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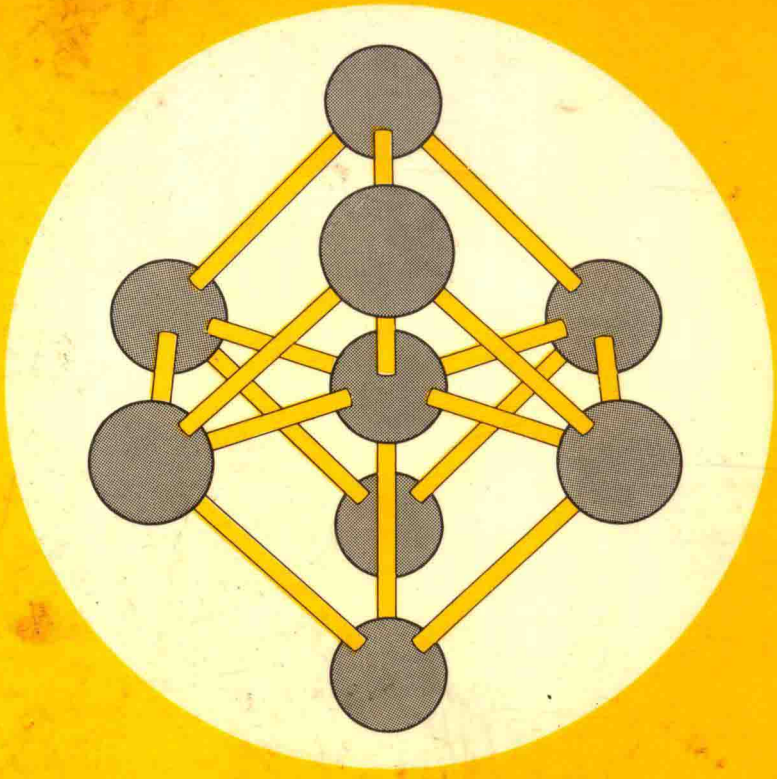
NUCLEAR POWER

Health implications of transuranium elements



NEPTUNIUM 93 Np	PLUTONIUM 94 Pu	AMERICIUM 95 Am	CURIUM 96 Cm

World Health Organization
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Copenhagen

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NUCLEAR POWER:

HEALTH IMPLICATIONS OF TRANSURANIUM ELEMENTS

Report on a Working Group
Brussels, 6–9 November 1979

WORLD HEALTH ORGANIZATION
REGIONAL OFFICE FOR EUROPE
COPENHAGEN
1982

This publication is dedicated to the memory of the late Professor S. Halter, Secretary-General, Ministry of Public Health and Family Affairs, Belgium, who initiated and strongly supported Regional Office activities on the health implications of nuclear power production

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Foreword

A major international debate on alternative forms of energy production is in progress and will undoubtedly continue. Two of its most important and often conflicting aspects are on the one hand cost, and on the other health, safety and environmental considerations. Risk assessment studies have shown that the older and more familiar forms of energy may, in fact, be more hazardous to those involved in their production and utilization than is nuclear technology, even though the older power industries have had a much longer time to establish and improve safety standards. Yet the public perception of risk is often different. It is clearly essential that all decisions concerning the potential effects on human health of different forms of energy production should be based on adequate scientific data, and it is equally important that accurate information should be available to the public in an accessible form.

No doubt largely due to economic recession, previous estimates of energy demand have proved to be excessive in many countries. This respite is at least allowing time for more thorough appraisal of the health and environmental implications of the various alternatives.

A particular dilemma faces the developing countries, where traditional industries often exist side by side with the most advanced technology. There are unfortunately no "third world", low-cost solutions to the problems created by high technology such as nuclear power; effective health protection for workers and the community by high standards of construction and operation and by comprehensive monitoring and control procedures must therefore be regarded as essential, whatever the level of economic development of the country which decides to adopt them.

This publication is the second of a short series being developed by the WHO Regional Office for Europe covering the health implications of different aspects of nuclear power production. The first, dealing with the overall fuel cycle, was published in 1978, and the present volume has as its subject health aspects of transuranium elements. A third volume, on highly radioactive waste, is expected to be published by the end of 1982. All have been prepared on the basis of the deliberations of expert groups, and the financial assistance of the Government of Belgium in convening these groups is gratefully acknowledged. Special reference must be made to the wise counsel and support provided by the late Professor Halter, Secretary-General, Ministry of Public Health and Family Affairs, Brussels.

The present volume is intended to provide guidance to public health administrations and others concerned with this important topic. The distinguished expert group involved in its preparation, under the chairmanship of Sir Edward Pochin, in addition to providing guidance on the health considerations at the nuclear power plant itself and in connexion with reprocessing facilities, also touched on problems related to transport of materials and threats of terrorism.

There is no such thing as absolute safety, but all new (as well as old) technology must attempt to reduce risks to a minimum. In the case of nuclear power production, safety standards have generally been extremely high and it is important that, at the very least, these should be maintained. It is hoped that this volume will help to clarify the public health issues concerning the transuranium elements, which are likely to be of increasing significance in relation to nuclear energy development.

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CONTENTS

Introduction

Public interest in the health aspects of transuranium elements, together with the need for an outline of the current factual position as regards these elements, led to the convening of a Working Group by the WHO Regional Office for Europe in collaboration with the Belgian Government on 6-9 November 1979. The objective was to consider, discuss and assess the health aspects of these elements.

This report, which is based on the collective knowledge and experience of the members of the Working Group, together with the extensive literature available in this field, provides general guidelines for national public health and environmental authorities. It does not provide detailed technical data, but instead broadly surveys the whole field, indicating the present position as assessed by members of the Working Group.

The meeting was the second in a series organized by the Regional Office on the health effects of nuclear activities; the first dealt with the health implications of nuclear power production and a report was issued in 1978 (*1*).

The meeting was attended by 26 participants from 12 countries. Four major disciplines (health administration, radiation biology, health physics, toxicology) and 5 professional categories (physicians, biologists, engineers, physicists, chemists) were represented, providing a comprehensive multidisciplinary approach to the subject. Representatives from intergovernmental and nongovernmental organizations were also present. The participants acted in their individual capacities and not as representatives of their own countries (see Annex 4).

The Working Group considered three major facets: physiological, toxicological and dosimetric aspects of the transuranium elements; the application of occupational health control, including health physics; and environmental behaviour together with public health implications associated with the transuranium elements. The intention was to cover all aspects relevant to health, with respect both to those who are occupationally engaged in work with such elements and to the general public who might be affected as the result of such operations. Consideration was also given to routine and emergency situations.

This report includes a brief summary of the physical and chemical properties of those transuranium elements considered by the Group to be

important (Chapter 2). Chapter 3 deals with the radiobiological aspects of these transuranium elements, including their metabolism, dosimetry and health effects. This is the most important section since the health implications of the transuranium elements depend on the fundamental parameters and assumptions discussed here, and these provide the basis for a quantitative evaluation of the associated health risks. Consequently, the data and arguments presented in this chapter are more detailed than in others. Chapter 4 deals with the sources of human exposure; these are considered in further detail in Chapter 5, which is concerned with occupational exposure, and in Chapter 6, in which the environmental aspects arising from possible releases of transuranium elements are considered. Since the latter topic involves the measurement of radioactivity levels in the environment as a result of such releases, Chapter 7 is concerned with environmental monitoring. Chapter 8 briefly discusses control and protective measures.

Sir Edward Pochin was elected Chairman of the Working Group, Dr W. Jacobi, Dr R.O. McClellan and Dr N. Wald Vice-Chairmen, Mr H. Howells Rapporteur, and Dr B.G. Bennett and Dr J.-C. Nénot Co-Rapporteurs. Dr M.J. Suess acted as Scientific Secretary.

On the basis of a preliminary draft prepared by the Rapporteurs and subsequent comments from the members of the Working Group, a drafting committee consisting of Dr Bennett, Mr Howells, Dr Nénot, Dr Pochin and Dr Suess prepared the final report.

1

Conclusions and recommendations

Metabolism and health effects

Conclusions

1. On the basis of existing knowledge about the behaviour and effects of transuranium elements within the body, it is evident that the radiotoxicity of this group of elements is not unique. The actual and potential conditions of occupational and environmental exposures of man are complex but, by taking into account the physicochemical properties of the transuranium elements as well as their metabolic behaviour, the radiation doses can be evaluated and the risk of health effects estimated with sufficient accuracy for the purpose of radiation protection. It is unlikely that the risk of death from cancers in irradiated organs is underestimated by such methods, and in some cases it may even be overestimated.

Recommendations

1. The development of new techniques and processes has led to the use of new transuranium compounds. These materials include plutonium-sodium compounds, finely divided plutonium particulates, and chemical complexes such as tributyl phosphate plutonium. Further study is needed of the behaviour and effects of such materials.

2. Further studies of metabolic processes associated with transuranium elements would increase the accuracy of predicting health risks under different circumstances and the capacity to deal more confidently with human exposures. In particular, the effect of human age on the metabolism and tissue distribution of transuranium elements after inhalation, and especially after ingestion, should be studied. Such data are needed to determine how the annual limit of intake varies for individuals of different ages.

3. Further research is needed into the possible combined effects of alpha irradiation, together with exposure to various other agents. The question of any such associated risks would be important, particularly for any occupational groups exposed to alpha radiation and to chemical carcinogens.

Health physics and occupational aspects

Conclusions

1. The general principles of monitoring industrial operations in which radioactive materials are used apply also to those operations involving transuranium elements. The techniques adopted must be appropriate to the particular radionuclides involved.

2. The optimal method for the prevention of significant radiation exposure of personnel working with transuranium elements lies in the provision of effective containment of the material.

3. For installations where the on-site medical services are unable to provide for workers exposed to transuranium elements, off-site medical services can only be expected to provide effective cover if special training, equipment and medication, including chelating agents, are kept constantly available in advance of need.

Recommendations

1. For major installations, such as fuel fabrication plants, nuclear power reactors and fuel reprocessing plants, the public authorities involved in the event of off-site emergencies should be informed in advance of any procedure they may be advised to follow. Planning arrangements to deal with off-site transuranium releases requires close coordination between local management and those agencies which might be involved.

2. The management of patients occupationally exposed to transuranium elements requires medical personnel with appropriate preparatory training, adequate equipment and the necessary medication, including chelating agents, to deal effectively with such cases.

Environmental aspects

Conclusions

1. The largest releases of transuranium elements to the environment have come from atmospheric nuclear testing. Releases from all nuclear fuel cycle operations are much smaller, and are largely limited to those from fuel reprocessing plants. Releases of transuranium elements from satellites and consumer devices, such as cardiac pacemakers, are even smaller in normal circumstances.

2. It is important to estimate the levels of fallout plutonium and americium in the environment, since any releases in the future from nuclear power installations or other sources would be superimposed on this background. The use of isotope ratios may often be useful in delineating contaminated areas.

3. The transuranium elements behave in the environment in a manner fairly typical of heavy metals; there appear to be no unusual transfers. The transuranium elements are not readily taken up by plants or absorbed through the gastrointestinal tract of animals or man, although absorption may be greater for organically combined forms, such as those that may occur in foodstuffs. For initially airborne releases, inhalation is generally the predominant route into the body.

4. Methods have been developed and are widely used to estimate the transfer of radionuclides through the environment. Fairly good estimates of the intake by man of transuranium elements, their distribution and retention by the body, and the consequent dose, can be obtained by proper use of the various parameters appropriate to the specific environmental situation.

5. Analytical and monitoring techniques are quite sensitive in determining trace concentrations of transuranium elements in the environment.

Recommendations

1. Precautions regarding the safe utilization of transuranium materials and the prevention of releases to the environment, which have been generally effective in the past, must be fully maintained in the future.

2. Although the behaviour of transuranium elements in the environment has been extensively studied and is fairly well understood, special environmental conditions and certain physical or chemical forms of the transuranium compounds require further attention.

3. It is particularly difficult to generalize about resuspension, since it may vary widely according to conditions. There are also many situations which are not well quantified, such as resuspension from water in spray, from sediments in intertidal zones, and from sediments into water, and further study of these processes is recommended.

4. As absorption of plutonium through the gastrointestinal tract may be greater for organically combined plutonium than for other chemical forms, it is important to determine the fraction of ingested intake which is in this form.

5. Because of the very long-term persistence of transuranium elements in the environment, periodic measurements of the amounts present in air, surface soil, sediments, foodstuffs, and man will be required to assess any change in the level of such activity reaching man or in levels of accumulation.

2

Physical and chemical properties

The transuranium elements are those that occur above uranium in the periodic table. They include neptunium, plutonium, americium and curium, all of which are generated in nuclear power reactors and are used in various industrial and medical products and procedures (see Chapter 4). The transuranium radionuclides are largely alpha particle emitters and generally have long radioactive half-lives. Their general properties have made them of special concern in assessing the health and environmental aspects of their utilization. The more specific physical and chemical properties of the transuranium elements are summarized below.

Plutonium

Trace quantities of "element 94" were first isolated in 1941 but the name "plutonium" was not coined until a year later. During the next few years it was produced in kilogram quantities. There are 15 known isotopes of plutonium, with mass numbers ranging from 232 to 246. Most of the isotopes with even mass numbers are alpha particle emitters, those with mass numbers 232, 233, 234, 235 and 237 decay by electron capture and those with mass numbers 241, 243, 245 and 246 decay by beta particle emission. Many of the isotopes that are alpha emitters also exhibit spontaneous fission, while in all cases of particle emission from these plutonium isotopes the decay schemes include X-ray and gamma-ray releases over a wide energy range.

Table 1 shows the major physical properties of the plutonium isotopes of interest (*2,3*) and Fig. 1 illustrates their mode of formation from uranium-238 (*4*).

Plutonium is a silvery-white metal, similar to nickel in appearance, which in an atmosphere of moist air rapidly oxidizes to form a mixture of oxides and hydrides. Under such conditions the metal is highly pyrophoric and consequently the pure metal is handled in inert dry nitrogen or argon atmospheres. It exists in six allotropic forms; at room temperature it is in the "alpha" phase with a mass density of about 19 000 kg/m^3.

The metal is readily soluble in moderate to high concentrations of hydrochloric acid, insoluble in nitric acid, and dissolves only slowly in sulfuric

Table 1. Physical properties of the major isotopes of plutonium, americium, curium and neptunium[a]

Isotope	Radioactive half-life[b] (years)	Principal mode of decay	Mean alpha energy[b] (J)	Specific activity (MBq/kg)	Mass per unit activity (kg/MBq)	Relative mass[c]
^{236}Pu	2.85	α	9.21×10^{-13}	1.97×10^{10}	5.08×10^{-11}	1.15×10^{-4}
^{237}Pu	1.25×10^{-1}	EC[d]	EC[d]	4.48×10^{11}	2.24×10^{-12}	5.08×10^{-6}
^{238}Pu	8.78×10	α, X-rays	8.75×10^{-13}	6.33×10^{8}	1.58×10^{-9}	3.58×10^{-3}
^{239}Pu	2.44×10^{4}	α, X-rays	8.25×10^{-13}	2.27×10^{6}	4.41×10^{-7}	1.00
^{240}Pu	6.54×10^{3}	α	8.25×10^{-13}	8.44×10^{6}	1.18×10^{-7}	2.69×10^{-1}
^{241}Pu	1.44×10	β	β	3.66×10^{9}	2.73×10^{-10}	6.19×10^{-4}
^{242}Pu	3.87×10^{5}	α	7.83×10^{-13}	1.41×10^{5}	7.08×10^{-6}	1.61×10
^{243}Pu	5.66×10^{-4}	β	β	9.62×10^{13}	1.04×10^{-14}	2.36×10^{-8}
^{241}Am	4.32×10^{2}	α, γ-rays	8.79×10^{-13}	1.20×10^{8}	8.32×10^{-9}	1.89×10^{-2}
^{243}Am	7.40×10^{3}	α	8.44×10^{-13}	7.36×10^{6}	1.36×10^{-7}	3.08×10^{-1}
^{242}Cm	4.47×10^{-1}	α	9.77×10^{-13}	1.22×10^{11}	8.16×10^{-12}	1.85×10^{-5}
^{244}Cm	1.79×10	α	9.29×10^{-13}	3.03×10^{9}	3.30×10^{-10}	7.48×10^{-4}
^{237}Np	2.14×10^{6}	α	7.66×10^{-13}	2.61×10^{4}	3.86×10^{-5}	8.75×10

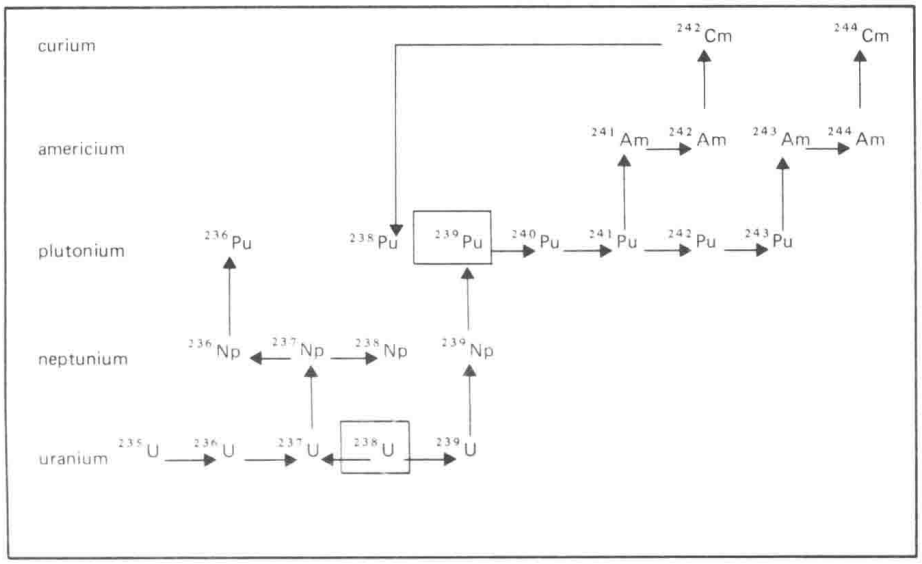
[a] After **Nénot & Stather** (*2*).

[b] After **Harte** (*3*).

[c] Mass per unit activity relative to ^{239}Pu.

[d] EC = electron capture.

Fig. 1. Formation of plutonium and transplutonium elements



Source: **Nénot (*4*).**

acid. The chemistry of plutonium is complex, though generally its properties are in line with other actinides (the series from actinium to lawrencium).

Plutonium can exhibit five oxidation states from III to VII. In aqueous solutions, plutonium VI is oxidized to plutonium IV, the most stable. The chlorides, nitrates and sulfates are all soluble in water. Plutonium dioxide (PuO_2) is the most important compound. It is a highly refractory material, usually green in colour, which melts at between 2200 and 2400 °C depending on the nature of the surrounding atmosphere. It is difficult to dissolve by normal methods but can be dissolved after heating in 85-100% phosphoric acid at about 200 °C. If the dioxide is prepared using high temperatures, it is even more refractory in nature. Because of its refractory properties and chemical stability this is the preferred form for storage and shipping of plutonium.

The complicated chemistry of plutonium is discussed in some detail by Wick (*5*), and Cleveland (*6*) covers more general aspects.

Americium and curium

The elements americium and curium (atomic numbers 95 and 96 respectively) were discovered in 1944-45. Americium has 13 known isotopes (atomic numbers 232, 234 and 237-247), of which 241 and 243 are the most important with half-lives of about 430 and 7400 years respectively; these have been produced in kilogram quantities. Americium-241 is the isotope most abundantly produced in nuclear reactors and it arises from beta decay

of plutonium-241. Americium itself decays by alpha particle emission with an accompanying gamma photon of energy of 60 keV from 40% of disintegrations. Curium has 13 known isotopes (atomic numbers 238–250) of which those of mass numbers 242 and 244 and half-lives of about 163 days and 18 years, respectively, are produced in significant amounts; these isotopes are also alpha particle emitters.

The isotopes americium-241, americium-242 and curium-244 are also considered to be fissile materials (*7*). Table 1 shows the principal physical properties of the important americium and curium isotopes.

Americium is a silvery-white metal which is malleable, ductile and melts at about 990 °C; it oxidizes slowly in air. Curium is a silvery, hard, brittle metal which melts at about 1300 °C and oxidizes rapidly in air. The oxides of both these elements are more soluble than plutonium dioxide. In aqueous solution the most stable oxidation state of these elements is the trivalent (i.e., americium III, curium III) and this is the most important state in relation to biological systems. General aspects of hydrolysis and complex ion formation are similar to those of plutonium. Several authors have reviewed the chemistry of americium and curium (*8–10*).

Neptunium

There are 11 isotopes of neptunium ranging in mass number from 229 to 239. Most of the isotopes have short half-lives, with the exception of neptunium-235, which has a half-life of about 396 days, and neptunium-237, which has a half-life of 2.14×10^6 years. Neptunium-237 is a radioisotope of some importance since it is used as the target material in a nuclear reactor to generate plutonium-238, which may then be used as a radioisotopic power source (*11*). Further, neptunium-237 is an important component of reactor waste because of its long half-life and its potential mobility in geological media after burial has taken place. The physical properties of neptunium are described in Table 1.

Neptunium was first isolated as a weighable quantity ($\sim 45\,\mu g$ of oxide) in 1944 by Magnusson & LaChapelle. It is a silvery, ductile metal with a melting point of 637 °C and a boiling point of 4175 °C. It has three allotropic forms below the melting point and at room temperature; in the "alpha" phase it has a density of 20 500 kg/m^3. Neptunium will slowly oxidize in dry air at room temperature but will rapidly oxidize if the temperature is raised, especially in moist air. It is readily soluble in hydrochloric acid at room temperature, and will dissolve in sulfuric acid at elevated temperatures, but has no visible reaction with nitric acid.

As for plutonium, compounds of neptunium can be prepared in five oxidation states (III–VII), which can exist in aqueous solutions.