

The Philosophy and Practice of Radioactive Waste Disposal

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This general review is largely concerned with the problems of disposing of the large amounts of highly-radioactive waste that will result from a large-scale nuclear power program employing fission reactors. The dangers of dilution-dispersal are outlined, and particular attention is drawn to the possibility of biological reconcentration of the dispersed radioactivity. The conclusion is reached that it is imperative to find some method of permanently storing the activity in a non-leachable solid form, if the fullest use is to be made of nuclear power production. At present the "temporary expedient" of tank storage in liquid form is the only practicable method, but by 1965 other methods will have to be found.

INTRODUCTION

Radioactively contaminated waste is potentially harmful, and must be disposed of in such a way that it cannot form a hazard to mankind. In practice we can recognise two schools of thought on the disposal of radioactivity—the "concentrate and store" school and the "dilute and disperse" school. At the present time each method is used to a greater or lesser extent, which is largely determined by expediency. There is as yet no international control of the disposal of radioactive waste, although this is becoming a matter of increasing importance and urgency as the number of atomic energy sites multiplies. Of the three types of waste involved, gas, liquid and solid, the second is of the greatest immediate importance, but in a large power program it will also be necessary to consider the escape of radioactive gases of long half-life to the atmosphere, in particular krypton 85. The toxicities of radioactive materials are many orders of magnitude higher than those of non-radioactive substances, and extraordinary care has to be taken to avoid contaminating air, water, and food supplies.

Although for a given absorption of energy the effect on the human body is the same whether the source is internal or external, from the point of view of waste disposal the ingestion hazard is the more important. It must, of course, be realised that there is slight natural radioactivity in the air we breathe and everything we eat and drink, deriving from minute traces of the natural radioactive elements. This background may vary from one place to another, and is in addition to the irradiation by cosmic ray particles to which we are continually exposed. The problem is to avoid increasing this background to a dangerous level, although unfortunately it is not easy to decide what level of activity constitutes a danger.

The followers of the "dilute and disperse" school derive their philosophy from the fact that we are all continually exposed to radiation throughout our lives, and they argue that small increases in the background will produce no measurable ill-effects in the exposed organism. Based on clinical observations dating from the

discovery of radium, and on experimental data acquired in the last 10 to 15 years, upper limits have been laid down for exposure to external radiation and for radioactivity in air and drinking water. These are the so-called maximum permissible levels or concentrations, usually designated m.p.l. or m.p.c. These are believed, on the basis of present knowledge, to be unlikely to produce detectable ill-effects in human beings continuously exposed to them. These maximum permissible concentrations are exceedingly minute by the ordinary standards of chemistry. Some of the more important ones are given in Table 1.

This philosophy would justify the disposal of radioactive liquid into a domestic water supply if this were done in such a way that the ingested concentrations did not exceed the maximum permissible ones for a large population. Furthermore, this might be achieved solely by dilution, as in the case of the activity induced in the Columbia River by the Hanford plutonium piles, or by a combination of chemical treatment and subsequent dilution as at Harwell, where the Thames provides the dilution and also part of the drinking water for London and other towns.

TABLE 1.—DRINKING WATER LEVELS FOR CONTINUOUS EXPOSURE OF LARGE POPULATIONS (I.C.R.P., 1955)

Nuclide	Drinking water level for continuous exposure for large populations (microcuries/ml)	ppm.
Pu239	1.5×10^{-8} microcurie/ml	2.4×10^{-11}
Sr90	8×10^{-9}	6×10^{-11}
Ra226	4×10^{-10}	4×10^{-10}
Rn222	2×10^{-8}	1.3×10^{-13}
I131	3×10^{-7}	2×10^{-12}
Cs137	1.5×10^{-5}	2×10^{-7}
Air level for continuous exposure of large populations		
I131	3×10^{-11}	
Rn222	10^{-9}	
A41	5×10^{-9}	
Xe135	2×10^{-8}	
Kr85	4×10^{-8}	

Compare these values with the following estimated Ra content of tap water supplies. (Lowden & Solon, 1956).

Location	Activity Present (microcuries/ml)
Deep wells, near Chicago, serving 100,000 people	$6-16 \times 10^{-9}$
Joliet, Ill. (pop. 50,000)	6×10^{-9}
Frankfurt a. M.	0.3×10^{-9}
Average for 42 U.S. cities (range from $0.0-0.17 \times 10^{-9}$ microcurie/ml)	0.042×10^{-9}
Thames, below AERE Harwell discharge point	0.01×10^{-9}

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On the other hand, the "concentrate and store" school believes that any increase in activity of the background is potentially dangerous, and that when the effects are integrated over a large population, they may materially add to the burden of suffering and disease to be borne by the community. The tolerance levels laid down by the I.C.R.P. have been gradually lowered over a period of years, and there now appears to be general agreement among radiobiologists that there is no threshold level of irradiation below which the exposed organism would be free from damage. The difficulty is to calculate the effects of chronic exposure to low levels of irradiation and to relate the hazard to other dangers of life which are normally accepted. Because the release of radioactivity is an irreversible process, and the rates of decay of the biologically significant nuclides very slow ($T_{1/2}$ Cs137 = 30 years, Sr90 = 28 years and Pu239 = 24×10^4 years), it is obvious that the emphasis should be on caution until further data have been collected. Furthermore, the calculations on which the so-called safe levels of intake are based can be upset by unexpected concentration steps which may occur in animal or vegetable matter forming part of a human food cycle. Certain organisms have extremely large concentration factors for some elements present in fission products, and this is particularly probable where the element is not normally present in sufficient quantities for it to be in equilibrium between the organism and its environment. Under such circumstances biological concentration factors up to 850,000 have been reported by Krumholtz (1954) (in this case for P32). Because of such possibilities, and because many more data are required on the human uptake of radioactive elements and on the long term effects produced on the body by chronic exposure to low concentrations, the "maximum permissible levels" should be regarded merely as *upper limits that are thought to be tolerable*, and until further information is available, actual discharge concentrations should be kept as far below these levels as possible. In practice, however, there seems to be a growing tendency for those responsible for waste disposal from atomic energy sites—engineers, and not biologists or geneticists—to look on the maximum permissible levels as routine discharge levels, to be approached as closely as possible at all times so as to reduce the amount of storage or chemical treatment that is needed. It must be emphasised here that the maximum permissible concentrations refer to intake only, not to discharge levels. In relating the two, concentration factors have to be taken into account. Rather too great a reliance is perhaps placed on these maximum permissible concentrations, without the gaps in the knowledge on which these figures are based being realised.

* The "maximum permissible concentrations" are laid down for occupationally exposed persons under medical supervision. For small non-occupationally exposed populations, the levels must not exceed 1/10th of the m.p.c., while for large populations the tacit agreement is that the levels must not exceed 1/100th of the m.p.c.

Almost complete containment of all radioactive wastes from atomic energy sites could be achieved if desired. (The qualification is necessary because a limiting figure for the amount discharged is set by the volume discharged and the limit of detection for the radio isotopes involved.) However, removal of all radioactivity down to such limits would be expensive and under present circumstances would probably make the cost of nuclear power unattractive. It is, therefore, up to a community to balance its need for nuclear power against the risk involved in raising the radioactive content of the environment. At the present stage of atomic energy development, the total amounts of activity that have been released from nuclear energy establishments are small, and the possibility of harmful effects probably negligible. (This is not true of H-bomb debris; because of the quantities of radioactive nuclides released and the wide dissemination, it is certain that their effect will *not* be negligible.) As the use of radioisotopes becomes more widespread and atomic energy sites multiply, however, the practice of dilution and dispersal may have to be modified considerably to prevent exposing large sections of the population to levels much above the background.

PRESENT PRACTICE

At present most major waste-producing sites effect a compromise. Very high level wastes (of the order of 100 curies/litre) are stored. Intermediate level wastes are chemically treated to yield a low-level liquid and a sludge containing most of the activity, which also has to be stored. The effluent is disposed of (with other low-level waste) by dilution and release to inland water, or to the sea. The usual chemical treatments involve flocculation with calcium phosphate or aluminium hydroxide, at a pH of about 11. Such a process may be used in conjunction with columns of vermiculite, or even wood sawdust, which will remove most of the remaining activity from solutions. Small quantities of short-lived isotopes, such as may be used therapeutically in hospitals or as tracers in small research laboratories, can be disposed of to domestic sewers, but the increase of this practice will have to be watched to prevent build up in pipes, sewers and sewerage treatment plants.

At present, the United Kingdom is one of the few countries practising dispersal on any considerable scale. At Windscale, for example, 6,000 curies of mixed beta-gamma emitters (mainly 9-month-old fission products, the bulk of which will be Sr90, Nb95, Cs137, Ce144 and Pm147), are discharged to the Irish Sea each month, while at the new Dounreay processing site it is hoped to discharge 1,000 curies per day of beta-gamma emitters. As an example of the somewhat unusual phenomena which may arise during waste disposal, the discharge levels at Windscale are thought to be limited by the uptake of ruthenium by seaweed growing nearby, which is eaten in parts of Wales (Farmer, 1956), and at Dounreay it will be limited by the accumulation of plutonium in the flesh of the considerable

number of lobsters caught for market in this area.

SEA DISPOSAL

At one time it was thought that very large amounts of activity could safely be injected into the sea, because this already contains considerable natural radioactivity. (Table 2.)

TABLE 2.—NATURAL RADIOACTIVITY IN THE OCEANS (REVELLE, 1955)

Nuclide	Total activity in oceans (curies)	Approximate specific activity	
		d/ml/sec	microcuries/ml
K40	4.6×10^{11}	1.2×10^{-2}	3.2×10^{-7}
U238	3.8×10^6	1×10^{-4}	2.7×10^{-9}
Th232	8×10^6	2×10^{-7}	5×10^{-12}
Ra226	1.1×10^6	3×10^{-5}	8×10^{-10}
H3	1.2×10^7	2.5×10^{-5}	7×10^{-10}

Ignoring the practical difficulties of effecting perfect mixing in any such disposal operation, it has, however, been shown by several authors (Rodger, 1954; Renn, 1955; Claus, 1955) that ocean disposal is out of the question as a remedy for the problems of waste disposal created by even a moderate-sized program of nuclear power production, because the quantities of radioactive material involved are enormous compared to anything previously experienced.

For example, it has been estimated that if the world production of electricity by fission reactors increased to the point where it was consuming 1,000 tons of fissile material per year, the equilibrium amount of artificial activity arising from Sr90 alone would be about 7×10^{10} c. requiring between 5 and 10 per cent. of all the oceans for dilution to drinking water tolerance (Rodger & Fineman, 1956). As Glueckauf has pointed out, this amount of fission product waste is probably an outside estimate of the amount with which we shall have to cope in the foreseeable future (Glueckauf, 1955a). However, the situation is still alarming, even for a much more modest program such as that for the Central Electricity Authority in the U.K. At the present rate of expansion this would call for the consumption of about 20 tons of fissile material per year by about 1958. After 50 years at this rate of consumption, the long lived fission products would have built up to the following approximate levels.

TABLE 3.

Amounts of fission products produced after 50 years by the continuous burn-up of 20 tons of fissile material per year (curies)	Sea water volume to dilute to drinking water tolerance level for large populations. (Cubic miles).
Nuclide	
Zr95 2.9×10^6	1.7×10^4
Ce144 2.4×10^6	1.4×10^4
Ru106 2.3×10^6	55
Pm147 1.2×10^6	30
Sr90 2×10^6	6×10^7
Cs137 1.8×10^6	3×10^4
Tc99 4.6×10^6	0.5
pu 239 6.5×10^4	10^3
Kr85 1.6×10^7	6.3×10^4 cu. miles of air to dilute to breathing tolerance for exposure of large populations.

Estimated total volume of oceans— 5×10^6 cu. miles.

WASTE DISPOSAL

Even for this program, the completion of which is perfectly feasible on the basis of present technology, the dispersal of the Sr90 alone would require 50 million cubic miles of sea water for reduction to drinking water tolerance for large populations. In any case, it is by no means certain that drinking water levels can be applied to sea water, because of the complicated biological chains involved and the probability of caesium and strontium concentration by plankton and by predatory animals, by mechanisms of which we are largely ignorant. The release of large quantities of Kr85 to the atmosphere would also be undesirable because of the possible formation of local high concentrations. It is, therefore, obvious that dilution alone is incapable of solving the waste problem, and that some method of safe permanent storage must be found. Such storage arrangements must be virtually everlasting, because up to 1,000 years would be required for some wastes to decay to harmless levels. It has been suggested by Glueckauf that the problem would be alleviated if the long-lived biologically significant isotopes Cs137 and Sr90 were removed for separate storage, because the remaining nuclides would have decayed enough for disposal by dilution after about 15 years' storage (Glueckauf, 1955a). At this stage it might then be possible to extract the most valuable disintegration products, such as the stable Xe, Ru, Rh, Pd and the Tc, and offset them against the cost of effluent treatment (Glueckauf, 1955b). However, this scheme of treatment ignores the fact that if the activity from Sr90 is reduced by a factor between 100-1,000, further reduction is of little help, because the controlling factor is then the concentration of the long lived alpha-emitters Np237 ($T_{1/2} = 2.2 \times 10^6$ years), Am241 ($T_{1/2} = 470$ years) and Cm242 ($T_{1/2} = 163$ days). The concentrations of these may be expected to rise as higher burn-up is achieved in reactor fuels or reactors using plutonium as fuel are brought into operation (McKay, 1956). Furthermore, even if the most hazardous isotopes could be effectively isolated in solid form from the short-lived nuclides, their fission heat might cause fusion and partial vaporisation unless continual cooling were applied.

GROUND DISPOSAL

As an alternative to dispersal, various schemes of "fully" or "partially" controlled containment have been put forward. Partially controlled containment is the name given in the United States to schemes in which wastes are discharged into the ground. At present this practice is being followed regularly only in the United States, although some ground disposal has taken place at Chalk River. Ground disposal was first used at Hanford to dispose of low level wastes containing up to 50 microcuries/ml. of total beta-gamma radiation, and by 1955 several hundred thousands curies had been disposed of in rock-filled pits.

At Chalk River, contaminated water from the NRX disaster was disposed of to a sandy area adjoining the Ottawa River, as an emergency measure. Subsequently, some 2,000 curies of Sr90 and 100 gm. of plutonium have been de-

posited there, largely from the fuel element trench water. At one time, liquid waste containing plutonium was pumped into a small lagoon in the sand, but it was discovered that the fauna in the area were exhibiting growing levels of acquired radioactivity in their bodies. The practice has now been stopped. At Oak Ridge National Laboratory ground disposal has been tried for "intermediate level" wastes, while low-level laboratory wastes totalling 150 curies/yr diluted in a flow of 700,000 gallons per day have been run for some years into White Oak Lake, an artificial lake formed on the Clinch River, which acts as a settling pond for the ORNL wastes. The results of this operation have not been encouraging for future ground disposals of this type. This has been shown by an ecological study made over three years on White Oak Lake, during which 25,000 specimens of the flora and fauna were analysed and the results compared with others for neighbouring lakes which had not received radioactivity (Krumholtz, 1954). The average level of activity of the water was estimated to be 5×10^{-5} microcuries/ml. and it was computed that some of the fish had received continuous doses of radiation of 50 rads/yr. The growth of all species of fish was found to be slower than those in nearby lakes and the average life span of at least seven species about 25 per cent. lower; two species had become extinct. While there was no positive evidence that this was due to irradiation the circumstantial connection was strong. Every fish examined had selectively accumulated radioactivity, with Cs137 primarily in the soft tissues and Sr90 in bones and skin, in amounts up to 20-30,000 times that in the water. Concentrations of P32 up to 850,000 times over that present in the water were shown by certain algae. Krumholtz (loc. cit.) remarked that the concentration factors for some other elements present only in traces may be even higher. Migratory water fowl appeared to have accumulated an average of 5 micro-curies, mostly P32, in the edible parts of the body over the three years of the survey. Most serious of all, one muskrat had acquired a total body concentration of 100 microcuries of Sr90—about 1 microcurie/gm. of bone. This animal had an advanced bone cancer with secondaries in kidneys and lungs. One species of tree had absorbed enough radio-ruthenium to cause irradiation damage to the leaves.

In spite of these results, which have been obtained with relatively small quantities and may serve as a warning of the difficulties and dangers of disposing of major amounts of active waste, various other schemes for the earth disposal of high level liquids have been suggested in the U.S. Among them are injection into deep wells, or storage in cavities in salt mines and in the old rock domes of discarded oil wells. None of these can, however, really be entertained as a responsible method of solving the problem since control over possible movement of the wastes is lacking. Pending some more permanent method of fully controlled storage, the bulk of high level waste is best held in suitably protected tanks.

TANK STORAGE

For a nuclear power program producing 20 tons of fission products per year, conventional solvent extraction processing with nitric acid as salting-out agent would produce about 200,000 cubic metres of high-level waste annually (Glueckauf, 1955a). After preliminary removal of the bulk of the nitric acid, it is possible to concentrate such a waste to about 1/400th of its original volume before deposition of solid begins. This would mean that an annual provision of about 500 cubic metres of very high-level tank storage capacity would have to be provided. This is a perfectly reasonable amount and the total cost of this operation has been estimated (Glueckauf, 1955a) at around £750,000. Though this is a large amount, it adds very little to the cost of the electricity generated.

The design of tanks suitable for holding such wastes is an extremely difficult and onerous task. One of the difficulties, apart from ensuring complete freedom from leaks and an extremely long life, is that the fission heat liberated is about 125 BTU/gallon/hour for a typical six-month-old fission product waste. This rate of heat evolution is enough to cause bumping and boiling of the contents unless continuous cooling is provided, and trapping devices are necessary to prevent the loss of active spray or vapour. Cooling might have to be applied for periods of 50 years or more. Such tank storage is potentially vulnerable to accidents (for example, earth movements) and to chemical corrosion, and the true life of the tanks under these conditions is unknown. However, it will almost certainly be less than the length of time necessary for the stored activity to decay to harmless levels.

This can, therefore, only be regarded as a temporary expedient, and a search is going on in the U.S.A., Canada and the U.K. for a truly permanent method of storage (Hatch, 1953, 1955; Amphlett, 1956; Evans, 1956; McVay, 1957). The proposal which seems to hold the greatest prospect at the moment is to evaporate the wastes to solid form, and—most promising of all—to reduce them to a solid containing the activity in non-leachable form (Durham & Watson, 1956; Durham, 1957).

FIXATION IN SOLID FORM

Although installation and operating costs are high, owing to the necessity for shielding and for obtaining decontamination factors (*) of the order of 10^6 , partial evaporation has been used for a considerable time to reduce the demand for high level tank storage. Worthwhile further reduction in stored volume could be achieved by conversion of the waste to dry solids, although the engineering difficulties of doing so would be very great. The present-day high level wastes that arise in a process using nitric acid as salting-out agent could be reduced to about 1/700 of their original volume if converted to nitrates, and this could probably be improved to 1/1000 by conversion to oxides. With such a concentration factor, the total yearly demand for storage of 20 tons fission products would be

* Decontamination factor = Activity present before treatment \div Activity present after treatment.

about 200 cubic metres of dry solids. In a typical waste of the type being discussed, the bulk of the solid present would be iron oxide from corrosion products. According to Amphlett (loc. cit.), spinel formation takes place during the conversion to oxide, with fixation of the bulk of the activity against subsequent leaching with water. As a further protection, conversion to a glass has been suggested by Chalk River workers (Durham, Durham & Watson, loc. cit.). The waste would be mixed with suitable proportions of base and flux and fired at temperatures between 1000° and 1500°C. to produce an aluminosilicate glass. Chalk River is examining the use of the mineral nepheline syenite for this purpose, while Amphlett, at A.E.R.E., Harwell, has obtained very satisfactory resistant glasses by using the local green-sand soil with fluxes such as sodium nitrate, sodium carbonate or borax. Although some encouraging results have been obtained in laboratory tests, a great deal of work remains to be done to determine the optimum conditions for the formation of the glass and to measure the leaching rates of the various isotopes when "fixed." In particular, the long-term stability of the glasses to high levels of radiation has still to be determined.

In spite of formidable engineering difficulties, plans for pilot plant-scale tests are going forward at several places in North America. Such a plant must be capable of being run remotely for long periods without maintenance at extremely high levels of alpha, beta, and gamma activity. The most ambitious approach to the problem is that being made at the Reactor Testing Station at Arco, where a combination of spray drying and fluidised-bed roasting is being set up for the conversion to oxides. The intention of the Arco workers is to leach the resultant oxides to remove caesium and strontium in almost pure form for use as radiation sources (Jonke, 1956). However, the plant might be used as a first step in the glass-forming technique.

The density of the resulting glass is such that its volume equals the volume of the original concentrated waste. Our hypothetical output of 20 tons of fission products would thus mean that we should have to store 200 cubic metres of solidified waste per year. Some kind of cooling would certainly be needed to prevent self-melting in the first five years (Coppinger and Tomlinson, 1956). Amphlett has suggested that the glass blocks could be stored in water-filled tanks and the leach liquor processed as low level waste before release.

Unfortunately, the storage situation is much less promising for some more recent types of alloyed fuel elements or for solvent-extraction processes where, for example, aluminium nitrate is used as salting-out agent. Such a process may yield a waste 2.5M in aluminium nitrate, with a fission product concentration of about 1 gm/litre. Conversion of a waste of this kind to oxide provides a meagre 6-10-fold volume reduction. The resulting demand for storage capacity would be serious. It is there-

fore perhaps not too much to suggest that future methods of fuel processing will have to be chosen with one eye on the waste disposal problems involved, which is hardly the case at present.

URGENCY OF THE PROBLEM

In spite of the number of possible methods of disposal that have been discussed, and the thousands of words that have been written about the subject for the past three or four years, the only practical way of handling high-level wastes at present is the method introduced as a temporary measure under wartime pressure, namely tank storage. Brightsen (1957), President of the Nuclear Science and Engineering Corporation, Pittsburgh, has recently observed that at present there are no other solutions to the problem in sight that could be brought into use in a short time. He also points out that the basic problem is one of biological hazard, not of economics, although this fact tends to be lost sight of frequently in the discussions of the subject. Research should therefore be directed towards the goal of lessening the biological danger, with cost a secondary consideration. Greatly increased efforts should also be made to assess the biological hazards of the nuclides involved.

Already, many hundreds of tons of concentrated fission product solutions are in store at various points in the world, representing a very great potential danger to neighbouring populations in the event of accidental release. The U.K. program alone is expected to yield almost two tons of solid fission products by 1965. In spite of the manifest difficulties, it is therefore essential that some practicable method of safe storage should be speedily forthcoming if the development of fission power is not to be seriously hampered.

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