

# RADIOACTIVE WASTE AND NUCLEAR POWER: SOURCES

The current worldwide expansion in the use of nuclear power for the generation of electricity has stimulated a renewed interest in the problems of radioactive waste management. It is generally accepted that at present nuclear fission provides the only viable alternative energy source for the early part of the coming century as world reserves of fossil fuels are gradually depleted (Yannacone 1974). However, if the full potential of nuclear power to provide a supply of energy which is limited only by investment capabilities is to be realized, techniques must be developed to cope with the radioactive wastes which arise at each stage of the nuclear fuel cycle. Since some components of these waste materials represent a hazard to man for many thousands of years or more, the problem of their secure containment presents scientists and engineers with a unique technological challenge.

The fundamental principles of the management of radioactive wastes within the United Kingdom were established by the Radioactive Substances Advisory Committee (1959) in a White Paper entitled *The Control of Radioactive Wastes*, and may be summarized as follows:

- (a) To ensure irrespective of cost, that no member of the public shall receive a radiation dose in excess of the limits recommended by the ICRP (International Commission on Radiological Protection);
- (b) To ensure, irrespective of cost, that the whole population of the country shall not receive an average dose of more than 1 R<sup>†</sup> per person in thirty years; and
- (c) To do what is reasonably practicable, having regard to cost, convenience and national importance of this subject, to reduce the doses far below these levels.

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In the first of two articles on radioactive wastes from nuclear power we look at the nature and origins of waste from present and future installations

These principles are implemented through statutory instruments embodied in the 1960 Radioactive Substances Act, and subsequent legislation, and are administered by the Department of the Environment (Radiochemical Inspectorate) and by the Ministry of Agriculture, Fisheries and Food via 'Authorizations' which limit the discharge of radioactive wastes to the

<sup>†</sup> An exposure of 1 R (roentgen) is approximately equivalent to a dose in tissue of 1 rad or  $10^{-2}$  J kg<sup>-1</sup>

environment. In addition, the accumulation of such wastes at individual locations is controlled under the terms of 'Nuclear Site Licences' administered by the Health and Safety Executive (Nuclear Installations Inspectorate).

Here we consider the nature and origins of the active wastes produced in each phase of the nuclear fuel cycle. The dominant sources of waste are associated with power reactor irradiation and the subsequent fuel reprocessing operations. Various categories of nuclear waste can be identified from their method of production and potential hazard to man; this article examines the characteristics of waste arising from a range of thermal and fast reactor designs and the problems likely to arise in the disposal of obsolete nuclear plant. Alternative methods of exploiting nuclear energy, other than current uranium or plutonium based fuel cycles will also be considered.

### The nuclear fuel cycle

The nuclear fuel cycle comprises the full range of operations associated with the generation of electricity from nuclear power and is illustrated schematically in figure 1. It commences with the mining and milling of the naturally occurring uranium (or thorium) bearing ores, prior to their concentration and refining for dispatch to the fuel fabrication plant. This forms the next stage of the cycle, in which uranium metal or  $UO_2$  fuels are manufactured from the ore concentrates and encased within appropriate cladding materials to form the reactor fuel elements. In the case of advanced thermal reactor fuels isotopic enrichment of the fissile  $^{235}U$  isotope, which has a natural abundance of only 0.71%, is also carried out at this stage.† Next comes the reactor irradiation and subsequent transport of the irradiated fuel to the reprocessing plant. This latter stage completes the nuclear cycle with the extraction of the unused uranium, and also the fissile plutonium produced during the reactor irradiation. These materials are then available for the fabrication of new fuels and recycle through advanced thermal or fast breeder reactor systems.

Radioactive wastes are produced at each stage of the nuclear cycle, beginning with the mining and milling operations. In the mining of uranium ores quantities of natural radioactivity comprising radon gas and its daughter products are released, and in underground installations these make it necessary to ventilate the mines. The contaminated ventilation air is rapidly diluted in the atmosphere where its  $^{222}Rn$  content is detectable

† Although most reactor types employ enriched fuel, a substantial fraction of the world's operating power reactors employ natural uranium fuel, for example the Magnox reactors in the UK, Italy and Japan, and Candu reactors in Canada

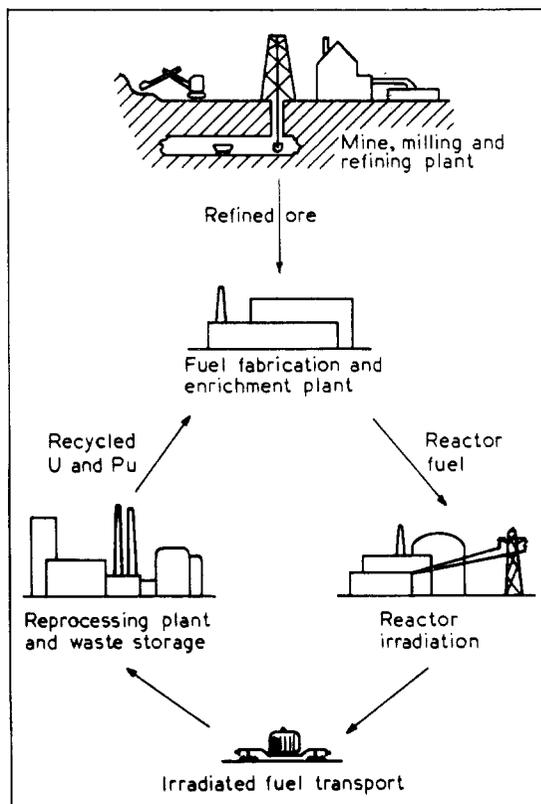
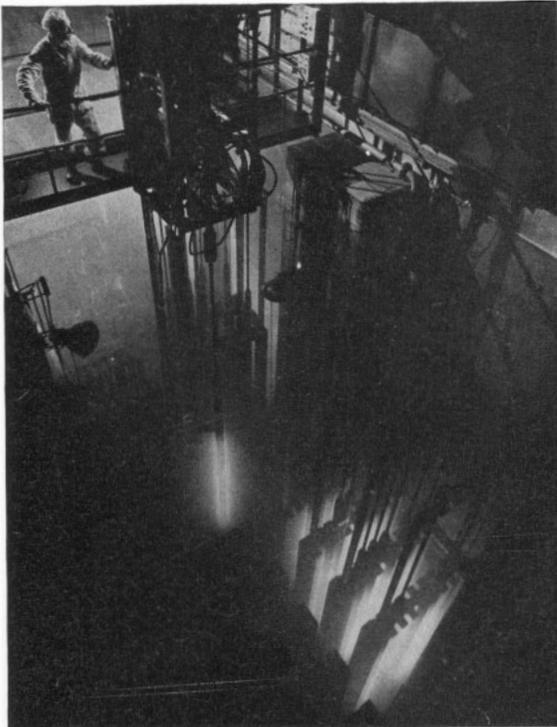


Figure 1 The nuclear fuel cycle

above normal radon levels only in the immediate vicinity of the mine.

Further radioactive wastes, principally the radium isotope  $^{226}Ra$ , are produced in the milling of the ore which concentrates the uranium from a naturally occurring level of 0.1–1% up to about 90% by weight. After crushing to reduce the size of the mined ore, the uranium is extracted by acid or alkali leaching, combined with chemical precipitation, ion exchange or solvent extraction. These processes result in solid and liquid wastes, known as tailings, which contain the naturally occurring active radium and are initially pumped into settling ponds. Apart from the radioactive content of the tailings, care also has to be exercised in the control of chemically toxic agents present, notably traces of uranium and other heavy metals such as zinc, copper and lead. Although the main radionuclide of interest,  $^{226}Ra$ , does not represent a significant radiological hazard, the residual solids ultimately require stabilization to minimize their long term dispersal in the environment (Eisenbud 1963).

In the fuel fabrication phase of the nuclear fuel cycle the main hazard arises from the chemical toxicity of uranium, the radiotoxicity of natural uranium being of secondary importance. Small quantities of uranium exist in the liquid



*Irradiated fuel in the storage pond of the Winfrith SGHWR. The glow that can be seen is Cerenkov radiation. (Photograph courtesy of UKAEA)*

wastes from fuel fabrication plants, while some processes involve potential exposure of operators to alpha-emitting dusts. However, the commercial value of uranium ensures that all operations are carried out with only minimal losses, and the amounts present in waste discharges are not normally detectable above the natural background levels (Hill 1973).

All of the activities considered so far are naturally occurring and in general their levels in the waste materials produced in the manufacture of reactor fuels do not present any major problems. Indeed, it is only in the reactor irradiation and subsequent phases of the nuclear fuel cycle that significant quantities of radioactive waste materials are produced. This situation is slightly modified in systems involving the recycle of recovered uranium and plutonium isotopes, and these will be discussed later where appropriate. In the following sections the emphasis is placed upon the artificial radionuclides generated during power reactor operations, many of which appear as the main contaminants in fuel reprocessing wastes. Others are retained within the reactor pressure vessel, or in shielded waste storage facilities on the site (Myatt 1974), and only constitute a waste disposal problem when the reactor is finally dismantled at the end of its life. (At least some degree of dis-

mantling will take place, and the CEBG has given public undertakings that reactor sites could be returned to former use if required, although it is more likely that the sites would be reused for future CEBG developments.) In addition, small quantities of radioactivity arise in the form of low-level contamination of gaseous and liquid effluents which are routinely discharged from nuclear installations under the terms of their nuclear site licences. These effluents are associated with normal operational and maintenance activities but the quantities of radioactivity involved are negligible compared with the high-level reprocessing and reactor wastes mentioned above.

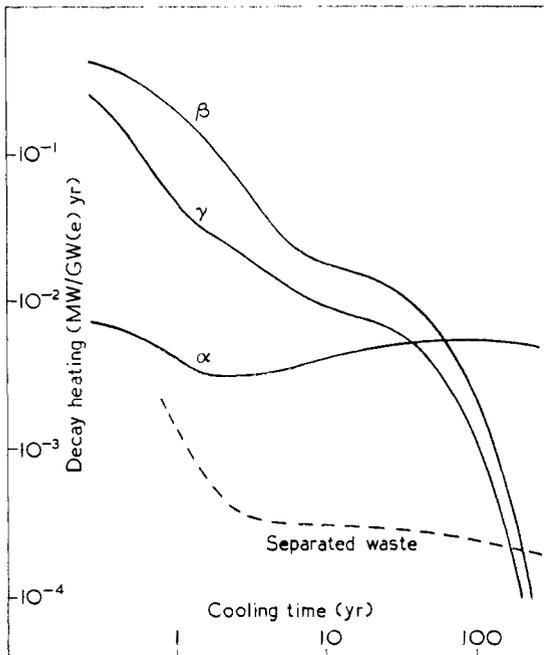
### **The origins and nature of nuclear wastes**

During irradiation of fuel within the reactor several hundred radionuclides are generated in the fission process, while others are produced via neutron activation of the fuel and also reactor structural materials. These isotopes have half-lives ranging from a fraction of a second up to millions of years, and in undergoing radioactive decay may emit alpha, beta or gamma radiations at energies up to a few MeV per disintegration. The aim of waste management activities is to contain these materials and limit their release to the environment in order to minimize the hazards to man from their transfer via air and water or through various food chains. In considering waste management problems it is convenient to identify in turn three main classes of radionuclides, namely fission products, actinide isotopes and activation products.

#### **FISSION PRODUCTS**

A highly readable account of the neutron physics of the fission process was presented in an earlier article in this magazine on the evolution of nuclear fuels (Boltz 1973), and only a brief outline will be given here. Fission product isotopes, together with the neutrons which sustain the chain reaction within a reactor core, are the products of interactions involving a neutron and a fissile heavy metal atom and their mass distribution follows the well known double-humped fission-yield curve, peaking at mass numbers in the range 85–110 and 130–150. The primary products of the fission reaction are normally short lived and rapidly decay via beta and gamma emissions through one or more longer lived isotopes to a stable species. Frequently the radionuclides of greatest radiological significance are the fourth or fifth members of one of these decay chains, as in the case of  $^{90}\text{Sr}$  and  $^{131}\text{I}$ , which occur in the decay schemes at the foot of the next page.

The build-up of activity within the fuel during a reactor irradiation and its decay following discharge of the fuel from the reactor, are thus determined by the combined effects of several tens of chains of the type illustrated above. On discharge the fuel is stored at the reactor for up



**Figure 2**  $\alpha$ ,  $\beta$  and  $\gamma$  decay curves for natural uranium Magnox reactor fuel irradiated to  $3500 \text{ MW d t}^{-1}$  at  $2.7 \text{ MW t}^{-1}$

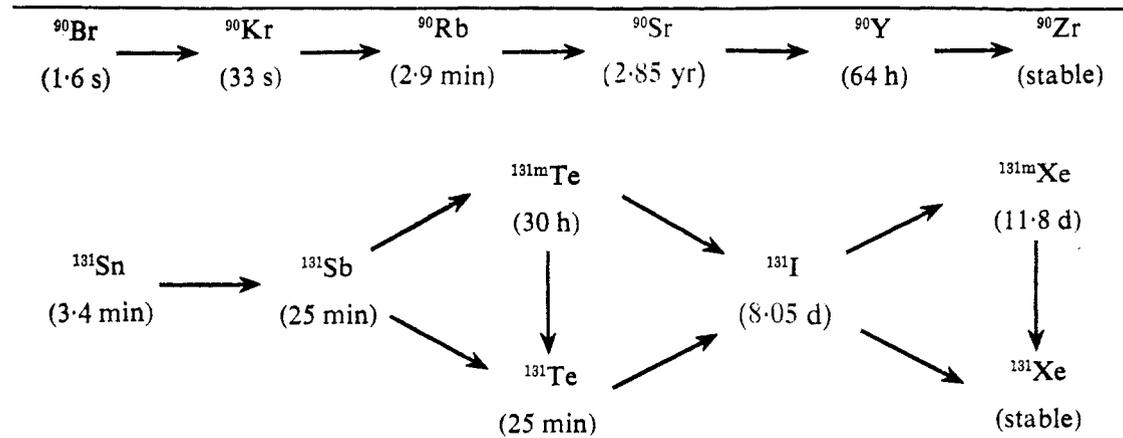
to three months, prior to transport to the reprocessing plant. This greatly reduces its radioactive emissions by allowing the decay of the majority of the shorter-lived isotopes, particularly those of the noble gases krypton and xenon, and also the volatile element iodine. The chemical reprocessing of the fuel normally takes place within a year of discharge from the reactor, when the remaining fission product isotopes are separated from the unused uranium and the plutonium isotopes produced during the reactor irradiation. Thus only those isotopes having halflives of a few hundred days or more make a significant contribution to the activity of the wastes

associated with the fuel processing.

The decay of the activity of a typical thermal reactor fuel is illustrated in figure 2, with the decay of the fission product isotopes being characterized by the beta and gamma curves. The former curve follows the decays of  $^{144}\text{Ce}/^{144}\text{Pr}$  up to about four years cooling time, and thereafter the decay of the bone-seeking  $^{90}\text{Sr}$  isotope. The gamma decay curve is more complex, having a large number of contributing isotopes during the first few years following fuel discharge, but at longer cooling times it is dominated by the decays of  $^{137}\text{Cs}/^{137\text{m}}\text{Ba}$ . The beta and gamma decay curves show that the fission product activities in reprocessing wastes fall by almost two orders of magnitude over a period of 100 years, and beyond this time continue to fall with a halflife of about 30 years, determined by the  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  decays.

The ultimate fission product activity of reprocessing wastes arises from a few very long lived nuclides, such as  $^{129}\text{I}$  with a radioactive halflife of  $1.6 \times 10^7$  years, but their activities are many orders less than that of the more hazardous nuclides mentioned above. An assessment of the relative hazards of the more active fission products is presented in table 1, in which the isotopes are listed in groups of similar halflife. The hazards, expressed as the volume of air or water in which the activity produced per unit generation ( $\text{m}^3/\text{GW(e)year}$ ) must be diluted to yield the maximum permitted concentrations for members of the public recommended by the ICRP (1960, 1964), are largely independent of reactor type, apart from minor variations associated with differences in fission yields of the various heavy metal isotopes. For example, the  $^{90}\text{Sr}$  hazard in a  $^{235}\text{U}$  fuelled thermal reactor is 2-3 times greater than that for a plutonium-fuelled fast breeder reactor, while in the case of  $^{106}\text{Ru}$  and the rare earth nuclides the fast-reactor hazards are greater by a similar factor. However, beyond the first few years after discharge of the fuel from the reactor the dominant fission product hazard is

#### Decay products from fission reactions



that associated with  $^{90}\text{Sr}$ , in terms of potential leakage to both the atmosphere and the aqueous environment.

#### ACTINIDE ISOTOPES

Apart from initiating fission reactions, the neutrons generated within a reactor core may interact with a heavy metal atom to produce another heavy metal nuclide. Such capture reactions are accompanied by the emission of gamma radiation or further neutrons, while the resultant heavy metal nuclide may undergo further neutron captures, or decay with the emission of alpha, beta or gamma radiations. Thus, during a power reactor irradiation successive neutron capture and decay processes result in the build up of a wide range of actinide isotopes, in addition to the fission products.

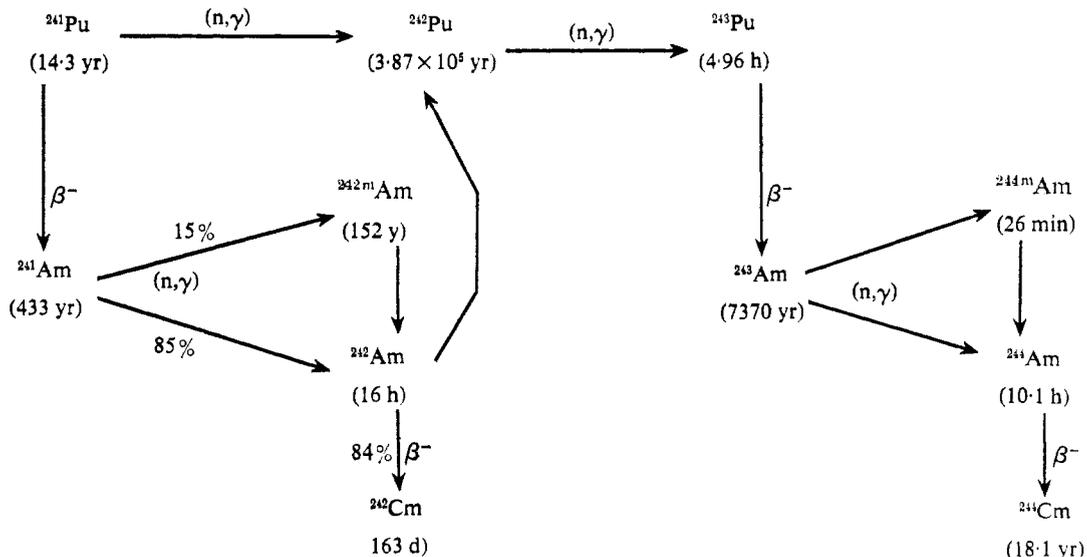
In the case of uranium-based reactor systems a broad spectrum of transuranic nuclides is present in the discharged fuel, the most important being the fissile isotopes of plutonium which are recovered during reprocessing for use in the fabrication of advanced thermal or fast breeder reactor fuels. Also present are higher transuranics, particularly the isotopes of americium and curium, which constitute the dominant source of alpha activity and decay heating in the reprocessing wastes following uranium and plutonium separation. The relative contributions of the fission product and actinide isotopes to reprocessing wastes are shown in figure 2, in which the total and separated-waste alpha-decay curves are plotted together with the associated fission product beta and gamma curves. At cooling times up to about two years the alpha decays are dominated by  $^{242}\text{Cm}$ , followed in the unseparated

wastes by a slow build up due to growth of the alpha emitter  $^{241}\text{Am}$  from beta decays of  $^{241}\text{Pu}$ . In the separated wastes remaining after uranium and plutonium recovery the main contributors to the alpha curves following the decay of  $^{242}\text{Cm}$  are its daughter nuclide  $^{238}\text{Pu}$ , together with the  $^{241}\text{Am}$  present in the fuel at the time of separation, which in the case of the data presented in figure 2 has been taken as 300 days post discharge.

Unlike the fission product activities and hazards per unit generation of electricity, the equivalent parameters for the actinide isotopes show a marked dependence on reactor type. On progressing from low burn-up uranium-fuelled thermal reactors to more advanced thermal or plutonium-fuelled fast breeder reactor designs, the main effect is an increased production of the higher transplutonium nuclides, notably  $^{244}\text{Cm}$  and  $^{243}\text{Am}$ , which manifests itself as a net increase in the alpha emissions relative to the fission product beta and gamma decays. This effect is associated largely with the higher fuel burn-up in these systems, but other factors such as the degree of neutron moderation and fuel enrichment can play an important role (Clarke *et al* 1975). The build-up of the curium and americium isotopes responsible for the alpha decays in reprocessing wastes occurs via two virtually independent routes, one controlled by the  $^{241}\text{Pu}$  activity within the fuel and the other by neutron captures in  $^{242}\text{Pu}$  as shown below.

As an example of the variability of actinide production, the build-up of  $^{242}\text{Cm}$  in a range of typical thermal and fast-reactor fuels is shown in figure 3. This clearly demonstrates the increased curium production in the higher burn-up thermal

#### Routes to the build-up of Cu and Am isotopes



reactor systems, as well as illustrating the dependence of the  $^{242}\text{Cm}$  build-up on the  $^{241}\text{Pu}$  content of the fuel through the characteristic shape of the fast-reactor curves. These show an enhanced curium production during the early stages of the fuel cycle, due to the small  $^{241}\text{Pu}$  content of the unirradiated plutonium fuel, in contrast to the uranium fuelled thermal systems where significant build-up of the plutonium isotopes occurs only in the later stages of the cycle. In addition, this effect is more marked for the long shelf-life fast-reactor fuel due to the formation of  $^{241}\text{Am}$  from decay of  $^{241}\text{Pu}$  during storage of the fabricated fuel.

Finally, activity and hazard values for the more important actinide isotopes present in Magnox reactor fuel are also given in table 1. In the un-separated fuel the actinide hazards are dominated by the isotopes of plutonium, but following the recovery of these during reprocessing the dominant hazards associated with the wastes arise from  $^{241}\text{Am}$  and  $^{242}\text{Cm}$ , or at longer cooling times the latter's  $^{238}\text{Pu}$  daughter. The data presented in table 1 show that the actinide hazards derived on

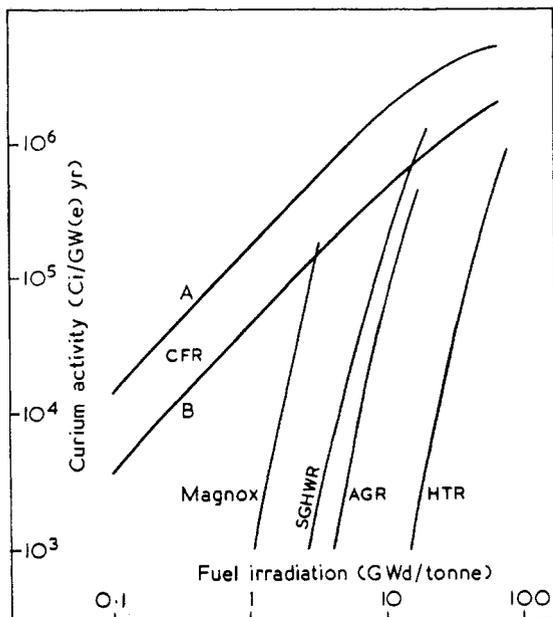
the basis of uptake via inhalation outweigh those associated with the fission product isotopes, particularly at longer cooling times. However, in terms of accidental discharges to the aqueous environment,  $^{90}\text{Sr}$  would dominate the hazard index for the first few decades following reprocessing, this period decreasing with the enhanced production of the higher transplutonium isotopes in advanced thermal and fast breeder reactors discussed above.

#### ACTIVATION PRODUCTS

The third class of radionuclides generated within power reactors, activation products, arise from interactions between neutrons and atoms within the coolant, moderator or structural materials. They are thus the products of capture reactions, but unlike the actinides do not in general form complex capture and decay schemes, decaying instead directly to stable species with the emission of beta and gamma radiations. Activation product inventories are strongly dependent on reactor type, even on detailed design features of individual reactors of a given type, and they may originate from trace impurities having large

**Table 1** Fission product, actinide and activation product activities and hazards for thermal power reactors. (a) Indicates values based on mean burn-up fuel irradiated to  $3500 \text{ MW d t}^{-1}$  at  $2.7 \text{ MW t}^{-1}$  and cooled for 100 d, (b) indicates representative values based on gas or water cooled reactors (see text)

	Nuclide	Half-life	Activity (Ci/GW(e)yr)	Hazard index ( $\text{m}^3/\text{GW}(\text{e})\text{yr}$ )	
				Water	Air
Fission product isotopes <sup>(a)</sup>	$^{91}\text{Y}$	58 d	$10^7$	$3.3 \times 10^{11}$	$10^{16}$
	$^{95}\text{Zr}$	64 d	$1.5 \times 10^7$	$2.5 \times 10^{11}$	$1.5 \times 10^{16}$
	$^{131}\text{I}$	8.05 d	$4.4 \times 10^3$	$2.2 \times 10^9$	$1.5 \times 10^{13}$
	$^{106}\text{Ru}$	369 d	$10^7$	$10^{12}$	$5 \times 10^{16}$
	$^{134}\text{Cs}$	2.1 yr	$1.1 \times 10^6$	$1.2 \times 10^{11}$	$2.8 \times 10^{15}$
	$^{144}\text{Ce}$	284 d	$2.8 \times 10^7$	$2.8 \times 10^{12}$	$1.4 \times 10^{17}$
	$^{147}\text{Pm}$	2.6 yr	$7.8 \times 10^6$	$3.9 \times 10^{10}$	$3.9 \times 10^{15}$
	$^{90}\text{Sr}$	28.5 yr	$2.9 \times 10^6$	$7.3 \times 10^{12}$	$7.3 \times 10^{16}$
	$^{129}\text{I}$	$1.6 \times 10^7$ yr	1.4	$3.5 \times 10^6$	$2.3 \times 10^{10}$
	$^{137}\text{Cs}$	30.1 yr	$3.7 \times 10^6$	$1.9 \times 10^{11}$	$7.4 \times 10^{15}$
	$^{154}\text{Eu}$	16 yr	$6 \times 10^4$	$3 \times 10^9$	$6 \times 10^{14}$
	$^{85}\text{Kr}$	10.73 yr	$3.4 \times 10^5$	—	$1.1 \times 10^{12}$
	$^3\text{H}$	12.3 yr	$3 \times 10^4$	$10^7$	$1.5 \times 10^{11}$
Actinide Isotopes <sup>(a)</sup>	$^{238}\text{Pu}$	86.4 yr	$9.4 \times 10^3$	$1.9 \times 10^9$	$1.3 \times 10^{17}$
	$^{239}\text{Pu}$	$2.4 \times 10^4$ yr	$3.7 \times 10^4$	$7.4 \times 10^9$	$6.1 \times 10^{17}$
	$^{240}\text{Pu}$	6580 yr	$3.2 \times 10^4$	$6.4 \times 10^9$	$5.3 \times 10^{17}$
	$^{241}\text{Pu}$	14.3 yr	$2.8 \times 10^6$	$1.4 \times 10^{10}$	$9.3 \times 10^{17}$
	$^{241}\text{Am}$	433 yr	$5.8 \times 10^3$	$1.5 \times 10^9$	$2.9 \times 10^{16}$
	$^{242}\text{Cm}$	163 d	$1.2 \times 10^5$	$6 \times 10^9$	$3 \times 10^{16}$
	$^{244}\text{Cm}$	18.1 yr	$1.7 \times 10^2$	$2.4 \times 10^7$	$5.7 \times 10^{14}$
Activation products <sup>(b)</sup>	$^3\text{H}$	12.3 yr	$10\text{--}10^5$	$3 \times 10^3\text{--}$ $3 \times 10^7$	$5 \times 10^7\text{--}$ $5 \times 10^{11}$
	$^{14}\text{C}$	5760 yr	50	$6 \times 10^4$	$5 \times 10^8$
	$^{41}\text{Ar}$	1.83 h	$10^5$	—	$2.5 \times 10^{12}$
	$^{60}\text{Co}$	5.3 yr	$10^5\text{--}10^6$	$3 \times 10^9\text{--}$ $3 \times 10^{10}$	$3 \times 10^{14}\text{--}$ $3 \times 10^{15}$



**Figure 3** Curium-242 build up in thermal and fast breeder reactors. A—fuel stored for 4 yr, B—fuel stored for 1 yr

neutron absorption cross sections, as well as from activation of bulk materials in and around the reactor core which are subject to neutron irradiation. Thus, while it is difficult to generalize about the activation product contributions to nuclear wastes, it is possible to discuss a few representative examples characteristic of a range of reactor types.

One of the most important activation products arising in nuclear power reactors is the 5.3 year half-life isotope  $^{60}\text{Co}$ , formed by thermal neutron captures in  $^{59}\text{Co}$  or fast neutron captures in  $^{60}\text{Ni}$ . These reactions occur in the various steels used as fuel cladding materials, in components of fuel assemblies and also in core support structures. For example, in the advanced gas cooled reactor (AGR) one of the major sources of  $^{60}\text{Co}$  is the activation of trace impurities in the stainless steel fuel pin clad, while for Magnox reactors it occurs in the cobalt-rich springs used to locate the magnesium-aluminium alloy clad fuel elements within the fuel channels. In the former case the cobalt activity is returned with the fuel for re-processing and thus appears in the wastes accumulated at that stage of the nuclear cycle, either as solid active waste or in liquid form depending on whether mechanical or chemical decladding techniques are employed.

In contrast, the Magnox fuel retaining springs are removed prior to fuel transport and are retained as solid active waste at the reactor site. This waste, together with the activated core structures and moderator, remain to be disposed of when the reactor is finally dismantled at the

end of its life. This operation is likely to be postponed for several years following reactor shut-down in order to allow decay of the shorter-lived radioactive species; thus, it is only the longer-lived activation products which are of interest in the context of the waste management of obsolete nuclear plant (Bainbridge *et al* 1974). For this reason  $^{60}\text{Co}$ , with its penetrating gamma emissions, is one of the more radiologically significant activation products and typical cobalt activities and hazards are included in table 1. At cooling periods up to a few years the  $^{60}\text{Co}$  values are comparable with those of the fission product and actinide wastes, but beyond this time the importance of the cobalt isotope is much reduced due to the relatively longer radioactive half-lives of these other nuclides.

Cobalt is also an important constituent in the radioactive waste materials arising in water cooled reactors, where  $^{60}\text{Co}$  is produced via activation of dissolved impurities and suspended particulate steel corrosion products (crud) present in the coolant. Deposition of activated crud around the reactor circuit is a major source of operator doses during routine operation and maintenance of both light water (LWR) and heavy water (SGHWR or CANDU) moderated plant, necessitating periodic decontamination of the system. In these circumstances  $^{60}\text{Co}$  is accumulated in the ion exchange resins and other spent filtration media which must be stored in the active waste facilities at the reactor site.

Of the other longer lived activation products which are normally considered in waste management studies, tritium and  $^{14}\text{C}$ , with half-lives of 12.3 and 5760 years respectively, are worthy of mention and typical parameters for these isotopes in power reactor systems are also given in table 1. The  $^{14}\text{C}$  values are based upon the build-up of this isotope within the graphite moderator of gas cooled reactors, while the wide range of tritium values quoted occurs because this isotope may arise from a variety of sources, including the fission process itself. The dominant activation sources of tritium are neutron captures in lithium impurities in graphite for gas cooled reactors, or in boron coolant additives in light water reactors and deuterium captures in the case of heavy water systems. Both isotopes decay via weak beta emissions and thus do not give rise to the gamma shielding problem associated with the  $^{60}\text{Co}$  decays, their main radiological significance being as potential sources of internal irradiation due to uptake by man following accidental leakage of waste materials to the environment.

Finally, one short-lived activation product which may be included under the heading of nuclear waste is  $^{41}\text{Ar}$ , which occurs in the biological shield cooling air of the earlier Magnox reactors. This isotope has a half-life of 1.83 h and is produced by neutron activation of naturally occurring argon, as well as argon im-

purities in the CO<sub>2</sub> coolant of Magnox reactors and AGRs. Dose rates due to routine argon discharges close to power reactors are generally small compared with those due to natural background sources (UNSCEAR 1972), while the hazard index for <sup>41</sup>Ar shown in table 1 is also of little radiological consequence.

The examples outlined here indicate that activation products may be accumulated at the reactor site or as part of the waste arising from the irradiated fuel reprocessing. In the former instance they are an important consideration in the context of the dismantling and disposal of obsolete nuclear plant. However, in the case of reprocessing wastes the activation product contributions to both activities and hazards are relatively insignificant compared with those from the fuel itself, and in particular those of the long-lived actinide isotopes.

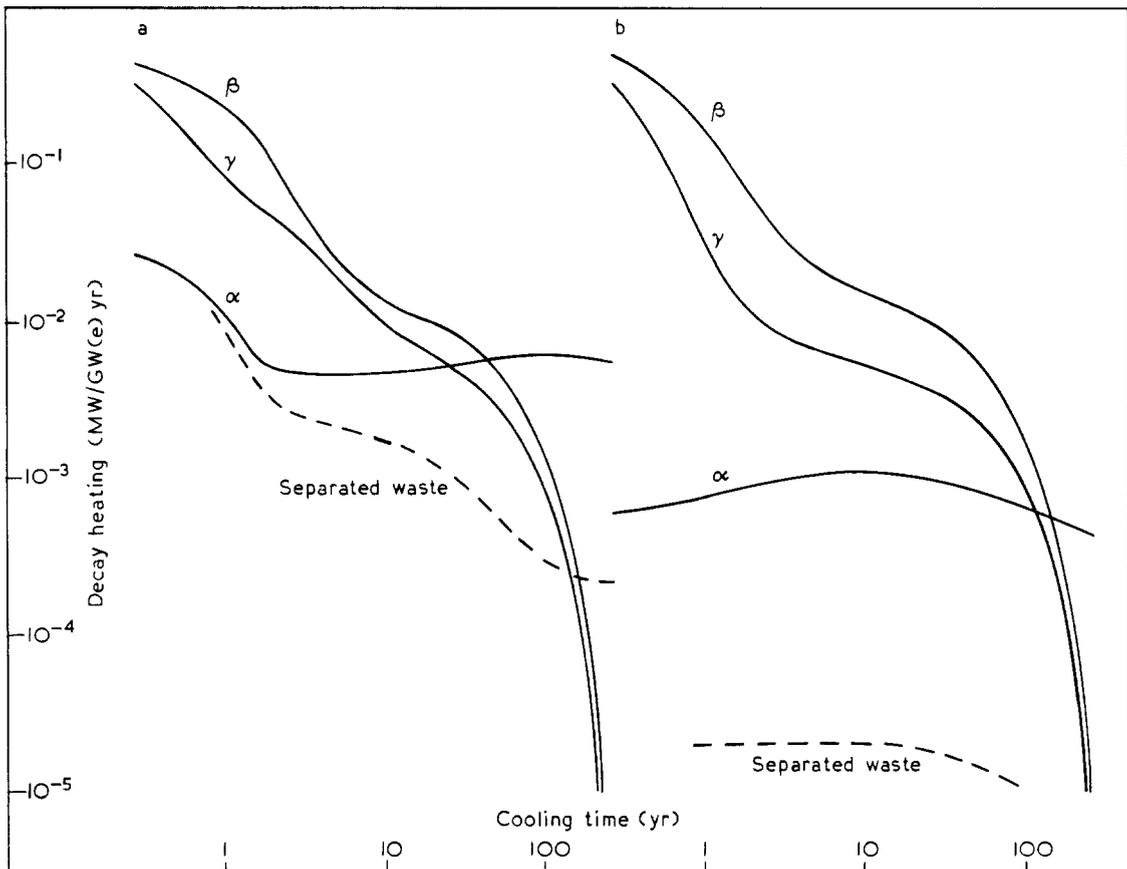
#### Alternative nuclear options

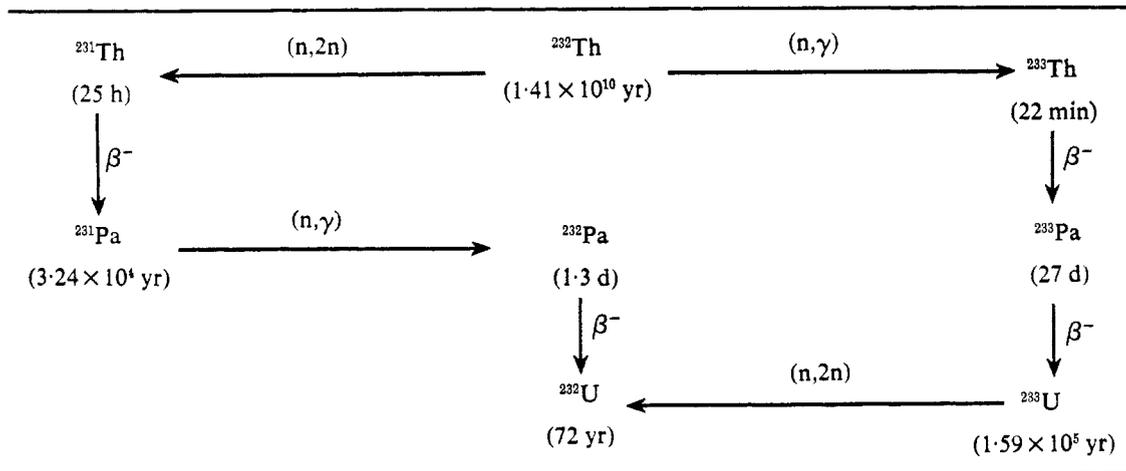
Power reactors based on the thermal fission of <sup>235</sup>U provide a relatively inefficient means of exploiting the world's uranium reserves, since this

isotope has a natural abundance of less than 1%. The situation may be improved by the development of fast breeder reactors, which use the fissile plutonium isotopes produced by neutron captures in the more abundant <sup>238</sup>U isotope, and ultimately have the potential of regenerating their fuel independently of the thermal reactors. However, both thermal and fast reactors employing uranium or plutonium fuels are characterized by the production of long-lived radioactive wastes of the type discussed earlier and these require isolation and containment for many hundreds or thousands of years beyond the useful lifetime of the reactor systems in which they originate.

An alternative to these conventional fuel cycles, which is attractive from both the energy resource and waste disposal viewpoints, is the development of thorium-fuelled advanced thermal or fast breeder reactors. Although the naturally occurring <sup>232</sup>Th isotope is not readily fissionable, when subject to reactor irradiation it undergoes neutron captures to form the fissionable <sup>233</sup>U isotope in accordance with the capture and decay scheme opposite.

**Figure 4**  $\alpha$ ,  $\beta$  and  $\gamma$  decay curves for <sup>235</sup>U and Th/<sup>233</sup>U HTR fuel (irradiation 80 GW d t<sup>-1</sup> at 100 MW t<sup>-1</sup>. a-6.67% <sup>235</sup>U, b-4% Th/<sup>233</sup>U





Capture and decay scheme for  $^{232}\text{Th}$

The production of  $^{233}\text{U}$ , and to a lesser extent  $^{232}\text{U}$ , from  $^{232}\text{Th}$  is analogous to the build up of plutonium isotopes via conversion of the  $^{238}\text{U}$  in natural uranium-based fuels and the fission product wastes generated during power reactor irradiation are very similar for the conventional uranium and thorium fuel cycles. However, an important difference in the latter case is the virtual absence of the build-up of the higher transuranic nuclides responsible for the long-term alpha-active wastes associated with the  $^{235}\text{U}$  or plutonium fuels. Instead, the actinide isotopes produced during reactor irradiation are precursors of the naturally occurring radioactive series and tend to decay via alpha or beta and gamma emissions. This results in a much reduced level of alpha activity in the fuel reprocessing wastes, particularly if the unconverted thorium and uranium isotopes are separated for recycle, as can be observed from the alpha decay curves

for enriched  $^{235}\text{U}$  and thorium/ $^{233}\text{U}$  fuels in a high temperature gas cooled reactor (HTR) shown in figure 4 (Clarke *et al* 1975).

The main contributors to the alpha activity of thorium based fuels are  $^{228}\text{Th}$ ,  $^{232}\text{U}$ ,  $^{233}\text{U}$  and  $^{231}\text{U}$ , or, in the case of the waste materials remaining following thorium and uranium separation,  $^{231}\text{Pa}$  and  $^{238}\text{Pu}$ . However, the activity and hazard values for these actinide isotopes, derived on the same basis as those presented in table 1, are small compared with those of the longer lived  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  fission product isotopes, as illustrated by the data for a thorium fuelled thermal reactor given in table 2. Thus the thorium fuel cycle offers the possibility of reduced long-term waste heating and hazard commitments, but this is at the expense of a shelf life problem associated with the fabricated  $^{233}\text{U}$  fuel. Unlike the case of  $^{241}\text{Pu}$  discussed earlier, this arises not from decay of the fissile material, but is due to the presence of

**Table 2** Fission product and actinide activities and hazards for a Th/ $^{233}\text{U}$  fuelled thermal reactor. Values are based on mean burn-up HTR fuel irradiated to  $80 \text{ GW d t}^{-1}$  at  $100 \text{ MW t}^{-1}$  and cooled for 100 d

	Nuclide	Half-life (years)	Activity (Ci/GW(e)yr)	Hazard index ( $\text{m}^3/\text{GW}(\text{e})\text{yr}$ )	
				Water	Air
Fission product isotopes <sup>(a)</sup>	$^{90}\text{Sr}$	28.5	$2.7 \times 10^6$	$6.6 \times 10^{12}$	$6.6 \times 10^{16}$
	$^{129}\text{I}$	$1.6 \times 10^7$	1.1	$2.7 \times 10^6$	$1.8 \times 10^{10}$
	$^{137}\text{Cs}$	30.1	$2.6 \times 10^6$	$1.3 \times 10^{11}$	$5.2 \times 10^{15}$
	$^{154}\text{Eu}$	16	$1.4 \times 10^5$	$7 \times 10^9$	$1.4 \times 10^{15}$
	$^{85}\text{Kr}$	10.73	$2.4 \times 10^5$	—	$8 \times 10^{11}$
	$^3\text{H}$	12.3	$2 \times 10^4$	$6.7 \times 10^6$	$10^{11}$
Actinide isotopes <sup>(a)</sup>	$^{228}\text{Th}$	1.91	$2 \times 10^3$	$2.8 \times 10^8$	$10^{16}$
	$^{231}\text{Pa}$	$3.24 \times 10^4$	$3.9 \times 10^1$	$4.4 \times 10^7$	$10^{15}$
	$^{232}\text{U}$	72	$6.1 \times 10^3$	$7.7 \times 10^9$	$6.8 \times 10^{15}$
	$^{233}\text{U}$	$1.59 \times 10^5$	$2.2 \times 10^4$	$5.5 \times 10^9$	$5.5 \times 10^{15}$
	$^{231}\text{U}$	$2.47 \times 10^5$	$9.6 \times 10^2$	$2.4 \times 10^8$	$2.4 \times 10^{14}$
	$^{238}\text{Pu}$	86.4	6.3	$1.3 \times 10^6$	$9 \times 10^{13}$

$^{234}\text{U}$  in the fuel. The decays of this isotope ultimately lead to the build up, on a timescale of a few months, of the high energy gamma emitting isotope  $^{208}\text{Tl}$ , which makes it necessary to employ shielded fabrication and handling techniques for the unirradiated fuel (Weissert and Schileo 1968).

Another nuclear alternative based on the fission process, which has been developed recently as a means of space propulsion, is the gaseous-core reactor. In this system the solid core of fuel elements with recirculating liquid or gaseous coolant of conventional reactor designs is replaced by gaseous  $\text{UF}_6$  which is recirculated through a large-cavity reactor vessel. The absence of neutron absorbing fuel cladding and core support structures enables high neutron energies and fluxes to be attained, and this leads to more favourable fission to capture cross section ratios in the gaseous-core system, with the result that there is a greater tendency for the long-lived actinide isotopes to undergo fission reactions. The reactor thus exploits the principle of induced nuclear transmutation in order to minimize the build up of actinide wastes by converting them to shorter lived fission product isotopes (Paternoster *et al* 1974). The gaseous-core reactor is currently at the advanced prototype construction phase in the US and offers the possibility of commercial power generation with relatively low actinide waste production towards the end of the century.

Finally, looking ahead into the next century, the development of fusion reactors could provide a viable alternative to fission systems, which is attractive on both economic and environmental grounds (Steiner and Fraas 1972). One of the most promising fusion reactor designs currently under consideration is based on the deuterium-tritium D-T fuel cycle, with the energy released in the D-T reaction being extracted by absorbing the 14.1 MeV neutrons produced in a liquid-metal lithium blanket. The system is characterized by a complete absence of fission product and actinide isotope production and the only radioactive wastes generated are activation products resulting from neutron interactions in the lithium blanket region and its associated structural component materials. The former result in the production of tritium, which is extracted for use as fuel, thus the system is a breeder reactor, while the activation product wastes generated in the containment structures depend markedly on the materials employed. These are likely to be the refractory metals niobium or vanadium, which possess the necessary corrosion resistance to liquid lithium, and the long-lived activation products of interest are  $^{93\text{m}}\text{Nb}$  and  $^{94}\text{Nb}$  with half-lives of 13.6 and  $2 \times 10^4$  years respectively. These arise from neutron activation of the bulk niobium or, if vanadium is used, of trace niobium impurities. However, the hazard levels associated with these niobium isotopes, and also the tritium inventory, of a fusion power reactor are typically

several orders of magnitude lower than those of the more significant fission product or actinide wastes in a corresponding fission reactor (Hall *et al* 1975).

Thus although significant quantities of radioactive wastes will be produced in current thermal and future advanced thermal or fast breeder reactors up to the year 2000 AD, it is possible to anticipate the amounts and composition of these arisings and ensure that adequate waste management provisions are made to deal with them. These centre largely around the development of techniques for the isolation and containment of the long-lived fission product and actinide isotopes which are reviewed in a companion article in this magazine. In addition, alternative nuclear options are available which offer the possibility of fission reactors with more favourable radioactive waste characteristics, while ultimately fusion power could provide an essentially unlimited source of energy with the production of only minimal quantities of long-lived wastes.

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