RD/B/N3523

Central Electricity Generating Board

Research Department

Berkeley Nuclear Laboratories

RADIOLOGICAL IMPLICATIONS OF PLUTONIUM
RECYCLE AND THE USE OF THORIUM FUELS
IN POWER REACTOR OPERATIONS

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RD/B/N3523

Job No: XJ034

January 1976

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Radiological Implications of Plutonium Recycle and the Use of
Thorium Fuels in Power Reactor Operations

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For inclusion in Nuclear Science Abstracts

SUMMARY

As economically attractive sources of natural uranium are gradually depleted attention will turn to recycling plutonium or the use of thorium fuels. In this study the radiological implications of these fuel cycles in terms of fuel handling and radioactive waste disposal are investigated in relation to a conventional ^{235}U enriched oxide fuel. It is suggested that a comparative study of this nature may be an important aspect of the overall optimisation of future fuel cycle strategies.

It is shown that the use of thorium based fuels has distinct advantages in terms of neutron dose rates from irradiated fuels and long term α decay heating commitment compared with conventional uranium/plutonium fuels. However, this introduces a γ dose rate problem in the fabrication and handling of unirradiated ^{233}U fuels. For both plutonium and thorium fuels these radiological problems increase during storage of the fuel prior to reactor irradiation. Finally, the novel health physics problems which arise in the handling and processing of thorium fuels are reviewed in an appendix.

CONTENTS

1.   INTRODUCTION

The use of natural uranium or ^{235}U enriched fuels in thermal power reactors is relatively inefficient in terms of its utilisation of the world's uranium resources since this isotope has a natural abundance of only 0.71%. The situation is improved somewhat by the production of thermally fissionable isotopes of plutonium, particularly ^{239}Pu, via neutron captures in the more abundant ^{238}U isotope during the reactor irradiation, and this extends the useful life of the fuel beyond that determined by the reactivity of its uranium content alone. However, the full realisation of the promise of nuclear power to provide a viable source of energy from fission reactors into the next century requires the development of a breeder system, such as that envisaged in current plutonium fuelled fast reactor designs. These employ plutonium produced within thermal reactors as their initial source of fissile material, but generate or breed their subsequent plutonium requirements via conversion of the fertile ^{238}U isotope which, together with the isotopes of plutonium, make up the bulk of the heavy element content of the fuel.

An alternative to the use of such uranium/plutonium fuel cycles in fission reactors is the thorium fuel cycle which offers the possibility of the development of both thermal and fast breeder systems (IAEA, 1966). The naturally occurring isotope of thorium, ^{232}Th, is not readily fissionable, but during power reactor irradiation may be converted to ^{233}U which can give a thermal neutron production ratio of 2.27, compared with corresponding values of 2.1 and 2.0 for ^{235}U and ^{239}Pu respectively, and during the past 10-20 years the feasibility of such systems has been demonstrated over a range of reactor designs (Weissert and Schileo, 1968). The thorium cycle has a number of advantages over conventional uranium/plutonium cycles, including more effective use of natural resources and more favourable long-term waste disposal characteristics (Clarke, Macdonald, Fitzpatrick and Goddard, 1975). However, these are at the expense of a gamma dose rate problem associated with unirradiated ^{233}U fuels, which imposes a significant shielding requirement during fuel fabrication and handling operations.

In this note the radiological aspects of plutonium recycle and the use of thorium fuels are examined, with special reference to fuel handling and irradiated fuel reprocessing waste management. Results of a comparative study covering enriched natural uranium, plutonium and thorium fuels irradiated in a conceptual advanced thermal reactor, based on a high

temperature gas cooled reactor (HTR) design, are presented and used to identify areas of potential health physics concern together with the relevant radionuclides of interest. Detailed considerations of factors such as fuel design, neutronics and fuel management schemes are not included since their influence will depend on specific reactor design features, but they are discussed in general terms in instances where they may affect the broad conclusions of this study.

2. ALTERNATIVE FUEL CYCLES

The reference fuel and irradiation history selected for this study was a 6.75% ^{235}U enriched UO_2 irradiated to 44,000 MWD/T at a mean rating of 55 MW/T in an HTR core environment. The effects of plutonium recycle were typified by a 5% Pu mixed PuO_2/UO_2 fuel employing magnox plutonium stored for a period of 10 years following separation, and the thorium cycle was represented by a 9.5% ^{235}U enriched mixed ThO_2/UO_2 fuel. In the latter case ^{233,235}U oxide fuels based on the mixture of thorium and uranium isotopes separated from the initial thorium fuel during reprocessing at 200 days after discharge from the reactor were also included. The fuel burn-up and rating parameters used in this study are intermediate between those currently projected for commercial water reactor and HTR designs, and thus in general terms may be taken as representative of a range of advanced thermal reactors. Indeed, it has been noted by Desoisa (1975) that the lower fuel burn-up and softer neutron spectrum characteristic of some water reactor systems may yield higher ^{233}U and plutonium conversion ratios than those achievable in an HTR, although considerable uncertainties exist in the basic nuclear data for many of the neutron capture reactions involved, particularly those associated with ^{233}U production (Clarke, 1971; Clarke et al., 1975).

The isotopic contents and radioactive emissions from the various fuels for the standard irradiation history quoted above were evaluated using the FISP4 and HYLAS2 computer codes (Clarke, 1971, 1972; Beynon, 1973). The appropriate fuel enrichments and variations in neutron fluxes and fractional fission rates with burn-up necessary to maintain a constant power rating relative to the reference UO_2 fuel were determined using the iterative technique described by Clarke et al. (1975). The resultant fractional fission rates, divided into thermal, resonance and fast groups, are summarised in figure 1, while the activities at 100 days cooling of some of the more important fission product and actinide isotopes are presented in table I.

Apart from minor variations associated with differences between the
fission yield curves for ^{233,235}U and ^{239}Pu, the fission product inventories
of the various fuels depend only on the burn-up and rating, and are rela-
tively insensitive to the fuel composition. This can be seen from the
similarity between the β and γ decay curves presented in figure 2, which are
dominated by the fission product contributions, and significant differences
are only apparent in terms of the activities of a few individual isotopes
such as ^{90}Sr and ^{106}Ru (see table I). In contrast, the actinide inventories
of the various fuels show marked variations both for individual isotopes
and in their nett α emissions (see table I and figure 2), which in the case
of the enriched ^{235}U and plutonium fuels arise from isotopes of plutonium,
americium and curium. At short cooling times the α decays are dominated by
^{242}Cm, while beyond the first few years after discharge from the reactor
^{238}Pu and ^{241}Am formed by decays of ^{242}Cm and ^{241}Pu respectively, and also
^{244}Cm, become important. For the thorium fuels the α emissions arise from
^{238}Pu built up during the reactor irradiation, together with ^{228}Th and its
daughter products formed by decays of ^{232}U, or if this isotope is removed
during reprocessing, ^{231}Pa and its daughters in the separated wastes. In
addition, the α emissions lead to significant variations in the neutron
emissions from the various fuels via (α,n) reactions with light elements
such as oxygen or fluorine present either as main constituents or trace
impurities in the fuel or cladding materials. These, as well as spontaneous
fission sources of neutrons, will be discussed in more detail below.

3.   FUEL HANDLING AND TRANSPORT

The radioactive emissions from reactor fuels are important in the con-
text of fuel handling and transport since they determine the decay heat
removal and shielding requirements. In the former case the relevant con-
sideration is the combined α, β and γ decay heating, which for irradiated
nuclear fuels is normally dominated by the fission product β and γ
emissions (see figure 2). As mentioned earlier, these are largely
independent of the fuel type and only in the case of plutonium fuels do the
actinides make a significant contribution to the nett decay heat commitment.

In the case of shielding requirements the emissions of interest are
the penetrating γ and neutron radiations which are the main source of doses
incurred during fuel handling and transport operations. The γ and neutron
emissions from the fuels included in this study as a function of time after

discharge from the reactor are presented in figure 3. Again, the γ decays are dominated by the fission products and show only minor variations with fuel composition, but marked differences occur in the case of the neutron emissions. For the reference UO_2 fuel the neutron production is dominated initially by spontaneous fissions of ^{242}Cm, with ^{244}Cm and ^{240}Pu taking over at cooling times beyond about 3 years, the (α,n) reactions accounting for only about 20% of the neutrons. The enhanced neutron production in the plutonium fuel is associated with its increased $^{242,244}Cm$ inventory and here again their spontaneous fissions are responsible for over 80% of the total neutron production. In contrast, the thorium based fuels exhibit much lower neutron production rates with 60 to 80% of the neutrons produced during the first year or so following discharge from the reactor arising from (α,n) reactions due to ^{238}Pu, and to a lesser extent ^{242}Cm and ^{228}Th and its daughter products.

The results presented in figure 3 show that the neutron emissions from the various fuels considered here range over approximately three orders of magnitude during the first few months post discharge and are thus an important consideration in the design of irradiated fuel handling facilities. This is emphasised by observation of the relative bulk of neutron and gamma shielding materials, approximately 3 cm of lead being required to attenuate 1 MeV γ rays by a factor of 10, compared with about 10-20 cm of hydrogenous material to produce the same attenuation for neutrons of a similar energy. In addition, the γ and neutron emissions can be an important consideration in the handling of unirradiated nuclear fuels, particularly those incorporating recycled plutonium or uranium isotopes derived from thorium. This is illustrated in table II which compares emissions from unirradiated and 10 day cooled fuels, both in absolute terms and relative to the irradiated reference UO_2 fuel.

In general, the γ emissions from the unirradiated fuels are small compared with those following reactor irradiation and for the plutonium fuel the main sources of γ radiation are the X-rays associated with α decays of ^{240}Pu, and the ^{241}Am daughter of ^{241}Pu. However, this is not a major shielding problem since X-rays are comparatively weak and in the main involve photon energies up to only 1-200 keV. Of greater radiological significance are the γ emissions from the thorium fuels which originate from small amounts of ^{232}U accompanying the ^{233}U incorporated during fuel fabri-cation. The α decays of ^{232}U, which has a radioactive half-life of 72 years

and represents 44 ppm by weight of the uranium content of the ^{233,235}U enriched fuel considered here, result in the production of ^{228}Th. This isotope is a member of the naturally occurring thorium radioactive series, which is illustrated in figure 4, and the ^{232}U leads to a gradual grow-in of the members of this series during storage of the fuel, with a resultant increase with time of penetrating γ emissions due to the production of ^{208}Tl and ^{212}Bi with γ ray energies of 2.62 and 0.73-1.81 MeV respectively. The build-up of ^{228}Th and its daughter nuclides with time via decays of ^{232}U is shown in figure 5, and is also reflected in the enhanced γ emissions from the ^{233,235}U enriched ThO_2/UO_2 fuel given in table II.

The importance of these effects may be illustrated by considering the γ dose rate at a distance of 1 m from a 1 kg fuel assembly for the various fuels listed in table II. For the 10 day cooled irradiated fuels this would be of order 500 R/hour, based on an extrapolation from measurements on irradiated magnox fuel elements, with corresponding values for the unirradiated magnox plutonium PuO_2/UO_2 and ^{233,235}U enriched ThO_2/UO_2 fuels of approximately 1 mR/hour and 5 mR/hour respectively. In addition, the latter γ dose rate would increase by about 30% after storage of the thorium fuel for a period of one year due to the presence of ^{232}U (see figure 5). Thus, although the absolute levels of dose rate quoted here are subject to some uncertainty associated with self-absorption effects and the influence of cladding materials in actual fuel assembly designs, these fuels would require γ shielding during storage and handling prior to reactor irradiation which is not necessary in the case of conventional ^{235}U enriched fuels. The plutonium based fuels may also require neutron shielding since the data presented in table II show that the neutron emissions from the unirradiated plutonium fuel are almost 20% of those from the irradiated ^{235}U fuel; these arise from spontaneous fissions of ^{240}Pu and (α,n) reactions associated with the α decays of ^{239,240}Pu and ^{241}Am present in the fuel.

Finally, the γ dose rate problems discussed above are also an important consideration in the design of fabrication facilities for plutonium and thorium fuels, leading to a requirement for the use of remote or semi-remote shielded handling techniques (see for example Weissert and Schileo, 1968). This is illustrated in figure 6 where the temporal variation of γ emissions at energies > 0.51 MeV for the U and Pu isotopes extracted from the reference ^{235}U enriched fuel during reprocessing at 200 days post discharge are compared with the corresponding emissions for the Th and U isotopes from the

ThO_2/UO_2 fuel. These results were obtained from γ spectra evaluated using the recently developed heavy element inventory code HYACINTH (Harte, 1976); the penetrating γ component of the uranium reprocessing streams in both cases arises from the ^{212}Bi and ^{208}Tl isotopes discussed earlier which grow-in during the first few years following separation due to ^{232}U decays. These isotopes are also responsible for the majority of γ emissions above 0.51 MeV for the thorium stream associated with the $Th/^{235}U$ fuel which show a more marked initial increase following separation originating in this case from the relatively short-lived ^{228}Th isotope (see figures 4 and 5), while the more gradual increase for the plutonium stream of the reference ^{235}U fuel is due to ^{241}Am formed via ^{241}Pu β decays.

In general the penetrating γ emissions associated with the Th and U isotopes of the $Th/^{235}U$ fuel are several orders of magnitude greater than those for the uranium and plutonium separated from the reference ^{235}U fuel. However, it is evident from figure 6 that if the separation and refabrication of $^{233,235}U$ fuels is carried out quickly the anticipated penetrating γ dose rates are not dissimilar to those from the plutonium stream of the reference ^{235}U fuel, and only in the case of the thorium stream is shielding significantly in excess of that encountered in the processing of conventional ^{235}U fuels required. Although not included in the present study, enhanced γ dose rates could also arise in the case of the ^{235}U fuel if recycle of depleted uranium were employed in order to improve the utilisation of the fissile ^{235}U isotope (Nunn, 1975); here the route for ^{232}U production is via decays of the 2.85 year half-life ^{236}Pu isotope formed during successive reactor irradiations (see table I). Thus the problem of increasing levels of penetrating γ radiation with time following separation of fissile materials is not confined solely to the thorium fuel cycle, although it is probably at its most acute in this case, and the curves shown in figure 6 emphasise the need for rapid processing and the elimination of long-term build-up of contamination within the fuel refabrication facilities.

4.   WASTE DISPOSAL ASPECTS

The waste disposal aspects of fuels originating from a range of commercial reactor designs of interest within the UK have been reviewed in a recent paper by Clarke et al. (1975), and the results of the present study are in agreement with the broad conclusions of that work. The decay heating commitment associated with the various fuel cycles considered here is

summarised in figure 2; on a timescale of tens of years this is dominated
by the fission product β and γ contributions, and in particular by the ^{90}Sr
and ^{137}Cs isotopes with radioactive half-lives of about 30 years. However,
in the long term the principal contributors are the actinide α emitters
which must be contained and isolated from man's environment for many hundreds
or thousands of years (Macdonald, 1976).

The long term α heating from the reference ^{235}U enriched fuel is
dominated by the isotopes of plutonium and ^{241}Am, or, following recovery of
the former for recycle, by ^{241}Am and ^{242,244}Cm in the separated wastes,
while for the magnox plutonium fuel the α levels are approximately an order
of magnitude greater with a much enhanced contribution from the curium
isotopes. These are produced via neutron capture reactions in the higher
plutonium isotopes during the reactor irradiation and this situation is
aggravated by storage of the fabricated plutonium fuel which permits the
build-up of ^{241}Am via ^{241}Pu β decays. The americium content of the fuel
prior to irradiation is an important factor in determining the subsequent
^{242}Cm build-up and may result in a further factor of five increase in the
waste α heating for long shelf-life fuels (Clarke et al., 1975).

The absolute levels of α heating associated with reprocessing wastes
from ^{235}U or plutonium fuels lie in the range from 1 to 10 kW per GW(e)yr
of electrical generation. The data presented in figure 2 indicate that a
reduction of two to three orders of magnitude in these levels may be achieved
by the use of thorium fuels. In these the main sources of long-term α
activity are ^{228}Th and its daughter products, together with ^{238}Pu, or in the
case of separated reprocessing wastes the 3.24×10^4 year half-life ^{231}Pa
isotope formed during the reactor irradiation. As an illustration of the
contributions to these α heating effects, figure 7 compares the actinide
activities as a function of cooling time for some of the more important
nuclides involved in the reference ^{235}U fuel and the mixed ThO_2/UO_2 fuels.
The variations in the activity levels with use of stored magnox plutonium
or the recycle of ^{232,233}U are also indicated; in general, the enhancement
of the plutonium and higher actinide activities in the former case is more
marked than that of the corresponding α-emitters present in the irradiated
thorium fuels. The only exception to this occurs in the case of ^{237}Np,
which builds up with repeated Th/^{233}U recycle, although this isotope is only
likely to represent a significant hazard in the separated reprocessing
wastes on a timescale of several thousands of years. However, as mentioned

elsewhere, a study of the influence of various fuel management options
would be required in order to specify in detail the importance of effects
of this nature in the recycle of plutonium or ^{233}U in a particular reactor
system.

In addition, the radiological hazards associated with the thorium fuel
cycle separated wastes are also significantly lower than those for conven-
tional ^{235}U or plutonium cycles at cooling times up to several thousands of
years (Clarke et al., 1975; Macdonald, 1976). These hazards, defined as the
volume of air or drinking water required to dilute the activity associated
with unit electrical generation to the recommended maximum permissible con-
centrations for exposure of members of the public (I.C.R.P., 1960, 1964) are
dominated by ^{90}Sr, ^{238,240}Pu and ^{241}Am, and are typically 10^{13} to 10^{17} and
10^9 to 10^{12} m^3/GW(e)yr for potential accidental discharges to the atmosphere
or the aqueous environment respectively.

However, the likelihood of the discharge of reprocessing wastes to the
environment must be relatively low, particularly once they are incorporated
for disposal into concrete, bitumen or glassy materials, and the main source
of human exposure will probably occur during fuel handling and processing
operations. In the latter instance active materials may be handled as
solutions or powders, possibly at elevated temperatures, and there is thus
an increased potential for exposure of operators via minor leakages or
spillage of materials. In the case of thorium fuels an added factor is the
presence of the gaseous radon isotopes which are present as members of the
naturally occurring radioactive series and may lead to the spread of con-
tamination associated with their solid daughter products, including the γ
emitting ^{208}Tl and ^{212}Bi isotopes mentioned earlier. These would lead to
increased doses during routine operation of plant and in the establishment
of thorium processing and fuel fabrication facilities it is important to
determine the radiological limits and monitoring procedures appropriate (see
Appendix I). These require techniques and experience not readily available
from the processing of natural uranium/plutonium fuels (Cofield, 1962; West,
1962), although the results of initial pilot experiments indicate that the
processing of thorium fuels with acceptably low operator doses is feasible
(Weissert and Schileo, 1968).

5.   CONCLUSIONS

In this paper, the results of a comparative study of the radiological

implications of plutonium recycle and the use of thorium fuels in advanced thermal reactor systems have been presented. The study does not represent a comprehensive assessment of the alternative fuel cycle options considered, but rather provides a demonstration of the type of information which may be obtained, and is intended more to establish guidelines along which detailed assessments of specific power programmes involving the use of recycled plutonium or thorium fuels should proceed. Such assessments would include consideration of specific reactor and fuel processing plant design features and fuel transport arrangements, as well as an examination of the influence of proposed fuel management schemes. The data obtained via this approach could then be used as part of the overall optimisation of future fuel cycle strategies aimed at a more efficient usage of natural uranium and thorium resources.

While the fission product inventories of the various fuels considered here are relatively insensitive to the fuel composition, the build-up of actinide isotopes depends strongly on this parameter, as well as on the details of the fuel management scheme employed. In recycled plutonium fuels the build-up of the higher transuranics, particularly the isotopes of curium, leads to significant α emissions which, together with spontaneous fissions, result in increased neutron emissions compared with conventional ^{235}U enriched fuels. These effects, which are important in the handling and processing of irradiated plutonium fuels, may be largely eliminated by the use of thorium fuels, but this is at the expense of increased γ dose rates during the fabrication and handling of the unirradiated fuel due to build-up of the naturally occurring radioactive series daughter products, notably ^{208}Tl and ^{212}Bi. For both plutonium and thorium fuels these problems are aggravated by prolonged storage of the fabricated fuel elements prior to reactor irradiation.

Finally, the actinides also play an important role in determining the long-term α decay heating commitment associated with fuel reprocessing wastes and here again the use of thorium fuels offers distinct advantages, although attention has been drawn to deficiencies in the basic nuclear data which are involved in predicting the build-up of these radionuclides.

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This appendix summarises the main areas of concern in the control of
health physics hazards associated with thorium fuels, based largely on an
analysis by Weissert and Schileo (1968):

(a) Criticality - this only arises when thorium is mixed with fissile
materials such as ^{233,235}U or plutonium. The hazards encountered are
similar to those arising in the case of enriched natural uranium or plutonium
fuels and are normally controlled via design features governing the storage
of the fissile materials and by administrative controls.

(b) Pyrophoricity - thorium and its compounds when finely divided,
such as in the form of powders or metallic swarf, are pyrophoric and thus
require special handling precautions. However, considerable experience
already exists, particularly within the U.K., in the handling of irradiated
uranium metal fuels which also exhibit pyrophoricity.

(c) Gamma and neutron activity - this gives rise to concern in the
case of thorium fuels containing ^{232,233}U and especially unirradiated
refabricated fuel (see Section 3). However, the dose rates involved are
much lower than those encountered in the handling of irradiated reactor
fuels and can be dealt with by adequate design provisions in the fuel
element fabrication and reactor handling facilities.

(d) Segregation of daughter products - this is a novel aspect of
thorium fuels whereby thorium, and the uranium isotopes produced from it,
are the parent nuclides of members of the naturally occurring radioactive
series. During reactor irradiation and subsequent handling and processing
this leads to the formation of their daughter products, some of which are
gaseous, while others readily segregate during particular operations
(Cofield, 1962).

As a consequence the health physics control of thorium fuels requires
special techniques not normally required in connection with conventional
uranium/plutonium fuels. Reactor irradiation coupled with chemical separa-
tions during fuel reprocessing and refabrication may disturb the daughter
product equilibrium and, as the maximum permissible concentrations in air
recommended by the I.C.R.P. for the various radionuclides involved range
over five orders of magnitude, the assessment of appropriate operational

limits is uncertain. Of particular importance is the ratio of the activities the two radium isotopes ^{224,228}Ra, which shows a marked dependence on time following thorium separation; the 3.64 day half-life α emitter ^{224}Ra builds up fairly rapidly via ^{228}Th decays, while the 6.7 year half-life β emitter ^{228}Ra builds up more slowly via decays of the very long lived naturally occurring ^{232}Th isotope (see figure 4). This necessitates the use of both β/γ and α counting techniques in the routine monitoring of contamination associated with the processing thorium fuels (West, 1962).

Another novel aspect of thorium fuels is the relatively long biological half-life of this element in man which influences the procedures employed in the personal monitoring. In general the levels of activity appearing in the excreta of individuals who have inhaled or ingested thorium compounds are below the limits of detection, and whole body γ counting techniques rather than for instance urine monitoring are needed. These techniques also enable the γ radiations due to the uranium isotopes which may accompany the thorium intake to be detected. A related problem is the metabolic behaviour of the thorium and uranium daughter products which is not well understood; current data normally assume that the daughters are retained in man in the same organ as the parent which in some situations leads to over-cautious estimates of the permitted body burdens for the latter isotopes (West, 1962).

Finally, careful assessment is also required in the control of inhalation hazards associated thorium fuels. The maximum permitted airborne concentrations recommended by the I.C.R.P. for occupational exposure to natural thorium and uranium isotopes when expressed as mass of material per unit volume are 0.02 and 0.2 mgm/m^3 respectively (Duggan, 1972). These levels are extremely low and are comparable with dust loadings encountered in many practical situations; indeed the typical natural aerosol levels found in an air-conditioned laboratory fitted with a filtered inlet system lie in the range 0.01 to 0.1 mgm/m^3, while those met in a factory environment might be of the order of a few mgm/m^3 or greater. Thus, although natural thorium and uranium have relatively low specific activities, it might prove difficult to demonstrate the absence of potential airborne hazards.

Thus, the novel aspects of thorium fuels require in general that they be handled in high integrity remote or semi-remote γ shielded facilities (Arnold, 1964). Routine health physics controls involve the use of continuous α and β/γ air sampling and monitoring of the working environment, while personal monitoring requires the provision of whole body counting

equipment. In addition, the build-up of contamination levels due to small quantities of uranium and thorium together with their daughter products during the lifetime of a facility requires careful assessment during the design phase; this is essential to minimise the problems encountered during decontamination and maintenance activities, as well as to avoid costly and difficult modifications to plant in later life once contamination levels have begun to build up.

TABLE I

FISSION PRODUCT AND ACTINIDE ACTIVITIES IN
100 DAY COOLED FUELS (Ci/gm)

Isotope	Half-life	6.75% ^{235}U enriched UO_2	5% magnox Pu enriched PuO_2/UO_2	9.5% ^{235}U enriched ThO_2/UO_2	9% ^{235}U 1.5% ^{233}U ThO_2/UO_2	Multiplier
^{90}Sr	28.1 y	0.95	0.42	1.27	1.29	x 10^{-1}
^{106}Ru	367 d	4.5	12.5	1.0	0.83	x 10^{-1}
^{131}I	8.05 d	2.0	2.4	2.2	2.2	x 10^{-4}
^{137}Cs	30 y	1.2	1.3	1.3	1.3	x 10^{-1}
^{144}Ce	284 d	1.2	1.0	1.5	1.5	x 10^{0}
^{228}Th	1.91 y	–	–	2.2	6.7	x 10^{-5}
^{231}Pa	3.25 x 10^4 y	–	–	1.3	1.3	x 10^{-6}
^{232}U	72 y	–	–	0.81	1.4	x 10^{-4}
^{233}U	1.62 x 10^5 y	–	–	1.2	1.6	x 10^{-4}
^{236}Pu	2.85 y	0.33	0.08	0.4	1.3	x 10^{-6}
^{238}Pu	87.4 y	0.52	5.3	0.61	2.0	x 10^{-3}
^{239}Pu	2.44 x 10^4 y	4.7	5.4	0.03	0.03	x 10^{-4}
^{240}Pu	6580 y	0.71	1.3	0.01	0.01	x 10^{-3}
^{241}Pu	14.3 y	2.0	6.7	0.01	0.01	x 10^{-1}
^{241}Am	433 y	0.28	2.4	–	–	x 10^{-3}
^{242}Cm	163 d	0.10	3.2	–	–	x 10^{-1}
^{244}Cm	18.1 y	0.11	4.1	–	–	x 10^{-3}

Fuel irradiation 44,000 MWD/T at 55 MW/T.

TABLE II

GAMMA AND NEUTRON EMISSIONS FROM UNIRRADIATED FUEL

AND FUEL COOLED FOR 10 DAYS FOLLOWING

IRRADIATION TO 44,000 MWD/T AT 55 MW/T

| Fuel	Cooling Time	Gamma Emissions (MeV/sec/gm)	Neutron Emissions (no/sec/gm)
a) Absolute Levels			
6.75% ^{235}U enriched UO_2	Unirradiated	8.6×10^{2}	2.1×10^{-2}
	10 days	3.2×10^{11}	1.3×10^{2}
5% magnox Pu enriched PuO_2/UO_2	Unirradiated	1.2×10^{7}	2.2×10^{1}
	10 days	3.5×10^{11}	4×10^{3}
9.5% ^{235}U enriched ThO_2/UO_2	Unirradiated	9.3×10^{3}	5.8×10^{-4}
	10 days	4.6×10^{11}	2.6
9% ^{235}U, 1.5% ^{233}U enriched ThO_2/UO_2	Unirradiated	1.5×10^{6}	5.5×10^{-1}
	10 days	4.6×10^{11}	5.4
b) Levels Relative to 10 day cooled ^{235}U Enriched UO_2 Fuel			
6.75% ^{235}U enriched UO_2	Unirradiated	2.7×10^{-9}	1.6×10^{-4}
	10 days	1.0	1.0
5% magnox Pu enriched PuO_2/UO_2	Unirradiated	3.7×10^{-5}	1.7×10^{-1}
	10 days	1.1	3.1×10^{1}
9.5% ^{235}U enriched ThO_2/UO_2	Unirradiated	2.9×10^{-8}	4.5×10^{-6}
	10 days	1.4	2×10^{-2}
9% ^{235}U, 1.5% ^{233}U enriched ThO_2/UO_2	Unirradiated	4.5×10^{-6}	4.3×10^{-3}
	10 days	1.4	4.2×10^{-2}
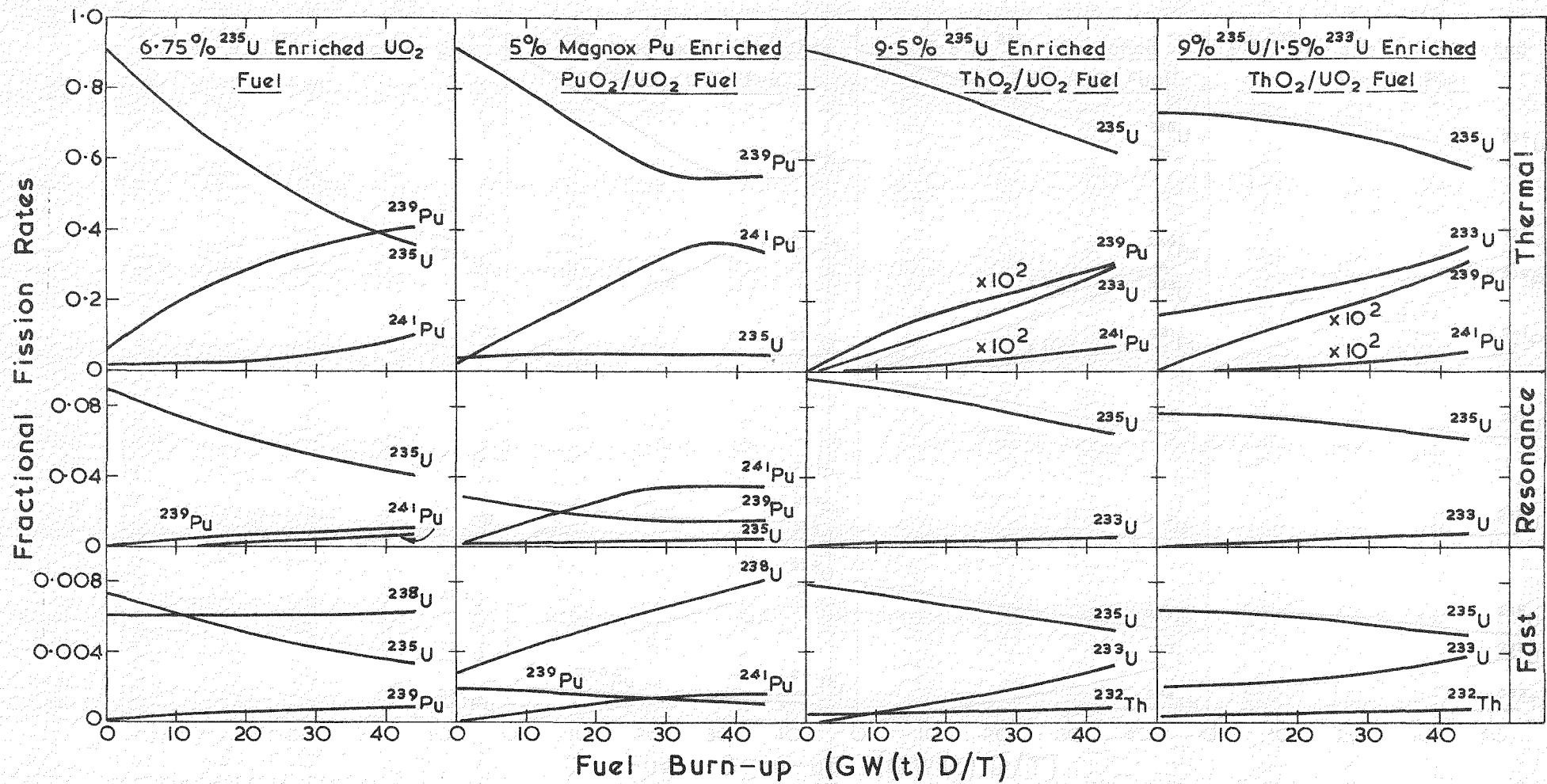
FIG. 1.   Fractional Fission Rates as a Function of Fuel Burn-up.
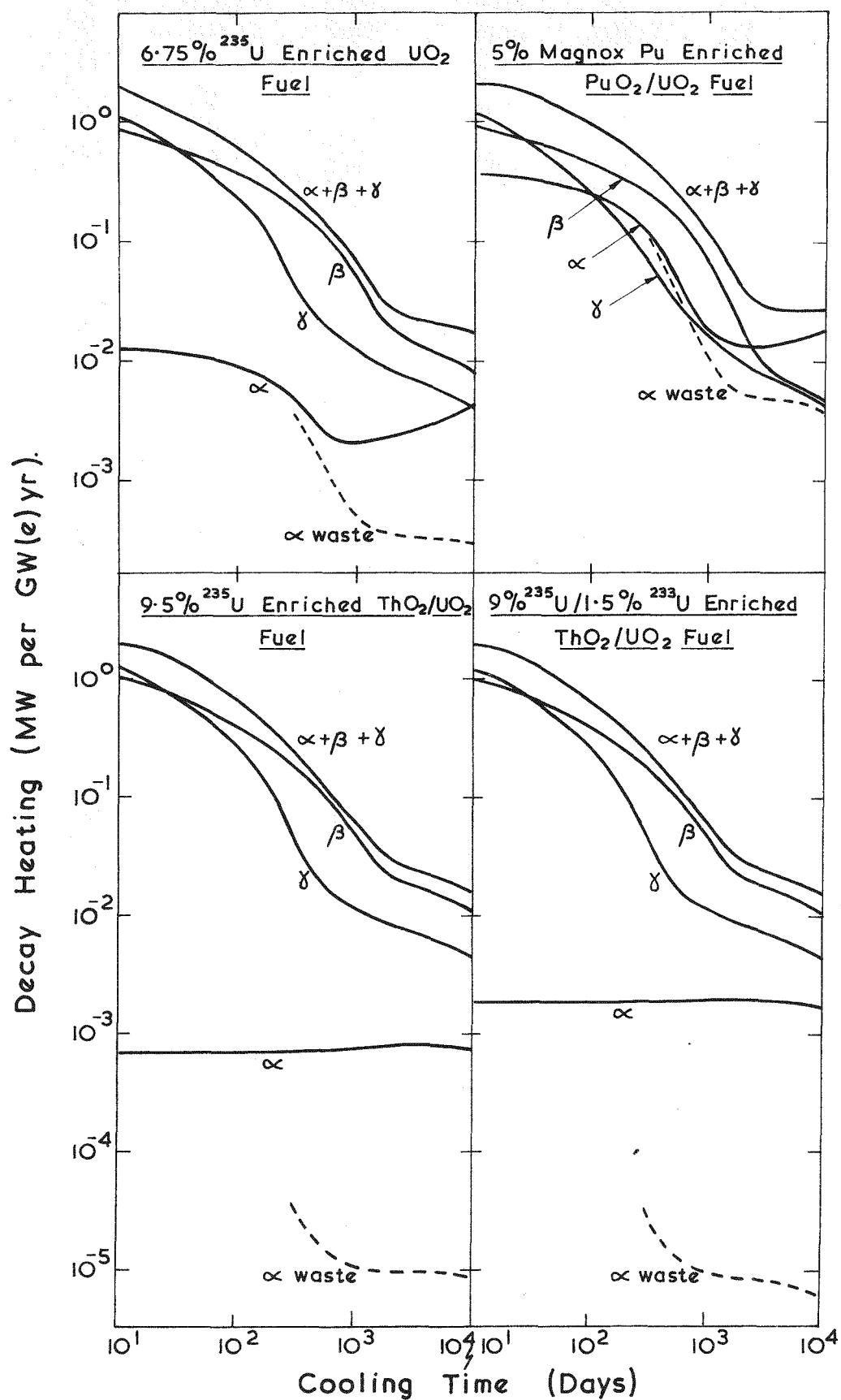
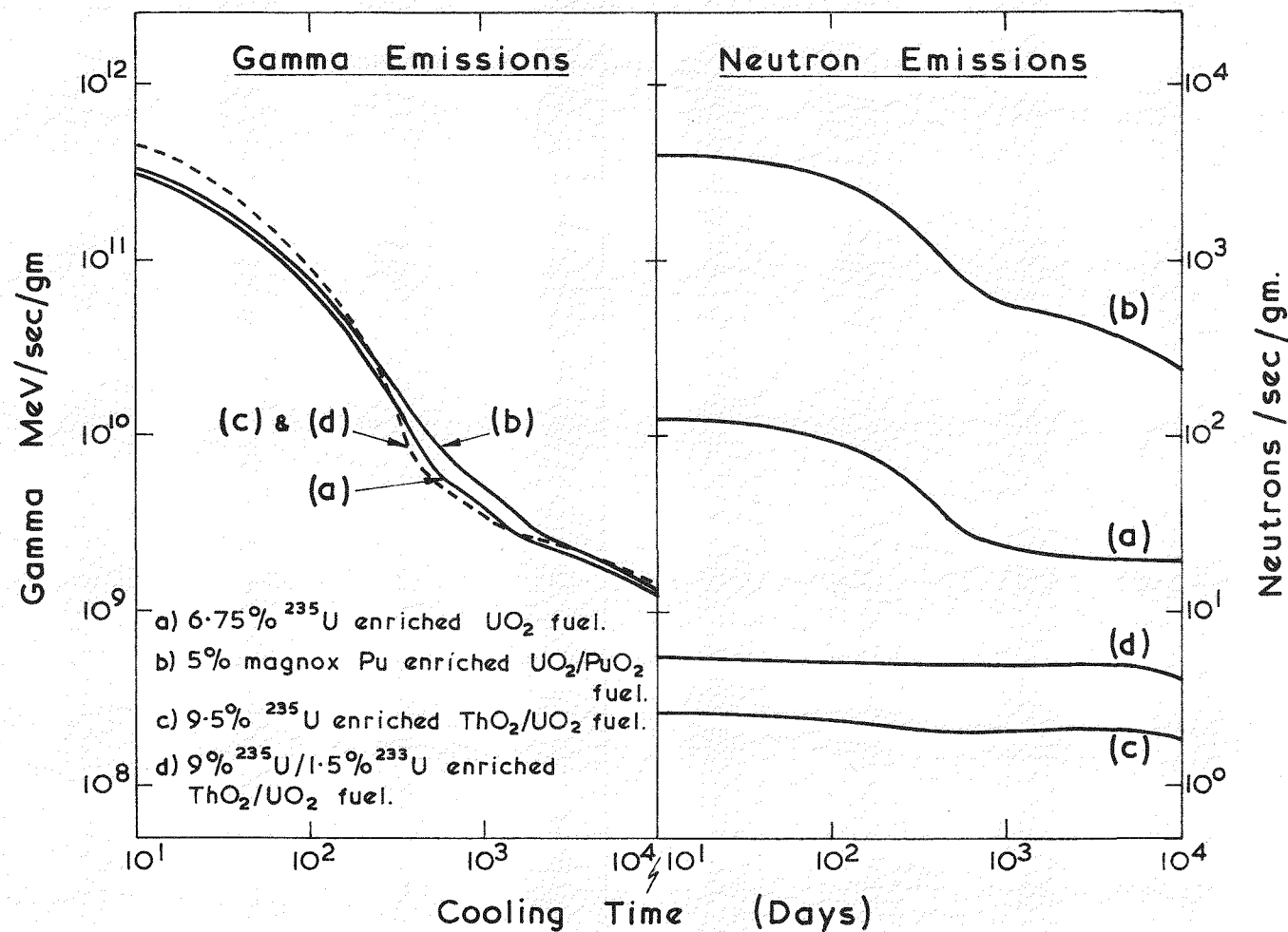FIG. 2. Decay Heating Commitment as a Function of Cooling Time for Irradiated Fuels.

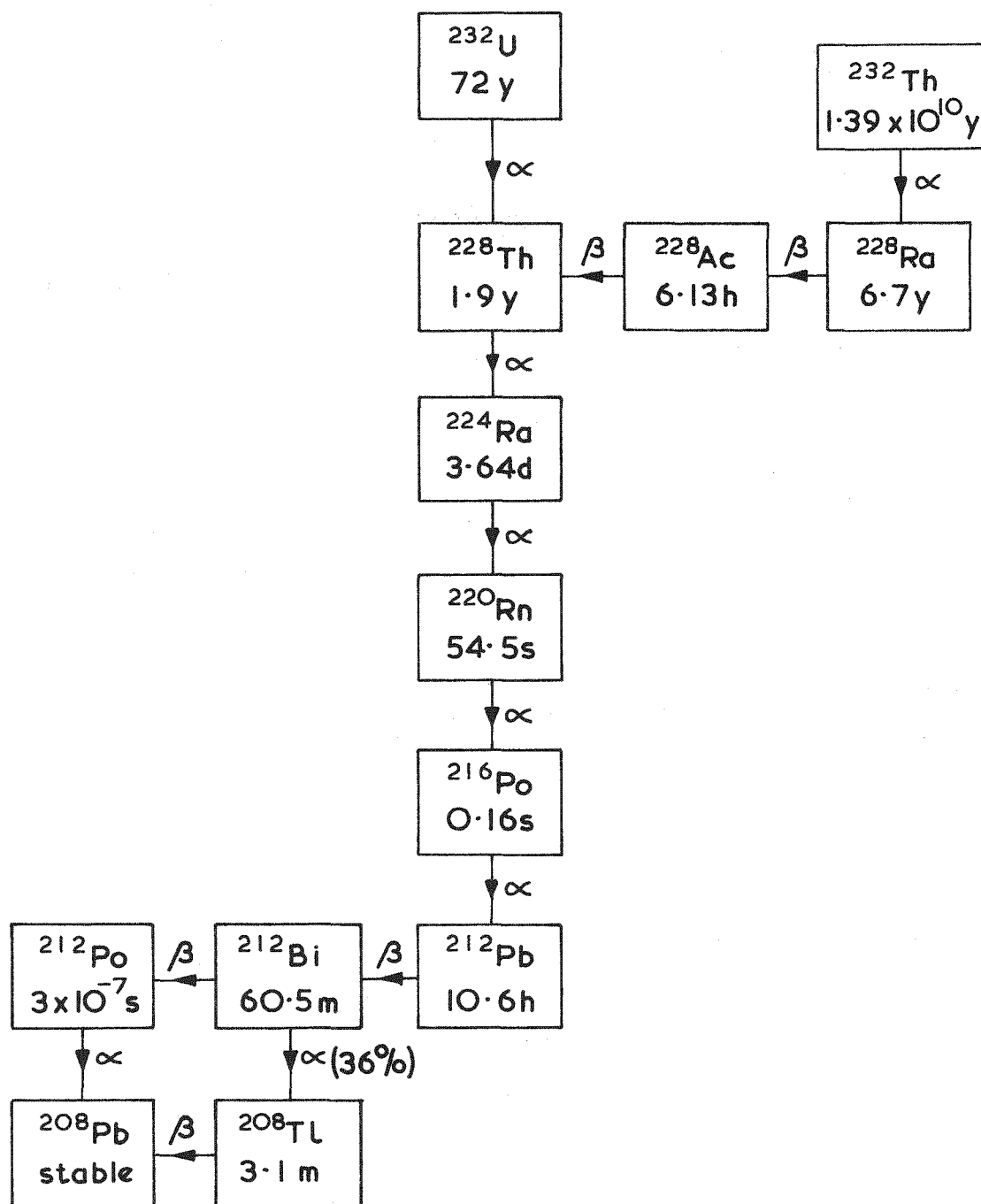FIG.3. Gamma and Neutron Emissions as a Function of Cooling Time for Irradiated Fuels.

FIG. 4. ^{232}U and ^{232}Th Natural Radioactive Decay Series.

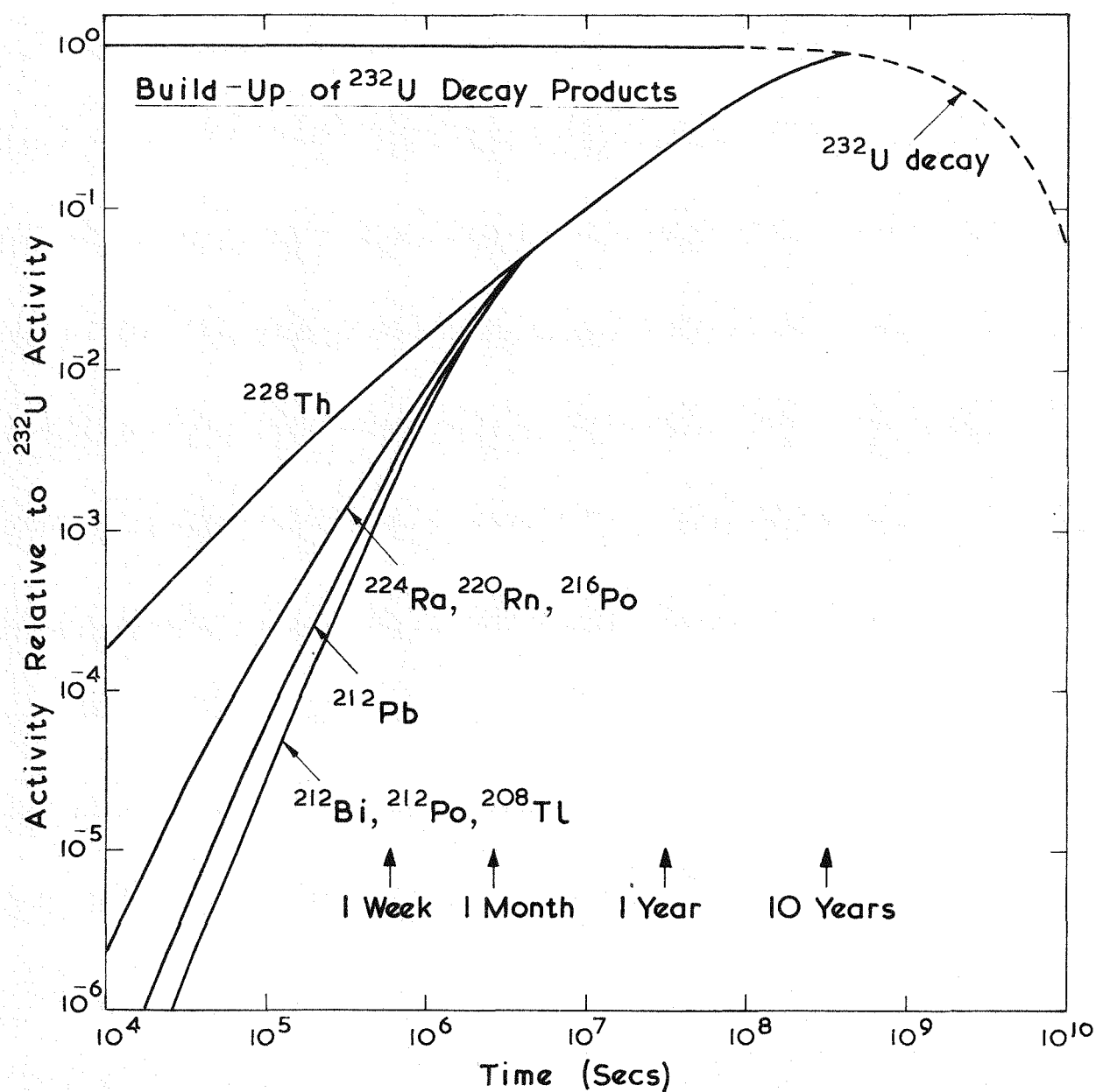FIG. 5. Build-Up of ^{232}U Daughter Product Activity as a Function of Time.

Penetrating gamma emissions from U,Th and Pu Isotopes separated from UO_2 and ThO_2/UO_2 fuels at 200 days cooling.

ThO_2/UO_2 Fuel- U stream

ThO_2/UO_2 Fuel- Th stream

UO_2 Fuel- Pu stream

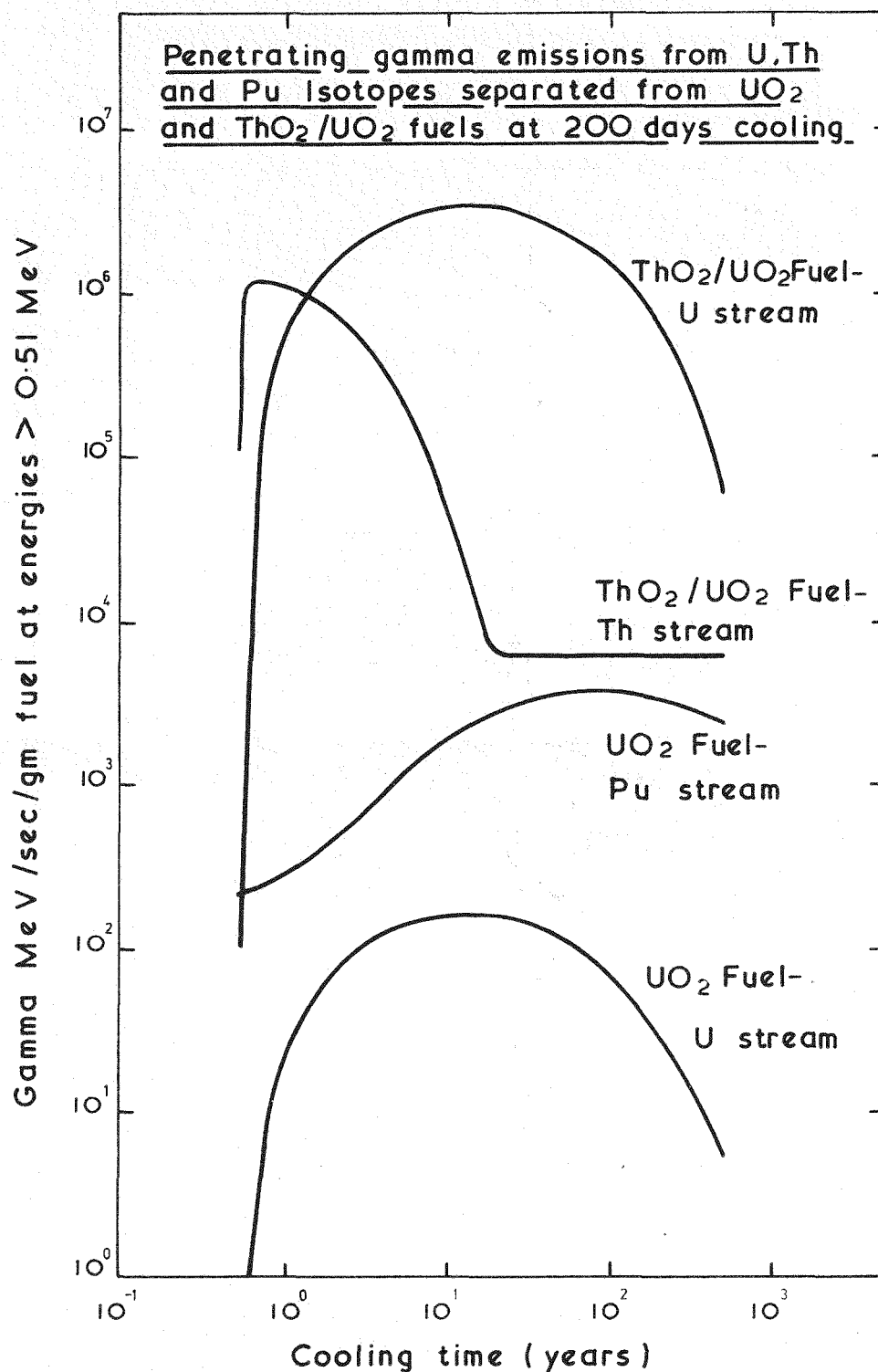UO_2 Fuel- U stream

Gamma MeV/sec/gm fuel at energies > 0.51 MeV

Cooling time (years)

FIG. 6. Gamma Emissions at Energies > 0.51 MeV from U, Th and Pu Isotopes in UO_2 and ThO_2/UO_2 Fuels as a Function of Cooling Time.
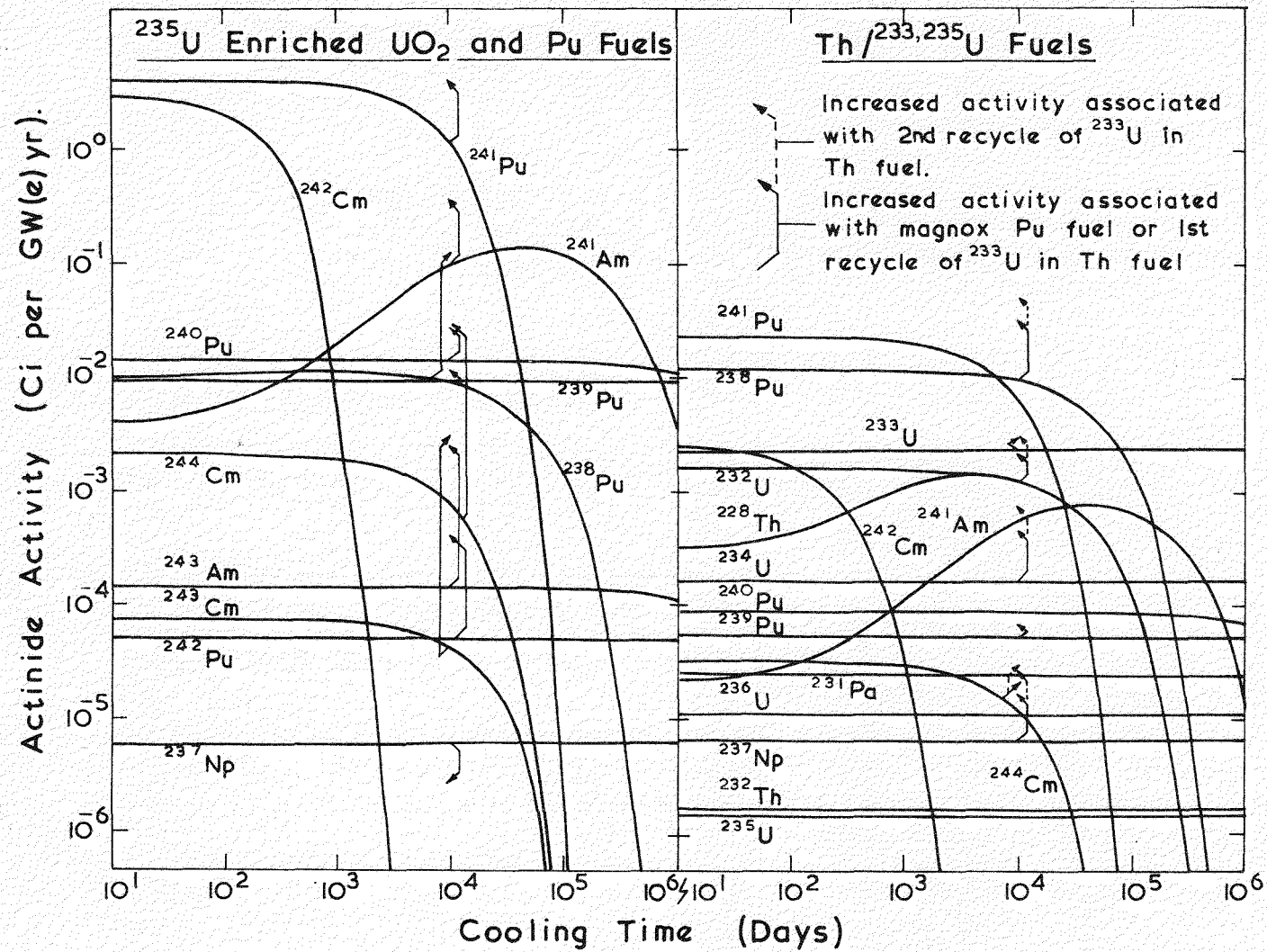
FIG. 7. Decay of Actinide Isotope Activity as a Function of Cooling Time for Irradiated Fuels.