELEASED WASTES

CASE 1. RADIOACTIVE WASTES IN RIVERS, STREAMS, LAKES OR OCEANS

1.1. Hazards related to use as drinking water

1.1.1.** Irradiation of gastrointestinal tract of man or animals.
1.1.2.* Transfer of radioactive materials to bloodstream and irradiation of blood.
1.1.3.** Transfer to critical organ of deposition (e.g. liver, kidney, bone or thyroid) as determined by the specific radioelement and its chemical and physical form.
1.1.4.** Irradiation of reproductive organs from materials passing through the body, deposited in specific organs, or in rare cases deposited in the reproductive organs.
1.1.5. Retention of filtrable radioactive materials in water purification systems, leading to external irradiation.
1.1.6.* Retention of non-filtrable materials in ion-exchange systems (e.g. water softeners) or in flocculent deposits in municipal water systems.
1.1.7. Scaly deposits of radioactive materials from boiling hard water.
1.1.8. Uptake by foods washed or cooked in domestic water.

1.2. Hazards arising from immersion in the water

1.2.1. External irradiation from the ambient water while swimming.
1.2.2. Additional irradiation from materials attached to mud, rocks, piling, aquatic plants; or, in fact, any surface.
1.2.3. Irradiation from materials deposited on the skin or clothing.
1.2.4.* Swallowing of water, equivalent to Case 1.1.
1.2.5. Irradiation similar to Cases 1.2.1. and 1.2.2. from use of sanitary water.

1.3. Hazards arising from close approach to the water

1.3.1. External irradiation from river, lake, reservoir or ocean.
1.3.2. External irradiation from mud, sand, vegetation, or salt deposits near the shore, on beaches, or tide flats.

---


* Minor contributor to total exposure

** Major contributor to total exposure
1.3.3. External irradiation from water while boating, water ski-ing, fishermen wading, etc.
1.3.4. Augmentation of 1.3.3. by radioactive deposits on the boat structure, fishing gear, navigation locks, etc.
1.3.5. Inhalation of contaminated spray. (→ Case 2.)
1.3.6. Deposition of spray on skin or clothing. (→ Case 2.)
1.3.7. Similar situations from domestic use of sanitary water.

1.4. Hazards arising from irrigation

1.4.1. Irradiation from water in canals or ditches.
1.4.2. Retention of radioactive material on the surfaces of canals or ditches, persisting in the absence of irrigation water.
1.4.3.* Atmospheric pick-up of radioactive materials from these cases or from irrigated soils. (→ Case 2.)
1.4.4. Direct spraying of plant surfaces with contaminated water by overhead irrigation.
1.4.5.** Immediate uptake of radioactive materials by plant roots from the water or delayed uptake following retention in the soil, in either case leading to incorporation in the plant structure; this can be followed by direct irradiation from the crops, or ingestion of the radioactive material by man or range animals.
1.4.6. Further transfer and dissemination of radioactive materials by birds or insects.
1.4.7. Transfer to the atmosphere by burning of crops.
1.4.8.* Radioactive milk, eggs, or meat from range animals or poultry fed as in 1.4.5.

1.5. Hazards arising from biological chains in fresh water

1.5.1.* Pronounced uptake of radioactive materials by attached algae or by plankton (free-floating organisms).
1.5.2.* Transfer to snails, insect larvae, etc. directly or from 1.5.1.
1.5.3.* Transfer to higher forms including edible fish.
1.5.4.** Ingestion of fish by man.
1.5.5.* Transfer to aquatic plants.

* Minor contributor to total exposure
** Major contributor to total exposure
+ Symbols such as (→ Case 2.) mean that this case leads also to some of the consequences of another case, such as Case 2. in this example.
1.5.6.* Transfer to waterfowl from aquatic plants, insects, etc.
1.5.7.* Ingestion of contaminated fowl by man.
1.5.8. More exotic food chains such as ingestion of contaminated insects by frogs and the frogs' legs by man.
1.5.9. Mass emergence of contaminated ephemeral insects, their infestation of dwellings and consumption by birds.
1.5.10. Secondary contamination of bird nests and the like from excreta.
1.5.11. Absorption of radioisotopes into fish eggs.
1.5.12. Perpetual recycling of isotopes through biological chains through death and decomposition of organisms.

In any of these transfers, the transmitter as well as the receptor will receive radiation exposure.

1.6. Hazards arising from biological chains in the ocean+
1.6.1. Uptake similar to 1.5.1. but including seaweeds.
1.6.2. Transfer to invertebrates as in 1.5.2. but shellfish are of greater significance.
1.6.3. Transfers to fish, birds and other animals as indicated in 1.5.3., 1.5.5., 1.5.6., 1.5.10, 1.5.11 and 1.5.12.
1.6.4.* Ingestion of radioactive materials by man from eating seaweed, shellfish, fish or fowl.

1.7. Hazards related to industrial processes
1.7.1. Direct irradiation equivalent to 1.3.
1.7.2. Preferential adsorption of radioactive materials on food products during washing.

1.8. Hazards related to sewage disposal
1.8.1. Accumulation of radioactive materials in drains later requiring service.
1.8.2. Accumulation in sludge and filter beds.
1.8.3. Accumulation in bacteria, affecting their function.
1.8.4.* Concentration and return to agriculture via fertilizers.

* Minor contributor to total exposure
+ Although marine organisms pick up radioactive materials by processes which are similar to those for fresh-water forms, the degree of biological concentration for any particular radio-element may be considerably different. For example, radio-caesium is highly concentrated in fresh-water organisms but apparently is not concentrated in substantial amounts from sea water.
CASE 2. RADIOACTIVE WASTES RELEASED TO THE ATMOSPHERE

2.1. Hazards related to inhalation

2.1.1. Direct irradiation of the lung due to radioactivity in the contained air.
2.1.2. Solubility of radioactive gases or vapours and transfer to the blood and specific organs of deposition. (→1.1.2. and 1.1.3)
2.1.3. Deposition of radioactive particles on the surfaces of lung, bronchi and upper respiratory passages, giving intense local irradiation.
2.1.4. Solubility of some of these particles. (→2.1.2.)
2.1.5.** Transfer of particles to lymph nodes of the lung.
2.1.6. Removal of particles by ciliary action, leading to ingestion. (→1.1.)

2.2. Hazards from radioactive material in ambient air

2.2.1.* External irradiation of the human or animal body or plant tissue.

2.3. Hazards related to deposition of materials

2.3.1. Irradiation from deposition on skin of man or animals or on clothing.
2.3.2. Transfer of radioactive materials from other surfaces to skin.
2.3.3. Secondary transfers to food, cigarettes etc. leading to ingestion.
2.3.4.** Deposition on crops, followed by transfer to man or animals.
2.3.5.** Secondary transfer of the radioactive materials through use of products such as milk, eggs and meat.
2.2.6.* Deposition on soil (→1.4.5.)
2.3.7. Deposition on bodies of water. (→Case 1.)
2.3.8. External irradiation from deposited materials.
2.3.9. Transfer from one animal to a predatory animal.
2.3.10. Washing by rain or floods into water systems. (→Case 1.)
2.3.11.* Real fall-out of individual highly-active particles or flakes onto hair or skin.

* Minor contributor to total exposure
** Major contributor to total exposure
CASE 3. GROUND DISPOSAL OF LIQUID WASTES

3.1. Hazards at the disposal site

3.1.1. If to surface ponds, the hazards are repetitious of many parts of Case 1, particularly 1.3. (The ponds may be particularly attractive to wildfowl.)

3.1.2. If contaminated surface is dried, transfer of radioactive materials to atmosphere by wind pick-up of soil (→ Case 2.)

3.1.3. Direct irradiation from a source just below ground.

3.1.4. Seepage of radioactive gases or vapours from below ground.

3.1.5. Flash floods removing superficial active materials. (→ Case 1.)

3.2. Hazards resulting from underground travel

3.2.1. Return of radioactive material to plant life from deep-rooted plants.

3.2.2. Subsequent leaching out of radioactive material apparently fixed in soil.

3.2.3. Chemical reactions releasing materials as incompatible wastes are later added.

3.2.4. Slow fall of wastes under gravity when apparently safely separated from the ground water.

3.2.5. Entry into ground water and subsequent recovery from wells (→ Case 1.)

3.2.6. Underground travel to springs or rivers. (→ Case 1.)

III. RADIATION DOSE CALCULATIONS

A. EXAMPLE CALCULATION OF THE PERMISSIBLE SIZE OF A SPECIAL GROUP

The ICRP has proposed a maximum permissible genetic dose to the whole population of 5 rem. This means that the dose to the gonads from all sources, other than natural background and irradiation given to patients in medical procedures, averaged over the whole population and including occupational and special groups, shall

* Minor contributor to total exposure
not exceed 5 rem at the mean age of child-bearing, which is assumed to be 30 years. The suggestion is made by the ICRP that the national regulatory bodies should decide what fraction of the total permissible population dose should be allotted to persons occupationally-exposed to radiation, to special groups such as those living near atomic establishments, and to the general population, leaving a reserve for future developments. An example is given in which 1.0 rem is allotted to occupationally-exposed workers, 0.5 rem to special groups and 2.0 rem to the population at large, leaving 1.5 rem as the reserve.

The size of the special group, relative to the size of the whole population, at which the genetic dose becomes more significant than the exposure of the individual, will depend on the apportionment of the genetic dose which is allotted to the special group and upon the variance of individual exposures.

A calculation of the permissible size of a special group can be made if the average exposure of the group is known. If there are N people in the whole population, and the apportionment for a special group B(c) is \( D_{30} \) rem, then that group may receive up to a total dose of \( D_{30} \times N \) rem over a 30 yr period. If the average gonad dose within the group is \( D \) rem/yr, and there are n people in the group, the total dose accumulated by the group will be

\[
\text{n} \times 30 \times D \text{ rem}
\]

Hence

\[
D_{30} \times N = n \times 30 \times D
\]

and

\[
n/N = D_{30} / 30 \times D.
\]

This is the fraction of the whole population that can be permitted to be in this group.

If the size of the special group is large, the maximum permissible dose to individuals in the group may need to be less than the 0.5 rem per year recommended for individuals of group B(c). Such lower dose limits can be calculated as follows: if we have a special group to which is apportioned a genetic dose of \( D_{30} \) rem, and this group constitutes a fraction \( n/N \) of the whole population, the average permissible annual gonad or whole body dose \( D \) to an individual member of the group must be less than

\[
D = \frac{D_{30}}{30 \times n/N} \text{ rem/yr}.
\]
For example, if $D_{30} = 0.5$ rem and $n/N = 0.1$ (i.e. 10% of the whole population) then $D = 0.17$ rem/yr. If the distribution of dose should be very uniform throughout the special group this latter limitation could prove more limiting than the individual maximum permissible dose of 0.5 rem/yr.

B. EXAMPLE CALCULATION OF EXPOSURE TO MAN LIVING IN A CONTAMINATED ENVIRONMENT

To deal with situations in which one source of exposure is contaminated food it is possible, in accordance with the ICRP, to assume that after ingestion the radionuclides contained in food will behave in the same manner as when they are contained in water. Thus it is possible to apply the MPC figures for water to the consumption of contaminated food. The problem resolves into two questions:

(1) What is the exposure ($D_a$) to the critical organ from a single isotope for an average individual (adult, occupationally or non-occupationally exposed, or a member of the public at large) in a given group of the population for a given period of time? This question can be answered only for adults, or more precisely, for the "standard man", since this is the only "average person" for whom the known biological parameters apply. In practice, safety factors must be introduced for assessing the exposure of children or individuals varying greatly from the "standard man".

(2) What is the exposure ($D_b$) to the critical organ from both internal and external sources of radiation and from more than one radionuclide? The internal exposure may result from many causes including the consumption of contaminated food. The external exposure may be due to contact with contaminated fishing gear, contaminated mud or sediment or simply living or boating on or by a contaminated river. In occupational exposure, added sources of external radiation such as neutron and $\gamma$-radiation etc. may also be present.

One way to answer these two questions would be to proceed as follows:

(a) The average level of contamination in food should be determined by actual analysis. It is essential to know in detail the food habits of the individuals in a group of people under investigation, in particular, the type, average quantity of the daily intake of the contaminated food and the duration for which it is envisaged that contaminated food will be consumed at a given level of contamination.
Great judgement and discretion should be exercised, because in practice the situation may be further complicated by many factors such as variations in the relative abundance of the different radionuclides in food, the relative abundance or scarcity in the diet of other radionuclides with similar chemical properties, the wide range of physiological differences, and differences in habits, age, sex and the chemical form of the radionuclides. Therefore, in the present state of knowledge, a word of caution should be stated regarding the modification and adaptation of values of the MPC’s for application to other situations than those specified.

(b) If a person drinks 2200 cm$^3$/d of water which is continuously contaminated at an average concentration of (MPC)$_{wA}^x$ (x is the critical organ, A denotes a given isotope, w refers to the fact that water is contaminated and the maximum permissible concentration is the 168 h per week value listed by ICRP), then the individual, at some time during the 50 yr period following commencement of intake, will accumulate the maximum permissible body burden of radionuclide A in critical organ x. At that time (and not before) organ x will be exposed to the maximum permissible (occupational) dose rate. Consuming a quantity g grams of food daily contaminated with a concentration $C_{muc/cm^3}$ of radionuclide A would be similar to consuming water contaminated with radionuclide A with a daily intake of water equivalent to 2200 cm$^3$ at a concentration equivalent to

$$\rho_{wA} = \frac{g\times C}{2200}. \quad (1)$$

(c) To answer question (1) we use the formula

$$[\rho_{wA}/(MPC)_{wA}^x] \times L_x = D_a \text{ rem.} \quad (2)$$

$L_x$ is the average weekly dose permitted to organ x in rem for an occupational worker. Substituting for $\rho_{wA}$ as computed in Eq. (1) and since $(MPC)_{wA}^x$ is known from ICRP tables and $L_x$ is known from the ICRP basic rules, then $D_a$ can be calculated.

(d) To answer question (2) a similar equation can be used. For an average individual who is exposed to concentrations $\rho_{A}$, $\rho_{B}$ and $\rho_{wA}$, $\rho_{wB}$ $\mu$e/cm$^3$ of radionuclides A and B in air and water (or food as computed in Eq. (1)) and also external sources giving doses $R_{\gamma}$, $R_{n}$ to a given organ x for $\gamma$ and neutron radiation respectively, and if $L_x$ rem is the average weekly dose (occupational) permitted to
organ x by the ICRP basic rules, then the total dose $D_b$ to organ x for that period of time is given by

$$\left[ \frac{\rho a_A}{(MPC)_{aA}^x} + \frac{\rho a_B}{(MPC)_{aB}^x} + \ldots + \frac{\rho w_A}{(MPC)_{wA}^x} + \frac{\rho w_B}{(MPC)_{wB}^x} \right] L^x + \frac{R_x^y + R_x^n}{\gamma} = D_b \text{ rem.}$$

Knowing $L^x$ from the basic ICRP rules and the MPC from the ICRP Table I of Committee II Report, computing $\rho w_A$ and $\rho w_B$ as shown in Eq. (1), and knowing $\rho a_A$ and $\rho a_B$ from measurements, make possible the calculation of $D_b$. If there is no inhalation exposure the $\rho a_A$ and $\rho a_B$ are omitted. A word of caution in dealing with a mixture of radionuclides should be mentioned. If the mixture of radionuclides irradiates a major portion of the body at somewhat comparable rates, the calculation is essentially as described except that the MPC values based on total body should be used. The formula would be:

$$\left[ \frac{\rho a_A}{(MPC)_{aA}^{T.B.}} + \frac{\rho a_B}{(MPC)_{aB}^{T.B.}} + \ldots + \frac{\rho w_A}{(MPC)_{wA}^{T.B.}} + \frac{\rho w_B}{(MPC)_{wB}^{T.B.}} \right] L^x + \frac{R_x^{T.B.} + R_x^{n,B.}}{\gamma} = D_{T.B.}$$

These calculations are based on the ICRP Committee II Report. For further detailed explanations see pages 23 to 27 of that report.

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