SOME CONSIDERATIONS ON THE MOBILITY OF RADIONUCLIDES*

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1. Introduction

Many toxic substances exist in this world; some are natural and others have been created by man through his activities. Toxic substances can harm living organisms in different ways; but in the end it does not matter if the damage is due to chemical or radiation effects. In fact, radioactive substances can be considered as a special class of toxic material.

It is intuitive that a certain risk results from the existence of toxic substances and that a direct relationship should exist between the quantity of toxic materials and the magnitude of the risk. However, it is also intuitive that many other parameters affect the risk and, in the end, the actual harm will be controlled by the availability of the toxic materials to man. A very simple example will suffice to show that "availability" is really the factor controlling the relationship between the potential harmfulness of substances and the actual health effects incurred by human beings. Alcohol and tobacco are relatively toxic and widely available; every year many thousands of fatalities are caused by their consumption. Chlorine is highly toxic; more than 10^{14} lethal doses of it are used annually by the chemical industry of the US [1], but since chlorine is not generally available the actual harm is very much smaller; as a matter of fact, the only exposure of members

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of the public to chlorine is the result of accidents.

Among the toxic materials used in industry, radioactive substances are probably the least available as a result of the reliable containment systems and the high safety standards of nuclear plants. Only a very minute fraction of the radioactive materials in the nuclear fuel cycle become dispersed into the environment as a consequence of planned or unintentional releases. Radioactive substances differ from other toxic materials since their radioactivity and, in most cases, their radiotoxicity, decrease at a fixed rate as a function of radioactive decay.

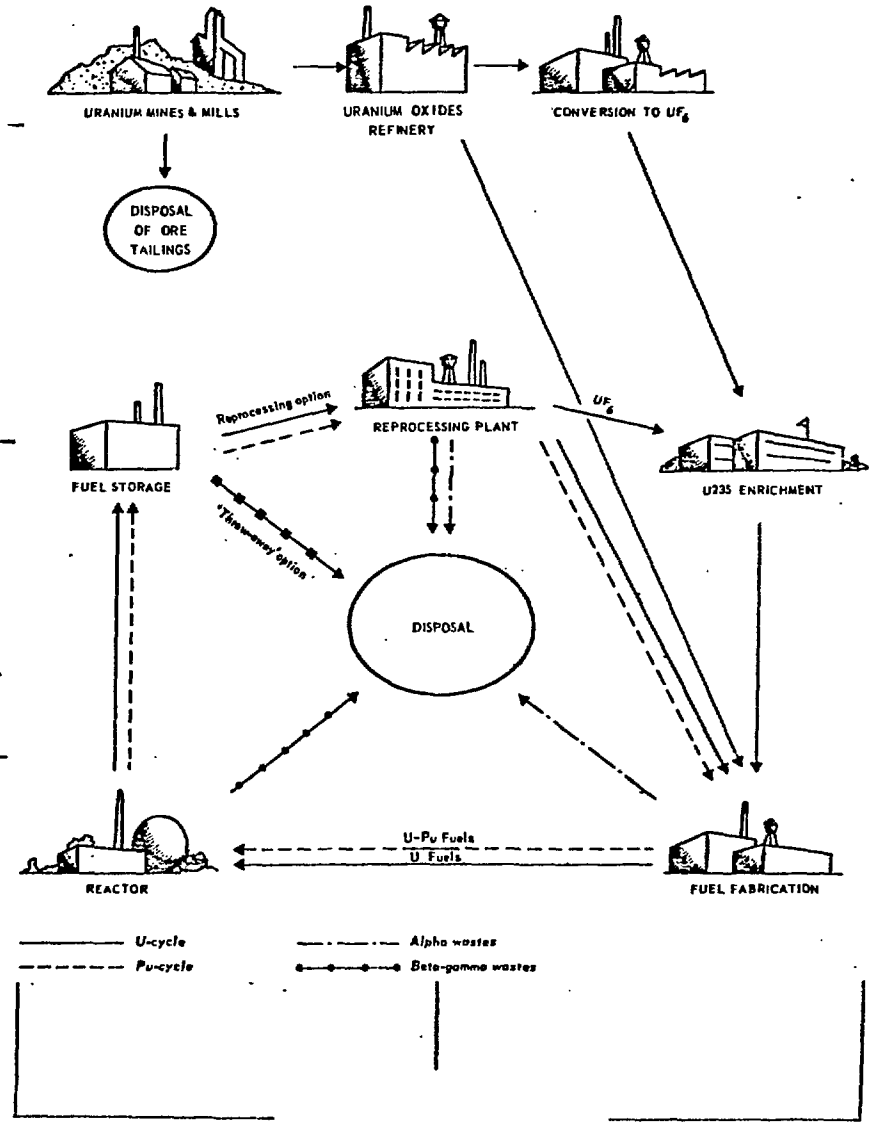
2. The Nuclear Fuel Cycle

Figure 1 shows the main steps in the nuclear fuel cycle. If we consider the uranium cycle, there is no artificial radioactivity before nuclear reactors. During irradiation in reactors, large amounts of radioactive substances are produced; consequently, radionuclide inventories in reactors, spent fuel storage facilities, reprocessing plants and waste storage and disposal facilities, are very large.

All plants in the nuclear fuel cycle are designed for maximum containment of radioactive materials; nevertheless, very small amounts of radionuclides are discharged to the environment, particularly from reprocessing plants, reactors, and uranium mining and milling facilities. The exposure of members of the public is usually well below the natural background and within the dose limits established by the competent regulatory authorities. The actual doses are a function of the magnitude of the discharges, of the population distribution and of the transfer along environmental pathways. The greatest part of radioactive materials is safely contained within the plants and is not available as a source of public exposure. The estimated probabilities of serious accidents which might lead to the dispersal of significant amounts of radionuclides in the environment, are extremely small. According to the Rasmussen

Figure 1

MAJOR STEPS IN THE NUCLEAR FUEL CYCLE



report, the frequency of short term fatalities resulting from the existence of 100 nuclear power reactors is comparable to the frequency of fatalities due to meteorite falls [2]. In the unlikely event of accidental release of radionuclides, the resulting doses would be controlled by the transfer to man along the environmental pathways.

By far the greatest part of radioactive substances in the nuclear fuel cycle will end up in radioactive wastes, either in solid or liquid form; the liquid wastes will eventually be solidified. The only viable alternative for the disposal of the bulk of radioactive solid waste is emplacement in suitable geological formations either on land or under the seabed. The only exception being the relatively minor amounts of low- and medium-level solid waste that can be disposed of by sea dumping.

The availability to man of radioactive nuclides after geologic disposal is the lowest in the entire nuclear fuel cycle, provided the disposal formations have been selected with care and the connections with the biosphere have been effectively plugged. The main questions refer to the long half-lives of some waste radionuclides and therefore to the unusual length of time over which long-lived wastes need to be contained. However, after disposal, several barriers are interposed between the waste and human beings, namely: (1) the waste form; (2) the waste containers; (3) the disposal formation; and (4) the retention mechanisms along the pathways from the disposal formation to the surface. Only after emergence from underground would the radionuclides become possible sources of exposure, always depending on their mobility along the environmental pathways.

In conclusion, environmental mobility controls the doses resulting from the entry of radionuclides into the environment; while mobility in the geosphere controls the amounts of radionuclides that can reach the biosphere from waste repositories after failure of the first three barriers.

3. Mobility in the Environment

A large amount of data is now available about the environmental behaviour of most significant radionuclides in the nuclear fuel cycle. The information is obtained from experiments, monitoring around nuclear plants, and fallout and geochemical data. Mathematical models have been developed with the capability of predicting doses resulting from the presence in the environment of any mixture of radionuclides [3-5]. The validity of the models can be verified by comparing calculations and observations.

Perhaps the best information on the behaviour of radionuclides in the global environment can be obtained by the fallout data, which have been collected and elaborated by a number of organisations, among which the Health and Safety Laboratory of the US Department of Energy and the United Nations Scientific Committee on the Effects of Atomic Radiation, deserve special mention. By means of fallout data it is possible to relate deposition levels with intakes by ingestion and inhalation. This information combined with the observation of radionuclides concentrations in the main environmental compartments enables the drawing of convincing inferences about the mobility of radionuclides in the environment at large [6, 7]. It may be interesting to review briefly the fallout data for two significant radionuclides.

Strontium-90

The total deposit of Sr-90 fallout on the earth's surface through 1970 and 1975 is estimated to be 12.2 and 11.7 MCi, respectively [6, 8]. Cumulative deposition in mid-latutude areas of the northern hemisphere through 1970 was in the range of 60 to 80 mCi/km^2 [6]. The resulting annual intakes through the diet in New York and San Francisco during 1975 were 3010 and 1190 pCi, respectively. Dairy products and vegetables accounted for 60 to 70% of the total intake [9].

Plutonium-239/240

The total deposit of Pu-239/240 on earth through 1970 was approximately 330 kCi. Cumulative deposition in the temperate regions of the northerm hemisphere through 1970 was in the range of 2 to 2.5 mCi/km^2 [10, 11]. The dietary intake in New York during the 1972-1974 period has been 1.6 ± 0.3 pCi/y. Bakery products, meat, fresh fruit and vegetables, and tap water were the most significant contributors to the intake; however, shellfish, with a mean concentration of 0.012 pCi/kg, was the food with the highest Pu content. During the same period, inhalation intake of Pu-239/240 averaged 0.2 pCi/y. However, since the uptake from the gastro-intestinal tract is extremely low and most ingested plutonium is excreted, it is calculated that the inhalation pathway contributed to body burden about 1000 times more than the ingestion pathway.

Consideration of the relative transfer from soil to man through food pathways can provide some indication of the relative environmental mobility of the two radionuclides. Pu-239/240 deposition in temperate regions is about 30 times less than Sr-90 deposition, but the dietary intake is 1500 to 2000 times less, confirming therefore the expected greater biological availability of Sr-90.

Table I summarizes the dose commitments from nuclear tests carried out before 1971, as reported by UNSCEAR [6].

4. Mobility in the Geosphere

In comparison with the environmental behaviour of radionuclides, much less is known about their mobility in the geosphere. It is difficult to generalise available information due to the great variability of geologic conditions and the long periods of time over which radionuclide migration could take place. However, data are being accumulated for a number of geologic media and environments.

TABLE I. DOSE COMMITMENTS FROM NUCLEAR TESTS CARRIED OUT BEFORE 1971 [6]

Source of radiation	Dose commitments (mrad) for the north temperate zone			Dose commitments (mrad) for the south temperate zone			Dose commitments (mrad) to the world population		
	Gonads	Bone lining cells	Bone marrow	Gonads	Bone lining cells	Bone marrow	Gonads	Bone lining cells	Bone marrow
External									
Short-lived	65	65	65	19	19	19	44	44	44
Cs-137	59	59	59	16	16	16	40	40	40
Kr-85	$2\ 10^{-4}$	$2\ 10^{-4}$	$2\ 10^{-4}$	$2\ 10^{-4}$	$2\ 10^{-4}$	$2\ 10^{-4}$	$2\ 10^{-4}$	$2\ 10^{-4}$	$2\ 10^{-4}$
Internal									
H-3	4	4	4	1	1	1	4	4	4
C-14(a)	12	15	12	12	15	12	12	15	12
Fe-55	1	1	0.6	0.3	0.3	0.2	0.7	0.7	0.4
Sr-90		85	62		23	17		57	42
Cs-137	26	26	26	7	7	7	18	18	18
Pu-239(b)		0.2			0.05			0.1	
Total(c)	170	260	230	55	81	72	120	180	160

(a) Dose accumulated up to year 2000. The total dose commitment to the gonads and bone marrow is about 140 mrad; it is about 170 mrad to cells lining bone surfaces.

(b) The dose commitment to bone lining cells for the north temperate zone has been taken to be equal to the integrated dose over 50 years to bone. A reduction by a factor of four has been assumed for the south temperate zone. Because of insufficient data, the dose commitments to gonads and to bone marrow have not been estimated.

(c) Totals have been rounded off to two significant figures.

All types of radioactive wastes can be disposed of underground. Low-level wastes have been buried in trenches in several locations in a variety of geologic media; as a result, some observations on radionuclides migration at shallow depths are available [13, 14].

High-level and alpha-bearing wastes will be emplaced at greater depth. The geological formations under consideration for the disposal of long-lived wastes include rock salt, argillaceous sediments and crystalline rocks.

In theory, several processes can cause the migration of radionuclides in geological formations, but it is generally agreed that leaching and transport by ground water is the only mechanism that could seriously affect the isolation of radioactive waste. Salt formations are normally free from circulating ground water. Argillaceous sediments are characterised by low permeabilities, but they contain some water, which normally moves as a function of the existing hydraulic gradient. Crystalline rocks are usually fractured; however, at depth, in tectonically stable areas, the fractures are closed by the overburden pressure and by secondary minerals, resulting in very low permeabilities. Ground water moves along the system of fractures, according to the hydraulic gradient. Therefore, in salt, no migration is possible until ground water reaches the waste and leaching takes place. In argillaceous and crystalline rocks, some ground water is usually present and leaching and migration should start as soon as water comes into contact with the waste. An additional difference between rock salt and the other potential disposal media is that very little retardation of radionuclides as a result of sorption processes would take place in the former, while many nuclides would be effectively retarded in crystalline rocks and especially in argillaceous formations.

In many cases the radionuclides would have to migrate through additional geological formations, probably over significant distances, before they could emerge at the surface.

Retardation along these geosphere pathways can contribute to the overall waste containment system.

The geologic conditions of potential disposal formations can vary drastically, to the point that a meaningful evaluation of the potential migration of radionuclides from waste repositories must be site specific. However, the information on underground migration of nuclides being generated from laboratory and field experiments, past waste disposal practices, and geochemical studies, is most useful since it broadens the data basis and will permit more reliable safety analyses when repository sites are chosen. The following list of observations and studies include the main sources of information on the migration of radionuclides in geologic media. Some references are provided as a small sample of the many publications existing in this field [15-21].

- Laboratory determinations of distribution coefficients (Kd) and retardation factors.

- Radionuclides distributions as a result of liquid waste disposal practices at Hanford, Washington, and at the Idaho Nuclear Engineering Laboratory, Idaho.

- Observations of radionuclides distributions at the Nevada Test Site around nuclear explosion cavities.

- Geochemical studies of the fate of fission products and actinides at the site of the natural fission reactors in Oklo, Gabon.

- Geochemical studies of distributions of specific elements around mineral deposits.

Several mathematical models and computer programs, which describe the underground migration of radionuclides, have been produced [22-27]. The theory seems to be well understood and

some models are quite sophisticated. In general, the limiting factor on the reliability of results of radionuclide transport calculations is not the model being used, but the quality of the geologic input data. It is obvious that some geologic input data are site specific and will have to be collected for particular repository sites, but more studies aimed at a better understanding of the physico-chemical reactions controlling sorption-desorption processes and of possible effects of the repository thermal loading on ground water flow are needed.

Due to their recent artificial origin and, in consequence, to the scarcity of geochemical evidence, transuranium elements are particularly unknown from the point of view of their migration in the geosphere. Nevertheless, some preliminary results are available and they indicate that geologic barriers should be quite effective. As a matter of fact, it appears that only nuclides with long half-lives and with unity retardation coefficient (no sorption) such as Tc-99 and I-129 could emerge at the surface with unacceptable concentration levels after 5 km transport in shale, granite or basalt [26]. If such a conclusion were confirmed by experimental observations, it would indicate that, for repositories in wet geological formations, long-lived radionuclides that migrate at the same rate as ground water, such as Tc-99 and I-129, might have to be removed from the waste and disposed of elsewhere. For I-129, a possible alternative could be seabed disposal, while Tc-99 might be incorporated in geochemically stable synthetic minerals and placed either under the seabed or in salt formations.

5. Conclusions

As for all toxic materials, availability to man of radionuclides is the key factor which controls the potential health effects. Radioactive materials are contained with great care and are not generally available to members of the public. However, they could become dispersed into the environment as a result of accidents both at nuclear plants and at radioactive

waste repositories. In the event of accidents, the mobility of radionuclides along environmental pathways and in the geosphere would control the exposure of individuals and populations. Some of the most hazardous alpha emitters generated by the nuclear industry, such as Pu and Am isotopes, are characterised by low mobility along environmental pathways. In relation to their potential migration in the geosphere, most radionuclides are characterised by low mobility as a result of the interactions with geologic materials. The only waste products for which geologic barriers would be relatively ineffective are long-lived nuclides which do not interact with geological media, such as Tc-99 and I-129. The radiotoxicity of these two nuclides is rather low. However, it is possible that they might have to be managed separately from the rest of radioactive waste products.

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