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PLUTONIUM

by

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ABSTRACT

Discovery of the neutron made it easy to create elements which do not exist in nature. One of these is plutonium, and its isotope with mass number 239 has nuclear properties which make it both a good fuel for nuclear power reactors and a good explosive for nuclear weapons. Since it was discovered during a war the latter characteristic was put to use, but it is now evident that use of plutonium in a particular kind of nuclear reactor, the fast breeder reactor, will allow the world's resources of uranium to last for millennia as a major source of energy.

Plutonium is very radiotoxic, resembling radium in this respect. Therefore the widespread introduction of fast breeder reactors to meet energy demands can be contemplated only after assurances on two points; that adequate control of the radiological hazard resulting from the handling of very large amounts of plutonium can be guaranteed, and that diversion of plutonium to illicit use can be prevented. The problems exist to a lesser degree already, since all types of nuclear reactor produce some plutonium. Some plutonium has already been dispersed in the environment, the bulk of it from atmospheric tests of nuclear weapons.

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DOSE LIMITS; FALLOUT DEPOSITS; HEALTH HAZARDS; NUCLEAR POWER; NUCLEAR WEAPONS; PLUTONIUM; RADIOACTIVE WASTE STORAGE

CONTENTS

PLUTONIUM

SUMMARY

Plutonium occurs in minute traces in natural uranium ores but this was not recognised until after the element had been made artificially by irradiating uranium with neutrons. This was accomplished in 1940, when military interest in the potential applications of plutonium was paramount. In consequence of this interest, nuclear reactors were built for the specific purpose of producing plutonium, of which the isotopic form, plutonium-239, is readily fissionable by thermal neutrons and hence, like uranium-235, is a suitable material for use as a fuel in nuclear power reactors or for the manufacture of nuclear weapons. In each of these applications, use is made of the very large amounts of energy which are liberated in the fission process, which can be a self-sustaining reaction.

The nuclear power reactors now in operation are fuelled by uranium, usually with its natural content of the isotope uranium-235 slightly enriched, but plutonium is generated in these reactors and fission of this plutonium makes a substantial contribution to the output of power. However, a considerable amount of plutonium remains in the fuel when its useful life is completed. This can be extracted from the spent fuel and put to use. Some of it can be recycled as fuel in similar reactors but its most effective use would be as fuel in a different kind of reactor - the fast breeder reactor - which can be made to generate as much, or more, plutonium from the common uranium isotope, uranium-238, than is consumed in the fission process. Since breeder reactors allow 60 or 70 times more energy to be extracted from natural uranium than is the case with the thermal reactors now in use, their introduction would prolong the duration of use of uranium reserves, as a major power source, from perhaps a century to millennia.

Prototype fast breeder power reactors are in operation but much anxiety has been expressed over the implications of safety, should they be developed on a major scale. The reason for concern is the extreme toxicity of plutonium which presents a radiological hazard comparable to that of radium. However, the quantity of radium which has been extracted from uranium ores in the past is minor, and little is now in

use, whereas the development of nuclear power will eventually mean the production of thousands of tonnes of plutonium. The introduction of fast breeder reactors will allow this stockpile to be used as an energy source, but it will also mean that large amounts of plutonium will be in production in fuel reprocessing plants or in transit.

The potential hazards associated with plutonium were recognised as soon as it became available for experiment, and appropriate standards of radiological safety were prescribed for its use. These were based on those already recommended for radium, which stemmed from extensive human experience. The major risk in each case is the induction of cancer. The standards for plutonium appear to have been satisfactory since, in about 35 years, no ill effects appear to have been attributable to it, although quite large amounts have been handled. It has been argued that these standards are quite inadequate but no good evidence backs this assertion. However, irrespective of the adequacy of safety standards, there is some concern that, when large amounts are processed industrially, plutonium may be dispersed in accidents or misappropriated.

There is already some plutonium in the environment. The testing of nuclear weapons up to 1962 released about six tonnes into the atmosphere and most of this was widely dispersed. Much the larger part of this has been deposited but traces can still be detected in the atmosphere. Other plutonium has been dispersed locally as the result of non-nuclear accidents, and we have a good deal of information on its behaviour in the environment. Discharges, other than accidental ones, of plutonium to the environment should be restricted to the limits derived from the accepted primary standards of radiological protection, and this should provide effective control.

The question at issue with plutonium is whether the risks associated with its accidental dispersion or its illicit diversion are sufficient to suggest that we should forego the undoubted benefits of the nuclear economy.

PLUTONIUM

1. INTRODUCTION

Much of the sound and fury engendered by contemporary planning for the growth of nuclear power production is centred on the man-made element, plutonium, which some think is too hazardous a material for mankind to employ. Discussion on this subject is often embattled defence of an established position rather than unbiased examination of possibilities, and presentations are sometimes intended to appeal to emotion instead of logic. For instance, a recent article by two of the more vocal polemicists on the supposed hazards of plutonium is captioned 'The Element of the Lord of Hell?', although it is not clear whether credit for this evocative title should go to Tamplin and Cochran, who wrote the article, or to the editor of the New Scientist, who published it. However, even at this level, there are two ways to look at the question. Hades, the brother of Zeus and Poseidon, ruled the Underworld but the Greeks, who were tactful in these matters, often preferred to call him Pluto, 'the rich one', who could send them corn from underground, corn being their primary standard of wealth.

So we may think of plutonium as an instrument of justice from Hades, who punished wrong-doers, or as a contribution to the necessities of life from Pluto. The reader will want to form his own opinion of which of these opposing views is the more appropriate, and the intention of this paper is to provide material on which to base this opinion. In doing this it will be necessary to say a little about the structure of atoms and the propensity of some of them to disintegrate in ways which are very productive of energy, to consider how this energy can be applied to useful purposes and, finally, to consider whether the use of one element, plutonium, whose atoms have desirable properties with respect to energy production, constitutes a hazard of such totally new dimensions that we might be better off without it.

2. NEW ELEMENTS

Each of the elements has characteristic properties which are a consequence of its atomic number, that is, the number of protons with positive electric charges in its nucleus, and the equal number of the orbital electrons (and their unique arrangement) around the nucleus. Many elements occur naturally as a mixture of isotopes, that is, varieties

of the element that have the same atomic number and chemical properties, but differing nuclear masses. For example, uranium, the 92nd and the heaviest natural element, occurs as a mixture of two isotopes, uranium-235 (0.7%) and uranium-238 (99.3%), the numerical suffix denoting the nuclear mass. Most of the isotopes occurring in nature are stable, but some are not and change spontaneously to a more stable atomic configuration - a different element of higher or lower atomic number - by the emission of beta or alpha particles respectively. Beta particles are electrons and alpha particles are helium nuclei. The change is 'radioactive decay' and occurs at rates characteristic for each radioactive isotope.

The discovery of the uncharged particle, the neutron, in the 1930s provided an efficient means of producing artificial isotopes. Neutrons, especially when slowed down until their speed and kinetic energy correspond to the temperature of the scattering medium ('thermal' neutrons), are readily captured by atomic nuclei to form unstable isotopes. Attempts were made to create new elements with even higher atomic number than uranium by irradiating uranium with neutrons. This objective was eventually reached but, before such transuranic elements were clearly identified, a discovery of enormous significance was made. This was an unexpected mode of decay of heavy elements - fission, in which the nucleus made unstable by acquisition of an extra neutron breaks into two smaller nuclei of about equal mass. Fission is accompanied by the emission of further neutrons and the release of substantial amounts of energy, approximately ten billion times that released by the combustion of carbon atoms in coal. The energy produced in fission corresponds to the net loss of mass by the interacting particles, the equivalence suggested much earlier by Einstein. This discovery had major implications for both war and peace that were soon followed up.

Renewed experiments in 1940 finally led to the predicted discovery of the new elements with atomic numbers 93 and 94. When uranium-238 captures a neutron, it forms uranium-239 which is unstable and changes by beta decay to element 93; this is also unstable and decays in turn by emission of another beta particle to element 94. By analogy with the naming of uranium from the planet Uranus, the 93rd and 94th elements were named 'neptunium' and 'plutonium' respectively - in the latter case without sinister intention. Plutonium-239 is radioactive but since its

rate of decay is low, with a 'half-life' of 24,400 years, it can readily be accumulated. Higher isotopes of plutonium, americium, curium and other transuranic elements have since been identified, but none has the peculiar importance of the isotope of plutonium with mass number 239.

3. NEUTRON CHAIN REACTIONS

Many heavy isotopes may be made to undergo nuclear fission by one means or another but three are of particular interest, uranium-233, uranium-235 and plutonium-239, which are all readily fissionable by thermal neutrons. Since fission is accompanied by the emission of several neutrons for each atom undergoing fission, the process of fission can be made self-sustaining, and energy will be generated continuously as long as the nuclear fuel is present and appropriate conditions are maintained. That is, a chain reaction is possible. This possibility for uranium was recognised in 1939 and verified in 1942.

Putting the concept into practice required a little care; would the result be an atomic explosion or a useful new source of power? Fortunately, the question could be answered in advance by study of the properties of the two isotopes of uranium and, in fact, both results were possible.

Uranium-235 will undergo fission with either thermal or fast (high energy) neutrons, those with thermal energies being the more effective. Uranium-238, however, can be fissioned only with fast neutrons and, since many of these are captured by atoms without producing fission, an explosive chain reaction in the naturally-occurring mixture is impossible. The neutrons released from atoms undergoing fission are fast; if they are moderated, that is, slowed down, and their numbers are regulated, a controlled chain reaction is possible. This reaction will be initiated by the atoms of uranium-235, although some of the uranium-238 atoms will capture neutrons and generate plutonium which can then contribute to the reaction. Since some neutrons are lost from the exterior of the system, the system needs to exceed a certain critical size for the reaction to be self-sustaining; the critical size depends on the material used as moderator, the presence of neutron-absorbing impurities, the presence of neutron-reflecting materials at the boundary, and the isotopic composition of the uranium.

The essence of a nuclear reactor is therefore a core of uranium or other fissionable element. Fission in the core of uranium is regulated

by control rods of neutron-absorbent material and can be initiated, maintained at any desired level, or shut down, at will. That is, fission is controlled and the design of reactors is such that uncontrolled 'run-away' fission, as in nuclear weapons, is not possible. In power reactors the heat produced by the fission process is used to generate steam which drives conventional turbines.

All present commercial power reactors are thermal reactors, although the moderating materials vary. It is also possible to design unmoderated, or 'fast', reactors in which fission is maintained by fast neutrons. Some reactors of this class offer the prospect of much more efficient utilisation of the energy latent in uranium, and will be considered further in relation to plutonium and nuclear power.

4. NUCLEAR WEAPONS

In nuclear power reactors the release of fission energy is a controlled process, but the alternative approach was apparent from the discovery of fission in 1939. If the reaction can be persuaded to proceed fast enough, and within a small volume, the result is an explosion, whose effectiveness needs no emphasis. The basic requirements are fairly well known; a mass of relatively pure fissionable material with appropriate properties, in excess of the critical size needed to sustain the neutron chain reaction, and absence of moderating materials, so that the fission process takes place as rapidly as possible. There are relatively straight-forward mechanical problems in assembling a super-critical mass of material sufficiently quickly without premature detonation, which would reduce the effectiveness of the explosion, and these were overcome in the initial firings.

Natural uranium cannot be used directly for the manufacture of nuclear weapons because the nuclear properties of uranium-238 are unsuitable. Manufacture of nuclear weapons from natural uranium demands that its content of uranium-238 be greatly reduced. To do this the proportion of the minor constituent uranium-235 is increased, through many stages of enrichment until it is the uranium-238 content which is very low. There are a number of ways of doing this, and by 1942 the theory of several such processes was well understood. By 1945, a large-scale gaseous diffusion plant for uranium enrichment was operating successfully in the United States, and by enriching the product still further with electromagnetic machines the manufacture of a uranium-235 bomb became possible.

There was another material from which nuclear weapons could be made. By late 1942 approximately 3 micrograms of the new element, plutonium, had been prepared by irradiating uranium with thermal neutrons. This was sufficient to establish its nuclear properties, and it turned out that not only was plutonium-239 fissionable by thermal neutrons, but its probability of fission was 30% greater than for uranium-235. Therefore plutonium-239 could sustain neutron chain reactions very well, and a nuclear weapon could be made with rather less material than was necessary with uranium-235. There was another advantage; since uranium and plutonium are different elements they can be separated by ordinary chemical procedures although a heavily shielded and remotely operated processing plant is required. Since in 1942 and 1943 it was far from certain that plans to separate the uranium isotopes would be wholly successful, reactor systems were designed and built in the United States for the single purpose of producing plutonium, the first coming into operation in 1944. This approach was also successful and nuclear weapons were made from both the possible materials, the one dropped on Hiroshima in August 1945, being made from uranium and that on Nagasaki from plutonium.

However, it should not be assumed that <u>any</u> reactor-produced plutonium is suitable for the manufacture of nuclear weapons. Not only plutonium-239, but also other isotopes of plutonium are produced, and they have properties which would greatly complicate the design of weapons and reduce the explosive yield. The plutonium for the early weapons was extracted from uranium which had been subject to low 'burn-up', that is, the extent to which the fissionable fuel is consumed. Its composition was 93 per cent plutonium-239 and only 6 per cent plutonium-240, because the low burn-up, equivalent to the consumption of only a few hundred grams of fissionable material per tonne of fuel, restricted the amount of the undesirable plutonium-240. The burn-up in today's commercial reactors reaches 30 kilograms of fissionable material per tonne of fuel, the plutonium produced has a plutonium-240 and plutonium-242 content as high as 25 to 30 per cent, and other transuranic elements are present. Plutonium of this grade is a good nuclear fuel but a poor nuclear explosive.

5. PLUTONIUM AND NUCLEAR POWER

Nuclear power reactors with an approximate aggregate electrical generation capacity of 60,000 megawatts (MWe) are in operation today. Total nuclear electrical generating capacity is expected to reach 2 million MWe by the year 2000. This is based on the assumption that the world's population will grow at 2 per cent per annum and be associated with a 4 per cent power demand growth. This implies a nuclear growth by at least a factor of 30 and assumes that the consumption of other energy resources (for example, coal) will still continue to grow. Current nuclear reactors require 2000 to 2500 tonnes of uranium over their lifetime for each 1000 MWe reactor installed.

The proven reserves of low cost uranium are adequate for the reactors installed so far. Much more uranium is likely to be found, but while the fissionable material in new reactor fuel remains only the uranium-235 isotope, only about 1 per cent of the energy potential of uranium is realised. A significant fraction of the power produced by current types of reactor comes from plutonium formed from the uranium-238 in the fuel while it is in the reactor, as much as half in the case of the Canadian reactors which use natural uranium fuel and are moderated with heavy water.

But conversion of large amounts of uranium-238 to plutonium-239 has a much broader significance than the contribution of some of this plutonium to the output of current power reactors; it has the potential to make available most of the fission energy of natural uranium, not merely the one per cent or so now utilised.

Fuel discharged from the present generation of thermal reactors contains 200 to 400 kilograms of plutonium per year for each 1000 megawatts of electrical power, but the reactors consume much larger quantities of uranium-235. It is possible to re-use some of this plutonium in thermal reactors, providing an alternative to the isotopic enrichment of uranium, and this could reduce requirements for uranium and for enrichment by about 15 per cent. However, even if plutonium is recycled in thermal reactors in this way, the total inventory of plutonium will continue to grow.

Re-use of plutonium in thermal reactors thus offers some economy in the consumption of uranium, but a much more efficient approach is possible. The nuclear properties of plutonium make it a suitable fuel for fast

reactors and offer the prospect of generating more fuel from uranium-238 than is consumed in the chain reaction.

It has been shown that plutonium fuel elements can be used in power reactors. For example, the Russian reactor BR5 has operated successfully since 1959 with plutonium dioxide fuel and, recently, the French fast reactor PHENIX has achieved very high burn-ups for its plutonium dioxide fuel.

The fast breeder reactor is so called because it will generate more plutonium, from uranium-238, than was initially provided in the fuel. Breeder reactors of commercial size, about 1000 MWe, will use about 2 tonnes of plutonium, as a 1:4 mixture with natural or depleted uranium, in their core. The production of plutonium from neutron capture both in the core and in the reflector is expected to exceed the plutonium fissioned by up to 20 per cent. The net amount of plutonium produced may be altered by reducing the amount of uranium in either core or reflector. With current designs, a net gain of 250 kilograms of plutonium per year is expected per 1000 MWe reactor. The design and operating practice of the fast reactor may be altered to stabilise, increase or decrease the plutonium stockpile.

The uranium-238 used in these reactors to create more fissionable material is referred to as the 'fertile' material. It is not the only such material; it is also possible in principle to use the more abundant thorium-232, but in this case the product fissionable by thermal neutrons is not plutonium-239 but uranium-233.

Many countries have delayed approving the use of plutonium as a fuel for thermal or fast reactors. This means that the stockpile of plutonium grows and an excellent nuclear fuel is being treated as a liability. The uncertain future for plutonium has delayed the construction of plants for commercial reprocessing and fabrication of plutonium fuel and this has aggravated the situation. Meanwhile, the considerable storage and safeguards costs accumulate; each gram of plutonium is estimated to cost $US0.50 per year to store.

6. HOW MUCH PLUTONIUM?

There is now something like 50 tonnes of plutonium in the world, all of it generated in nuclear reactors, and it is being accumulated at a rate of about 20 tonnes a year. At present, not much spent reactor fuel is being processed to recover its content of plutonium, most of it

being simply stored, but the plutonium remains and can be extracted when required.

At the rate of expansion of nuclear power generation now in prospect, the total stock of plutonium will be of the order of 1000 tonnes by the end of the century unless positive decisions are made to use plutonium as a reactor fuel. The total could later reach several thousands of tonnes but will eventually attain a peak value, either because the depletion of low cost uranium reserves has made thermal reactors economically unattractive, or because the introduction of breeder reactor makes it possible to begin using the plutonium stockpile as an energy resource. A thousand tonnes of plutonium should be thought of as the energy equivalent of more than 2,000,000,000 tonnes of coal - about enough coal to supply the whole world with its electric power for a year at current rates of consumption. But with plutonium fuel in fast breeder reactors, each power station would be generating enough new plutonium for its own future refuelling, plus a little more to put aside as the initial fuel for a new power station.

Although spent reactor fuel is not at present being reprocessed on a large scale, most such fuel is valuable not merely for its acquired content of plutonium, but also for its residual uranium which usually contains rather more uranium-235 than natural uranium and can usefully be fed back into an enrichment plant. There is therefore an economic incentive to process most spent fuel, and to separate its plutonium from its uranium, even if this plutonium is not immediately re-used in nuclear fuel elements.

Critics of plans for the growth of nuclear power are often misinformed on the relation between plutonium stocks and the introduction of fast breeder reactors which are fuelled with plutonium, and it is necessary to make the points that plutonium is produced in all reactors and that the introduction of breeder reactors will allow the stockpile to be reduced, rather than increased. However, the quantity of plutonium processed in a year will be greater.

7. PLUTONIUM WASTES

The total stock of plutonium in the world may not be a very important statistic, since most of it will be securely held, and there is already enough of it in existence to make trouble if the custodians are so minded, or simply careless. Scaling up the quantities will not

materially change the situation. What may be more important are the quantities of plutonium, in production in fuel reprocessing plants or in transit, which can conceivably be stolen or dispersed as the result of accidents, and the fractions which are lost to the environment by one route or another or remain as residual wastes requiring prolonged storage. The possibility of theft or of illicit diversion of nuclear materials, the requirements for safe transport, and the management of residual wastes, are considered in other papers of this series; in this section a brief review is given of past dispersions of plutonium, possible future dispersions, and the behaviour of plutonium in the environment.

The wastes which result from the reprocessing of spent reactor fuel contain two principal categories of radioactive materials; fission products and transuranic elements. All fission products, the pairs of elements into which the uranium-235 or plutonium-239 split in the fission reaction, are unstable and undergo radioactive decay, or a short series of decays, mostly by the emission of beta particles. The fission products are responsible for most of the radioactivity initially present in the wastes, but their activity is comparatively short-lived and, after a few hundred years, the major hazard will come from plutonium and other transuranic elements with longer lives and greater radiotoxicity.

An infinitesimal amount of plutonium is formed naturally in uranium ores, as was recognised after the discovery of artificially made plutonium, but it is of no possible consequence. Artificially produced plutonium has been dispersed to the environment in several ways and can be detected at very low concentrations in air and soils.

The testing of nuclear weapons up to 1962 released about 400 kilocuries*, or six tonnes, into the atmosphere and this has become widely dispersed. More than 98% of it was deposited by 1965, about one quarter locally at test sites and the rest as world-wide fallout with 80% in the Northern Hemisphere. Another contribution came in 1964 from the atmospheric burn-out of an artificial satellite, which had a thermoelectric power source fuelled by one kilogram, about 17 kilocuries, of plutonium-238. Plutonium-238, which is much shorter lived than plutonium-239, is formed in irradiated reactor fuel as an unwanted by-product, not being fissionable with thermal neutrons, and constitutes about 1% of commercial

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* The curie is defined as a unit of activity defining the number of spontaneous nuclear disintegrations occurring per unit time; 1 curie = 3.7×10^{10} disintegrations per second; whence picocurie, microcurie and similar sub-multiples. One curie of plutonium-239 is about 16 grams.

plutonium. Since the burn-out occurred at a height of 40,000 metres in the Southern Hemisphere, the distribution of plutonium from the satellite debris has been quite different to that from weapons tests. By mid-1970, 95% of the satellite plutonium had been deposited on the Earth's surface, with over 75% going to the Southern Hemisphere. The total quantity is much less than that from weapons tests.

Dispersed plutonium can be detected at very low concentrations in the atmosphere. Concentrations in surface air in 1974, measured at points in both hemispheres, are in the region of 10^{-17} curies of plutonium-239 per cubic metre. Cumulative fallout levels in soils in the temperate United States amount to about 2 millicuries per square kilometre. It is possible to calculate the respiratory intake of plutonium from breathing the recorded concentrations in air; the calculation, for Northern Hemisphere data, suggests (for man) a lung content of around 2×10^{-13} curies and a bone content building up to a similar level. Published data on tissue concentrations are in reasonable agreement with this at 4.5×10^{-13} curies per kilogram for the lung and a rather lower figure for bone. Concentrations of this magnitude are of no significance to human health.

Plutonium has also been dispersed in limited areas additional to those involved in weapons tests. These include the non-nuclear detonation of plutonium-bearing weapons as a result of aircraft accidents, at Palomares in Spain and near Thule in Greenland, losses in controlled effluents from fuel-processing plants, and some minor uncontrolled releases from wastes and from accidents. In all these cases, and in some planned experiments, the behaviour of the dispersed plutonium has been carefully studied. There is therefore a considerable amount of information relevant to the environmental behaviour of plutonium, and much of it is being extended by continued observation. There is in fact a reasonable foundation on which to base environmental protection standards commensurate with the toxicity of plutonium.

The question of how much plutonium will need to be stored as waste in the future cannot be answered with any precision, since the pattern of nuclear power development is not yet clear, nor is the efficiency with which plutonium will be recovered. However, on present indications it is likely that unusable plutonium, accompanied by other transuranic elements and mostly associated with fission products in high-level wastes, will amount to a few tonnes by the year 2000. These wastes have

generated almost as much heat in discussion as in their storage tanks, but there is no reason to think that their disposal presents an insuperable problem. Several alternatives are considered in another paper of this series, dealing with radioactive waste management (AAEC/IP3).

6. DISCHARGE AND DOSE LIMITS

Some plutonium is deliberately released to the environment in controlled waste discharges and it is appropriate to ask whether this procedure is reasonable and safe. There are well established methods for setting safe limits to radioactivity in waste discharges, and plutonium is not essentially different to other radioactive materials; it is not even the most hazardous when considered on the weight basis generally used by the prophets of doom. A radiation worker is allowed a maximum of 0.64 micrograms of plutonium in his body, but only 0.1 micrograms of the commonest isotope of radium, radium-226. There are corresponding limits for other radioactive isotopes, which take account of their relative toxicity, and reduced limits applicable to members of the public whose radiation dose does not come from occupational exposure.

The primary standards of radiation protection are set down by both national and international bodies, and incorporated into legislation in the form of allowable annual maximum doses of radiation, with some variation in the dose limits for different organs. These limits do not specify the kind of radiation nor, if it comes from radioactive materials, restrictions on the doses from particular radioisotopes. The limiting quantities of radioactive elements, such as those given above for radium and plutonium, are the amounts that will not cause the permissible radiation doses to the various organs to be exceeded. From these limits it is possible to derive tertiary limits to govern the intake of radioactivity to the body in food, drink or respired air, so that allowable body burdens of radioactivity in exposed persons will not be exceeded. These derived limits provide a guide to how much radioactivity may safely be released to the environment; in practice releases are kept as far below the limit as is practicable.

In any practical situation where the release of radioactive effluents is contemplated, there will usually be many, often complex, environmental pathways by which these may ultimately cause some radiation exposure to man. It will always be possible to identify some radioisotopes and pathways as being more important than others; these are the 'critical'

isotopes and pathways, since they will determine the limitations on discharges to the environment. Within these pathways there will exist one or more groups of people whose characteristics in some respect: age, location, eating habits for example, will cause them to receive greater radiation doses than the rest of the population; these will constitute 'critical groups', and their response will provide the limits of discharge.

If a critical group cannot be identified, a hypothetical group can be postulated, with characteristics such that its radiation dose will exceed that of any real group. This procedure, the identification of a critical group of exposed persons, is generally the most convenient way to ensure that no individual receives an excessive dose of radiation as the result of environmental discharges of radioactivity; if the dose to members of the critical group is within the accepted limit, it cannot be excessive to anyone else. Limits to the total radiation dose to an exposed population may also be prescribed. In that case, the average dose to all exposed individuals must be estimated, as well as the maximum dose to any one person.

Discharge authorisations are often, and conveniently, expressed in the general form:

$$\Sigma_i \frac{x_i}{(\text{m.p.m.d.})_i} \leqslant 1$$

a summation where x_i is the quantity of the i-th radioisotope discharged during a month (in curies) and $(\text{m.p.m.d.})_i$ is the maximum permissible monthly discharge of the i-th radioisotope (in curies) - the amount which will not cause the tertiary limits referred to earlier to be exceeded. In other words, the formula recognises that more than one radioisotope may be present, and lowers the permissible amounts of individual radioisotopes accordingly, so that the acceptable discharge of radioactivity is not exceeded however many are present. The formula quoted above is a slightly simplified version of the one used to regulate the discharge of radioactive effluents to the Woronora River from the Research Establishment of the Australian Atomic Energy Commission at Lucas Heights (NSW). The reason for quoting it is to bring out the point that discharge formulae need not be written in terms of any particular radioisotope - there is no mention of specific radioisotopes such as those of radium or plutonium. What matters is the total radiation dose, however it is made up, and plutonium, or any other specific

element, is automatically included in the reckoning if it is present in the discharge. If the formula is complied with, no radioisotope, including plutonium, will present any difficulty.

Formulae of this sort have worked effectively so far, and there seems no reason to think that they will not continue to be effective if properly applied. In other words, our present standards already cater for plutonium; if they are adequate - and the historical evidence suggests that they are - no fresh problem arises from the use of plutonium. And if it should be shown that there are peculiarities about plutonium, they can readily be accommodated by revision of the allowable bodily intakes and corresponding adjustments to the value of the m.p.m.d. for plutonium in discharge formulae. There is no present indication that such revision is necessary. The monthly regulation implied in this formula is simply a matter of convenience in operation; quarterly or annual summations could also be used.

9. ENVIRONMENTAL DEPOSITION OF PLUTONIUM

The environmental deposition patterns of plutonium released in the upper atmosphere have been about the same as for other radioactive materials present in fallout, dependent on latitude of injection into the stratosphere and on the scavenging effect of rain and snow but nevertheless fairly similar. The same will not be true for plutonium released locally, either in accidents or in controlled discharges; these discharges may be terrestrial or marine. In terrestrial ecosystems most plutonium is likely to accumulate in soil; uptake by plant species is quite small, the concentration factor from soil being about one ten-thousandth for most species which have been examined, and small terrestrial animals take up very little at all - in fact some observations indicate that plutonium is not accumulated to any measurable extent in the tissues of small mammals from areas contaminated with plutonium. Soil plutonium may be transported by groundwaters and it can be moved about by atmospheric resuspension of particles from disturbed soil, and there are still some uncertainties about the precise influences of these factors. To provide some perspective on what has been said about the terrestrial dispersion of plutonium, it might be noted that the top metre of the Earth's crust is estimated to contain about 1000 tonnes of radium - the equivalent in radiotoxicity of 6000 tonnes of plutonium.

The behaviour of plutonium in aquatic environments will be rather different, and behaviour in marine and fresh water systems can differ. The concentration factors observed for fallout plutonium include values of 100 for mussels and other molluscs, 1000 for zooplankton and 10,000 in seaweeds. This suggests that plutonium concentrations decrease along the foodchain, although some exceptions have been noted. The accident at Thule, when about 25 curies of insoluble plutonium oxide was released into a marine environment, has allowed continued observation of plutonium dispersion over several years. Three years later, about 10% of the plutonium had been dissolved, and that remaining had been dispersed in the sediments as far as 30 km from the original site. Bottom-living marine animals were still contaminated with plutonium, the concentration depending on their proximity to the original site, but concentrations of plutonium in seawater, seaweeds, zooplankton, seals and seabirds in the region did not exceed levels attributable to fallout.

It is apparent that the growth of nuclear power requires a long-term strategy to ensure that the radio-ecological and other environmental information necessary for the management of plutonium and other long-lived nuclear wastes is gathered and is available when wanted. It does not seem likely that there will be any serious difficulty in catering for these wastes.

10. THE BIOLOGICAL HAZARDS OF PLUTONIUM

As noted earlier, there are standards for radiation protection which are accepted nationally and internationally; an account of their origins and evolution is given in another paper of this series (AAEC/IP1). These standards appear to have been perfectly adequate; another paper (AAEC/IP8) reviews their effectiveness in the nuclear industry, including their application to people handling plutonium occupationally, and suggests that not only does the nuclear industry have a good safety record but also that its record is actually better than that of other industries in this respect. The question then arises, why is there currently so much concern over the expansion of nuclear power and, in particular, why is there so much concern over the supposedly unmanageable hazards of plutonium. This concern is of two main kinds; can plutonium be diverted to the manufacture of illicit nuclear bombs or for use as a means of radiological blackmail in some way (see AAEC/IP6), and is plutonium so unimaginably toxic that we should not contemplate its use at all? The second question is considered here.

The radiation protection standards in common use emanate from the International Commission on Radiological Protection (ICRP), a highly respected body with an excellent reputation in scientific matters. The views of the ICRP have been endorsed, with little variation, by national bodies of equivalent stature - The National Council on Radiation Protectio and Measurements (NCRP) of the United States, and the Medical Research Council (MRC) of Great Britain, to name but two. These organisations have based their recommendations on the work of experienced scientists working in the field and publishing their results in the open scientific literature. Their collective minds are not ossified but are open to change when new evidence suggests this to be warranted; indeed all three bodies quoted have revised their views occasionally over the years. Quite recently in fact the MRC has reviewed the toxicity of plutonium, and found it necessary to suggest only quite minor revisions to standards for its use.

It has to be said that the clamorous views to the contrary which have been expressed recently do not stem from any new data not available to the ICRP, MRC, NRCP and other organisations of their sort, and they do not stem from observations or experiments by scientists with a reputation in this field which have been published in the orthodox manner. They do stem from hypotheses which have not been tested by observation or experiment, nor received the usual kind of scientific publication. There is in fact no more evidence that plutonium has all the sinister attributes it is now credited with than there was that strontium-90 in fallout had those attributed to it a few years ago by an earlier anti-nuclear pioneer, Sternglass. However, there are some similarities in the techniques of information, for example the use of evocative titles; 'The Death of All Children' was used to emphasise the alleged properties of strontium and other fallout radioisotopes.

Plutonium is of course a highly radiotoxic and hazardous material; no reputable scientist pretends otherwise, but its effects are not qualitatively different to those produced by other radioactive materials and there is no reason to think that our safety practices will somehow become ineffective in the future. There is not space here to deal with all the statements made about plutonium, but one of them can be examined briefly. This is the 'hot-particle hypothesis', publicised by the two polemicists referred to at the beginning, in support of their contention

that the ICRP standards for allowable plutonium in the lung must be reduced by a factor of over 100,000. The unit of radiation dose, the rem, is defined in terms of energy deposited per unit of mass. In calculating dose to the lung, the ICRP simplifies the procedure by calculating the average dose to the whole lung mass and, in fact, anything else would be inconsistent with its prior assumptions of a linear dose-response function and an absence of dose-rate effects. The ICRP did recognise that this procedure might not be appropriate where there was extreme inhomogeneity of dose - a condition which is present with particulate radioactivity of high specific activity - but an ICRP Task Group investigating this point subsequently concluded that averaging the dose from particulate sources would introduce a safety factor, if anything. The hot-particle hypothesis affirms that the ICRP practice is grossly in error.

The hypothesis has these components:

(i) There may be a very high radiation dose close to an alpha-active particle, i.e. within 50 microns.

(ii) This dose will disrupt tissue structure near the particle.

(iii) Tissue disrupted in this way has a high probability of becoming cancerous. It is supposed that a 1000 rem dose, which extends only over a microscopic domain, is sufficient to produce the local injury and that the risk of cancer appearing in the site is from one in one thousand to one in two thousand.

(iv) Following from this, the presence of 1000 or more alpha-active particles in the lung will lead to a cancer risk approaching unity. Since the particles can be extremely small and still produce a dose of 1000 rem at their surface, the allowable contamination must be very small, and the reduction factor of 100,000 is obtained by selecting the smallest particle size to meet the dose conditions.

The first two components of the hypothesis are statements of fact, the rest is unsubstantiated inference which was prompted by some earlier observations on the induction of tumours in rat skin by irradiation with electrons. There is no reason to think that these experiments have any bearing on the induction of lung tumours by insoluble alpha-active

particles, and the hypothesis does not rest on experiments with alpha-active particles in lungs, nor on any other study of radiation-induced cancer in the lung. The concept that tissue injury precedes the development of radiogenic cancer is an old one, dating from the early development of X-rays, but was abandoned about 25 years ago when it was found to be untrue. The hot-particle hypothesis therefore has a very insubstantial foundation. Several arguments bear against it, as well as the observed lack of harmful effects from plutonium under our current standards of protection.

The allowable body burden of plutonium is derived from a more substantial foundation - the value for radium. The value for radium-226 was set some 40 years ago, at 0.1 microcuries (in the case of radium-226 this is also 0.1 micrograms), this level not having produced harmful effects in people contaminated with radium. Observations since then on the hundreds of people available for study have not indicated any need for change. When plutonium was identified it was expected to have toxicity of a similar order (in terms of curies); animal experiments suggested that it might be 5 times as effective an inducer of cancer in bone as radium - presumably because of different micro-anatomic distribution - and this factor of 5 was allowed for in determining the allowable body burden of plutonium. The limit is therefore quite conservatively based. Now the 'hot-particle' argument can hardly be confined to the lung; other organs must also be supposed susceptible if the tissue injury concept is correct. In the early work on radium it was recognised that its distribution in bone is quite uneven, and that some foci receive doses up to 100 times the mean dose. At the body burdens known to be without effect, local doses in these hot spots (as they were termed) will reach the levels postulated as hazardous by the hot-particle hypothesis. However, there has been no effect.

Experimental studies in animals do not support the hot-particle hypothesis; in fact, they suggest that particulate alpha sources may, in some instances at least, be less effective in inducing cancer than more soluble materials. Nor do theoretical studies of the number of cells put at risk by soluble and insoluble alpha sources provide any support for the theory.

There is in fact no reason, on the grounds of experience, experiment, or theory to suppose there have been major errors in setting

safety standards for plutonium. Minor changes may be desirable on occasion, but drastic revision is not called for.

11. MISUSE OF PLUTONIUM

A nuclear weapon can be made from about 6 kilograms of weapons-grade plutonium; if the manufacturer is aware of all the technical requirements for obtaining efficient explosion and the means of achieving them, and has sophisticated facilities for manufacture and assembly, he can expect to produce something like the Nagasaki bomb. If his design is effective, he may produce something much less efficient but still equivalent to a great deal of conventional high-explosive and, if he is a rank amateur, his bomb will probably not explode at all. If the available plutonium is 'reactor-grade', with perhaps only 70% of plutonium-239, and high proportions of plutonium-240 and other transuranic elements with unwanted properties, an explosive device can still be made but necessarily of much larger physical dimensions and of low overall efficiency. The calculations required to design such an explosive are much more complex than those for weapons-grade material. In the extreme, an explosive device could be made, in principle, from the 20% plutonium-uranium fuel proposed for breeder reactors but 300 kg would be necessary and effective assembly would be so difficult that it would be simpler to separate the plutonium chemically. All such proposals as these presume that the illicit manufacturer can manage the health problems involved and has some means of delivering his bomb. It might be noted that even efficient weapons use only about 5 per cent of their plutonium in fission; the rest will be vaporised and dispersed in the atmosphere.

Stolen plutonium has also been suggested as a convenient weapon for large scale blackmail. To be most effective in this way, the plutonium would have to be dispersed as a fine aerosol and confined to a fairly restricted area. It could, for example, be introduced into the ventilation system of a large building with unfortunate results. However, if it were accompanied by a threat, it would be a simple matter to turn the ventilation off. Anyone contemplating blackmail with plutonium in this sort of way would be better advised to use such agents as botulinus toxin or anthrax spores, which are much easier to make, are more effective weight for weight, and produce their results at once - not 20 to 30 years later as is the case with lung cancer from plutonium.

12. THE BALANCE SHEET

The credit side of the balance sheet is quite simple. Firstly, a substantial part of the world's electricity demand is already met by plutonium, since it provides 25 to 30 per cent of the output of the present generation of nuclear reactors, depending on type. However, its potential contribution is more important. The thermal fission reactors now being installed in most parts of the world utilise only 1 per cent, or a little more, of the energy latent in natural uranium, and do not use the energy in thorium at all.

The world's resources of uranium at economic prices are not unlimited and it hardly seems likely that they would see out the next century if used only in thermal reactors. The introduction of plutonium-fuelled fast breeder reactors would completely change this prospect. In a sense, the plutonium acts as a catalyst to allow fission of the uranium-238 which is the great bulk of the natural element and increases sixty or seventy-fold the energy recovered from a tonne of uranium. The life of reserves would then stretch into millenia, even without the further introduction of thorium as a fertile material. This is a very substantial credit, and it can also be seriously argued that the production of power by nuclear means will reduce the overall environmental damage associated with meeting our demands for power. At present no alternative in clear sight has these virtues.

The debit side must include the complex controls and regulations necessary to ensure the safe use of plutonium, and heavy responsibilities must rest on those who administer these controls. However, at present, reflection on the issues arising from the use of plutonium tends to be bedevilled by fear. Fear that the persons to whom we entrust the duty of prescribing standards of safety are incompetent, fear that if they are competent we shall be unscrupulous and not adhere to their standards, fear that terrorist or other groups will steal plutonium and blackmail the rest of the world with it and, above all, fear that the growth of nuclear power will somehow make it more likely that nuclear weapons will be used again.

Many of the fears expressed about nuclear matters are as soundly based as those of the people who were reluctant to travel behind Stevenson's Rocket, but they are mostly genuine and come from deficient or faulty information. A sensible choice between the two aspects of plutonium

requires us to be well informed on both, but we should always be cautious about the source of information.

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