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EUROPEAN ATOMIC ENERGY COMMUNITY - EURATOM

NON-FUEL APPLICATIONS OF α-EMITTING NUCLIDES

by

J. KOOI

1967

Transplutonium Elements Program

Directorate-General for Research and Training
Research Directorate

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SUMMARY

A critical survey is given of existing data concerning the application of heavy element α-active isotopes, other than their application as a new nuclear fuel. An attempt is made to extrapolate present data and trends, both from a technological as well as from an overall economic point of view. Attention has been drawn to a number of problems which urgently require further study.

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C O R R I G E N D U M

Page 23 paragraph 2 line 4 please read

" Sould be carried out in <u>isotope</u> production or experimental reactors "

Page 27 Table V

Fully enriched uranium	
<u>direct</u>	<u>via target</u>
$559^{c)}$	$0^{a)}$

NON-FUEL APPLICATIONS OF α-EMITTING NUCLIDES(+)

INTRODUCTION

In recent years tremendous progress has been made in our knowledge of the elements beyond uranium. The initial stage in the development of commercial nuclear power has made available substantia amounts of plutonium with important concentrations of isotopes heavie than 239. The development of fast breeder reactors and the assessment of their role in an integrated nuclear power system -heavily emphasizing the importance of plutonium as a nuclear fuel- requires detailed knowledge of some of the heavier elements. At the same time it has generally been realized that nuclear power generation will inevitably produce impressive amounts of potentially valuable heavy element by products.

At least equally important have been the research programs on transplutonium elements. These have not only provided basic knowledge on many dozens of nuclides of the elements beyond plutonium, but have also brought about the pilot scale synthesis of several kilograms of isotopes which are predestined to play an increasingly important role in the nuclear industry.

These developments have greatly stimulated interest in finding practical applications of certain isotopes of the heaviest elements. A number have been found and some of these offer indeed unique solutions to existing problems. In fact already in a number of cases today's demands are higher than what might possibly become available in years to come. In addition to the above one has become aware of the possibility that substantial benefits may be acquired by the nuclear power industry by actually producing isotopes which until recently were considered valueless and/or undesirable.

The aim of this report is first of all to present in a suitable form a representative selection of existing data pertinent to applications of heavy element isotopes other than that as a nuclear fuel. Secondly an attempt is made to correlate these data in order to facilitate extrapolating recent findings and ideas into a foreseeable future, both from a technological as well as from an overall economic point of view. Finally, it has been tried to define the problems which urgently require to be studied more extensively.

(+) Manuscript received on March 14, 1967.

Chapter I.

BASIC DATA

Apart from some very special isotopes discussed in chapter IX, the first group of isotopes considered in this report is of interest because of the fact that they emit high energy α-particles, the energy of which can be converted into useful forms. A second group consists of those nuclides which are starting materials for or precursors in the synthesis of the first group of isotopes. In some instances an isotope may be considered as belonging to both categories. For the purpose of this discussion the first group comprises the isotopes ^{238}Pu, ^{242}Cm and ^{244}Cm. All other isotopes discussed are considered as belonging to the second group.

Tables I and II present the characteristic data on both groups of isotopes.

Table I. Characteristic data of main Radioisotopic Heat Sources

Isotope	^{238}Pu	^{242}Cm	^{244}Cm
halflife, years	86.4	0.45	17.8
α-decay energy, MeV	5.45 (72%)	6.11 (74%)	5.80 (76%)
	5.36 (28%)	6.07 (26%)	5.76 (24%)
specific activity, Ci/g	17	3310	84
specific power, W/g	0.54	120	2.8
spontaneous fission occurrence	2×10^{-7}%	6×10^{-9}%	1.3×10^{-4}%
neutrons/sec per gram	2.9×10^{3}	2×10^{7}	1.1×10^{7}
thermal capture crosssection, barns	403	25	15
thermal fission crosssection, barns	18	<5	-
compound used	PuO_2	Cm_2O_3	Cm_2O_3
density compound, g/cm^3	10	11.75	11.75
active nuclide content in compound, % [a]	88	91	91
specific power of compound [a], W/g	0.47	109	2.55
power density of compound [a], W/cm^3	4.7	1280	30
max. body burden, uCi	0.3	0.2	0.3
M.P.C. in air, critical organ, uCi/cm^3	7×10^{-3}	4×10^{-11}	3×10^{-12}
shielding	minor	minor [b]	minor [b]
critical mass, kg	32	-	58

a) calculated for pure isotope

b) neutron shielding may be required

Table II

Characteristic data of Starting Materials and Precursor Nuclides

Nuclide	^{236}U	^{237}Np	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{243}Am
halflife, years	2.4×10^{7}	2.14×10^{6}	2.44×10^{4}	6.6×10^{3}	13.2	3.8×10^{5}	433	7.7×10^{3}
decay properties[a]	α 4.50 SF 10^{-7}	α 4.75 γ 0.03, 0.09 SF $<2\times10^{-10}$	α 5.15 SF 4×10^{-8}	α 5.17 SF 5×10^{-6}	β 0.02	α 4.90 SF 5×10^{-4}	α 5.49 γ 0.06, 0.03 SF 1.5×10^{-10}	α 5.28 γ 0.07
thermal capture cross-section, barns	9	170	286	290	390	19	620	184
thermal fission cross-section, barns	-	-	750	<0.1	1000	-	3	-
specific activity, Ci/g	6×10^{-5}	7×10^{-4}	6×10^{-2}	0.23	115	3.9×10^{-3}	3.45	0.19
neutrons/sec per gram	-	-	0.03	935	-	1670	-	-

a) energies given in MeV; SF: occurrence of spontaneous fission in %.

Chapter II

APPLICATION POSSIBILITIES

The data presented in Table I clearly demonstrate that ^{238}Pu and ^{244}Cm are very useful source materials for radioisotope heat sources. Due to their long halflives and the high energy of the emitted α-particles they are able to provide over long periods a constant heat output from small volumes. In common with other isotopic heat sources they require essentially no maintenance, which makes them very suitable for power supply units in remote and difficultly accessible locations. An added advantage, in particular of ^{238}Pu, is the absence of γ-radiation and of the need of shielding. Therefore, ^{238}Pu is ideal for applications in medical engineering.

Curium-242, on the other hand, is one of the two highest specific power nuclides under study for use in isotopic heat sources. It is of special interest that according to the most recent calculations Polonium-210, having a specific power of 141 W/gram, is more expensive by a factor of 1500[1]. For relatively short lifetimes ^{242}Cm seems to be an ideal base material for small volume - high power output sources.

As will be discussed later, ^{242}Cm is in many cases produced along with ^{244}Cm, thereby greatly reducing the favorable time-independence (only 16% decrease in 5 years) of the energy output of the latter nuclide. Most authors therefore provide for relatively long cooling time for ^{244}Cm to improve the ^{244}Cm quality. After sufficient cooling pure ^{244}Cm and ^{238}Pu are isolated. This not only results in loss of ^{244}Cm and in financial losses (storage and investment) but also sacrifices what might become considerable amounts of useful energy. As has been pointed out by Lang[2] heat utilization from ^{242}Cm is considerably improved by using isotopic devices containing more than one heat source, one of which is replaced at regular intervals. His calculations, given in Table III, show that the percentage of heat utilized may be improved tenfold under certain conditions.

Table III

Useful heat from device using ^{242}Cm

Number of sources	Interval for replacement of 1 source	Percent heat utilized
1	2 years	6.7
2	1 year	33.3
4	6 months	50.0
8	3 months	70.0

Lang also indicated that the presence of equal parts of ^{244}Cm in ^{242}Cm increases the heat utilization of ^{242}Cm from 6.7 to 9.0 percent over a 2 years period. It therefore seems to be quite clear, that curium, containing both isotopes should be used first for relatively short periods in deviced with successively decreasing power output and which may be serviced at regular intervals. After essential depletion of the ^{242}Cm the source material should be reprocessed to yield pure ^{244}Cm and ^{238}Pu for long life sources.

Several years ago the launching of a U.S. satellite equipped with an isotopic generator containing a kilogram of ^{238}Pu attracted considerable attention, but already in 1964 Seaborg[3] stated: "Huge requirements, difficult to comprehend by today's standards, will exist for ^{238}Pu, ^{244}Cm and other transuranium isotopes". Very recently, while the first massive demand estimates have been formulated (see chapter III), Seaborg[4] foresees requirements of tons of ^{238}Pu, alone for the U.S. space programs and much larger needs for applications in medical engineering. In addition, the utilization of tons of ^{244}Cm in a not too distant future is expected.

Indeed, many applications of isotopic heat sources for power generation have been proposed and a number of these are under development. The fact that no or very little maintenance is required combined with long life time and the high power density and, consequently, small volumes and weights enable in many instances unique solutions.

During the UKAEA/ENEA Symposium on "Industrial Applications for Isotopic Generators" a number of papers have been presented[5] which give a fairly complete survey of specific applications of isotopic generators in general. They may be summarized as follows:

1) Space applications

For propulsion and navigation in space isotopic generators seem to offer very attractive possibilities (see also ref.6). Power requirements, e.g. for communication satellites are in the order of 10-20 kW, corresponding to 80 - 160 grams of ^{242}Cm.

As regards production of on board power at least 8 satellites have been launched and first experience has been obtained. Power output requirements range from "smallest conceivable" to many kiloWatts (e.g. Stirling-machines). In this case, the advantages of isotopic generators will have to be fully developed for making them competitive with solar cells which are already in a more advanced stage of technological development.

2) Terrestrial communication applications

For power supply to relay stations (50 W to some kW), submarine cable repeaters and radio beacons on board airplanes and in landing systems isotopic generators will in many cases offer very attractive possibilities.

3) Remote or difficultly accessible locations

Many examples can be found were the inherent advantages of isotopic power sources will prompt their early applications. Amongst these may be mentioned automatic meteorological stations, undersea stations and marine buoys, all in the 0.1 to 100 W range. In the 100 W to 50 kW range undersea vehicles and dwellings may be near future customers.

Special mention may be made of the power supply for manned stations on the moon and other planets. For example, both for heating during the lunar night and cooling during the lunar day considerable amounts of power will be needed. For a long time to come the weight of the generators will have to be as small as possible so that nuclear reactors will not be feasible. Isotopic power sources and in particular those requiring little or no shielding seem to offer the only practical solution.

4) Medical applications, together with the use of small generators (in combinations with conventional ones) in instrumentation might likely become the main consumer of isotopic power. Numerous applications in instrumentation will be found, while some are already under study! Also, a ^{238}Pu-fueled pacemaker is under

development, whose power output will be in the order of 0.1 mW. The quantity of ^{238}Pu required -the only isotope which can be used- amounts to roughly 10 mg. Although it is very difficult to estimate annual demands, it is clear that a large market exists. As soon as satisfactory artificial organs have been developed the demands will be higher by several orders of magnitude.

Figure 1, page 9, partly taken from reference[7] summarizes the power spectrum covered by isotopic power generators. At left a number of devices under development or already in operation are indicated, together with the names of some of the firms which are active in this field.

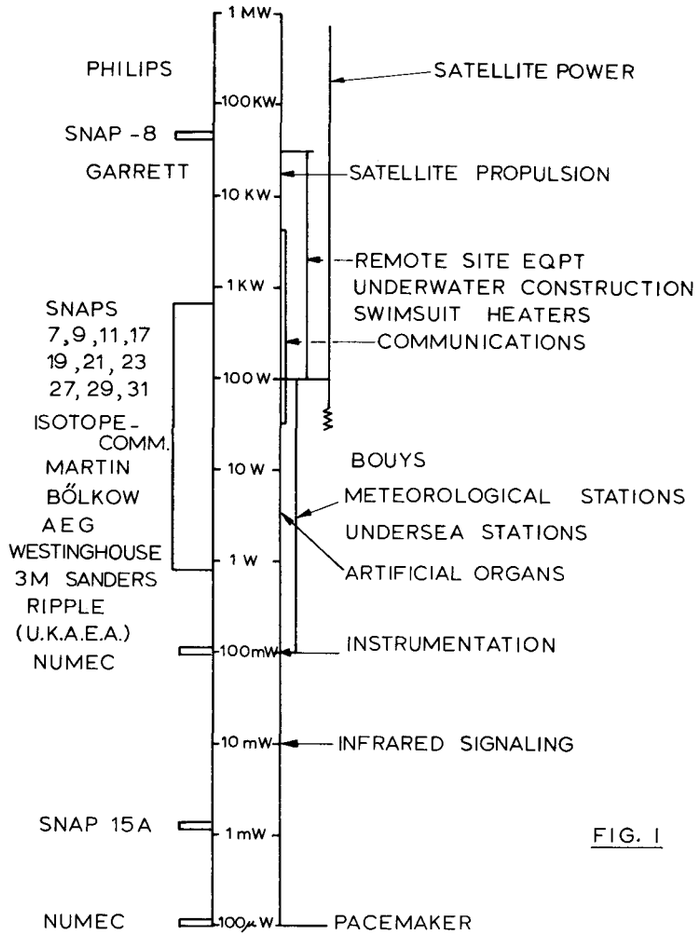


FIG. 1

Chapter III

DEMAND ESTIMATES

All nuclides discussed in this report have in the past twenty years proceeded from trace and microgram laboratory curiosity to the status of rare base material for industrial development. Some already are acquiring the characteristics of industrial pilot-scale raw materials. As has been pointed out this is due to their unique properties, but equally important for this fast development have been the spectacular developments in fields varying from space exploration and astronomy, meteorology and oceanography to biology and medical science.

As a consequence many demands for power supplies are beginning to be formulated, which could or should be met by isotopic power generators. Every month new projects are announced, stepping up the demands. Developments in medical engineering seem to be appearing which will surpass all other requirements.

Under these circumstances a realistic estimate of the future annual demand is impossible. Therefore only those demands will be reviewed here, which have been announced or can be derived from published data. Consequently this estimate has to be considered as a (very) low minimum. In addition, it will be clear that these figures concern almost exclusively the United States, the cradle of isotopic power generators, which may be considered to be several years ahead of other potential users.

Hennelly and Hood[8] quote 1965 N.A.S.A. estimates as amounting to 500 kW (thermal), equivalent to 1000 kilograms of ^{238}Pu before 1980. As much as 250 kW (thermal) might be needed annually of combinations of ^{238}Pu, ^{244}Cm, ^{210}Po and ^{147}Pm. No specific indication was given as to the preferred distribution over these four isotopes. However, from general considerations and other information it may be safely assumed that preference largely goes to ^{238}Pu and, in a later stage, also to ^{244}Cm. These figures have lately been confirmed by A.E.C. Commissioner Tape[9], who stated that NASA's estimated needs alone would be some 900 kg of ^{238}Pu by 1980. As an example, one of the specific sources reported under study is a 20 Watt ^{238}Pu-fueled generator for at least five years of operation, while it is also known that needs for one Transit satellite are in the order of 1 to 2 kgs of ^{238}Pu.

Recently it has been announced that the U.S. Navy may be buying several hundreds of SNAP generators in the next 4 to 5 years[10]. Most of these will be in the output range of upto 100 Watts, some going to 30 kWe for the Man-in-the-Sea programme. It is most likely that at least part of these will be fueled by ^{238}Pu. A 500 Watts swimsuit heater, fueled with the same isotope, is due to be delivered early in 1967. A rough estimate of ^{238}Pu needs derived from these data gives a probable cumulative demand of several 100 kilograms for the early seventies.

An estimate for the demands to be expected from medical applications must necessarily be based on an estimated incidence rate of illnesses, organ defects etc. and the possibilities of developing reliable instruments. It has been stated that a pacemaker requires about 10mg of ^{238}Pu. It seems that already at present about 2 pacemakers are installed annually per 10.000 people in urban areas in the western world. Assuming that electronic and medical technology would be perfected so that the use of ^{238}Pu as power source would be warranted, this would mean that Western Europe and the United States would need in the order of 1 kilogram of ^{238}Pu per year for the fabrication of pacemakers alone.

As mentioned above, however, many experts expect the successful development of fully artificial organs, as e.g. hearts and kidneys. In the United States several teams are working on these projects and the successful operation of implanted artificial hearts has been reported several times. The power needs for these instruments are rather important and they seem to require in the order of several hundred grams of ^{238}Pu. Therefore, even if the incidence rate of the application of artificial hearts will be in the order of one percent of that of pacemakers, the western world alone would require several hundred kilograms of ^{238}Pu per year, only for this one application. On a world basis and taking account of the development of other artificial organs many tons would be needed annually.

Up to now no estimates, other than those of NASA, have been made for ^{242}Cm and ^{244}Cm. The main reason is, of course, the less developed technology and the fact that much less is known about the prospects of large scale production. Again, it should be stressed that specific demands will certainly be formulated with developing technology.

Chapter IV

PRODUCTION CHARACTERISTICS

In this chapter the data will be reviewed which govern the synthesis of the three nuclides of primary interest - ^{238}Pu, ^{242}Cm and ^{244}Cm - under different irradiation conditions from available starting materials. The influence of variations in nuclear physics data will be considered. In the next chapter different production methods will be discussed, in particular the influence of factors such as reactor cycle, burn up, recycling, etc.

Figure 2 illustrates the paths along which ^{238}Pu may be obtained, either in nuclear fuel or in separate irradiations of the base material. Figure 3 presents the same information for ^{242}Cm and ^{244}Cm. As can be seen from Fig. 2 the main path for the production of ^{238}Pu goes from ^{235}U via ^{237}U through ^{237}Np. A minor contribution to the ^{237}U yield comes from the (n,2n) reaction on ^{238}U. ^{238}U, however, is the starting material for the production of ^{239}Pu via ^{239}Np. It will therefore be clear that, if fuel is used with no or very little ^{238}U, the resulting plutonium will be high in ^{238}Pu (high Q)*. Fuel mainly consisting of ^{238}U will result in plutonium with low Q-value and thus unsuitable for direct use in isotopic generators. Such fuel, therefore, will require separation of the neptunium and its subsequent irradiation. It will also be clear that any ^{238}Pu produced and subjected to neutron irradiation will be isotopically diluted with undesirable heavier isotopes. The (n,2n)-reaction on ^{239}Pu has generally been neglected. Its contribution to the yield of ^{238}Pu will be small and its influence on the quality of ^{238}Pu negligible. Also the (n,2n)-reaction on ^{237}Np, giving ^{236}Pu, has not been indicated, although its influence on the quality of ^{238}Pu may in some cases be important through the formation of the γ-active daughter product ^{208}Tl. On the other hand, very pure ^{238}Pu may be obtained from the decay of short lived ^{242}Cm, even if separated from mixtures with ^{244}Cm, due to the relatively long half-lives of the latter isotope and its daughter ^{240}Pu.

From Fig. 3 it is clear, that the production of ^{242}Cm and ^{244}Cm takes place simultaneously in the irradiation of plutonium in a nuclear reactor. The production scheme involves the successive capture of a number of neutrons and is more complicated because of the

* The quality, Q, of ^{238}Pu may be defined as the percentage of ^{238}Pu in the total plutonium obtained: $\frac{\text{weight of } ^{238}Pu}{\text{total weight of all Pu-isotopes}}$.

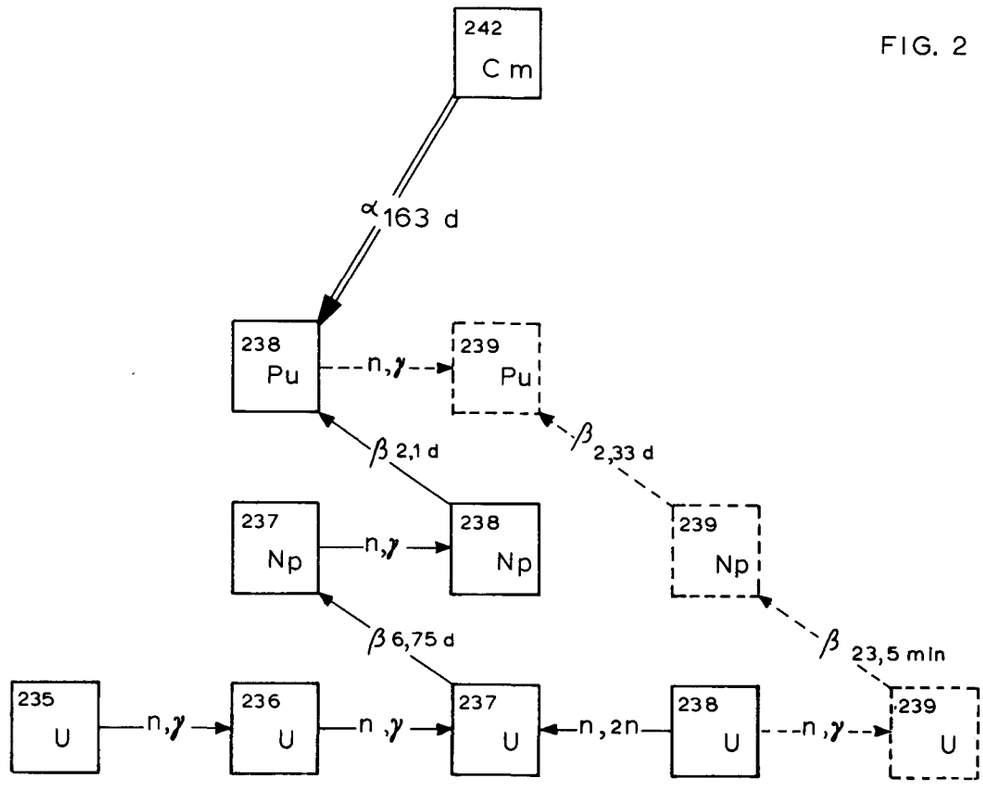

242 Cm
α 163 d
238 Pu
n,γ
239 Pu
β 2,1 d
β 2,33 d
237 Np
n,γ
238 Np
239 Np
β 6,75 d
β 23,5 min
235 U
n,γ
236 U
n,γ
237 U
n,2n
238 U
n,γ
239 U

FIG. 2

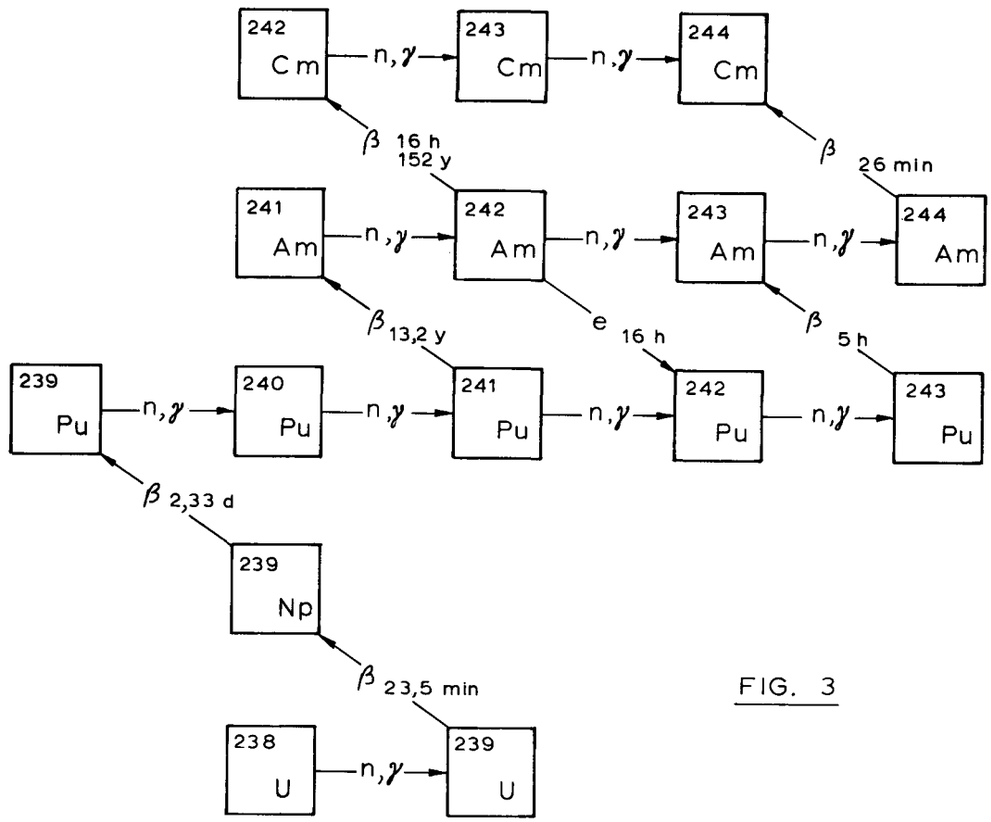

242 Cm
n,γ
243 Cm
n,γ
244 Cm
β 16 h 152 y
β 26 min
241 Am
n,γ
242 Am
n,γ
243 Am
n,γ
244 Am
β 13,2 y
e 16 h
β 5 h
239 Pu
n,γ
240 Pu
n,γ
241 Pu
n,γ
242 Pu
n,γ
243 Pu
β 2,33 d
239 Np
β 23,5 min
238 U
n,γ
239 U

FIG. 3

occurrence of a number of side- and decay back-feed reactions.

It is also clear that the composition of the initial plutonium starting material will greatly influence final product composition and the time required for producing a certain quantity of curium per unit weight of plutonium. The composition of the final curium product, in the case of ^{244}Cm again expressed in terms of product quality, Q, will also considerably vary with cooling time and time required for processing and source fabrication. (c.f. also chapter II)

Relatively pure ^{242}Cm can be obtained by irradiating pure ^{241}Am, resulting from the decay of ^{241}Pu during longer periods. Pure ^{244}Cm may result from the irradiation of ^{243}Am or the irradiation of ^{241}Am and ^{242}Cm during sufficiently long times in order to assure the disappearance of these two nuclides. With $Q^{244}Cm$ = 98% the drop in the heat output during the first year of operation amounts to 35%, while ^{244}Cm may be considered satisfactory for constant output long term operation if the ^{242}Cm content is not higher than 0.25%. As is well-known from the transplutonium production programs, however, very high values of integrated flux (several times 10^{22} nvt) are required to achieve this purity by irradiation of ^{241}Am.

In Fig. 2 and 3 no data are included on the cross-section (σ) for the different reactions. As is well-known fairly large uncertainties and discrepancies still exist in cross-section data for these heavy elements as found in literature. Interesting examples may be found by comparing the basic data used for the calculation of the yield of transplutonium elements by different authors. For detailed information see the reports quoted in reference 11).

Although in some cases large deviations also exist in the experimental values of specific cross-sections, by far the largest uncertainty is due to the lack of knowledge concerning the energy-dependence of σ. Experimentally determined data usually concern either thermal neutron cross-sections, 2200 m/sec cross-sections or pile cross-sections. In most calculations however, one wants to take into account the possible epithermal and fast thermal (resonance) contributions, in order to evaluate an "effective" cross-section for a specific reaction in a specific (although often very poorly defined) neutron spectrum.

The neutron energy dependence is, in general, important in two aspects. First of all sometimes very large resonance capture may occur in the epithermal energy range, thereby greatly enhancing the (n,γ) reaction. Secondly, induced fission cross-sections often decrease with increasing neutron energy, thereby increasing the effective $\alpha = \frac{\sigma_{capture}}{\sigma_{fission}}$ of a specific nuclide. This not only decreases material losses but also fission product activity, thus improving overall reaction yields and facilitating reprocessing.

As will be seen from the data presented in chapter VI a harder spectrum, like that of a F.W.R., is beneficial to the production of higher actinides as compared with the somewhat softer spectrum of a BWR-type reactor. Similarly it is expected that the production of transplutonium isotopes in the somewhat harder spectrum of H.F.I.R. at Oak Ridge, will surpass the yields calculated on the basis of yields obtained in the M.T.R.[12] Also Epel[13] has early drawn attention to the beneficial effect of a larger epithermal component for the production of heavy elements while a dramatic demonstration has been given by Ice[14], who indicates a 100-fold increase in ^{252}Cf production by irradiating ^{242}Pu in a socalled resonance reactor.

A very interesting table (Table III) has been given by Deonighi and Eschbach[1] clearly demonstrating the importance of resonances and of flux ratio $\emptyset = \varphi_1/\varphi_2$ = epithermal flux / thermal flux. If only 2200 m/sec cross-sections had been used, the yields of the nuclides given would have been grossly underestimated (e.g. also data from Table II).

As may be seen, exploiting the influence of $\emptyset$ on the key base materials leads to considerably higher product yields, while product losses are not increased or even lowered.

In general it may be said that also an increase in average neutron temperatures enhances product yields.

Table III

Influence of flux ratio Ø *

Nuclide	Effective Cross-section (barns)					
	Ø = 1	Ø = 3	Ø = 6	Ø = 1	Ø = 3	Ø = 6
^{235}U	602	603	597	0.19	0.22	0.25
^{236}U	23	68	11.8			
^{238}U	5.0	9.3	14.4			
^{237}Np	180	221	260			
^{238}Pu	453	425	397			
^{239}Pu	1565	1759	1914	0.50	0.53	0.55
^{240}Pu	559	1215	1981			
^{241}Pu	1602	1601	1573	0.37	0.38	0.39
^{242}Pu	88	276	497			
^{241}Am	874	1248	1605			
^{242}Am	7220	7201	7140			
^{243}Am	146	340	553			
^{242}Cm	18	18	18			
^{244}Cm	14	14	13			

* The effective cross-sections include the effects of thermal flux depression, increasing with increasing Ø. The fuel enrichment at each Ø was sufficient to attain a fuel exposure of 25 Mwd/kg.

It is evident that an increase in flux, ψ, will, in first approximation, correspondingly increase the product yield of a specific reaction per unit of time. This may be essential in overcoming certain bottle-necks in the production paths of a number of the heaviest isotopes. A large amount of data and very extensive calculations have demonstrated this in great detail in connection with the transplutonium element programs for which reference may be made to the detailed reports[11]. However, since also the product nucleus is subjected to the higher flux the yield of higher isotopes and the losses due to fission and side reactions increase also. Moreover since, especially the first irradiations for obtaining the immediate precursors of the desired nuclides will have to be carried out in power reactors with rigid long period fueling schemes an increase in 'results usually in an increased burn-up of starting materials, sometimes in an increased yield of product but always in a (much) lower product quality. A number of examples have been quoted in literature. Figure 4 presents data from Lang[2] for Q of ^{238}Po for P.W.R. Pressurized Water and Heavy Water reactors, respectively as a function of flux and irradiation time.

Chapter V

PRODUCTION METHODS

The production of the α-emitting nuclides under discussion will become naturally integrated in the commercial production of nuclear power. This not only because the financial profits are sufficient to incite reactor operators to include this production in their power production schemes, but also because only in that way the supply of the gratuitously or purposely produced material will possibly become sufficient to meet the rising demands.

As will be clear from the discussion in chapter IV the production of these nuclides may be considerably enhanced by exploiting changes in physical parameters through modification of reactor characteristics. Examples mentioned include reactor design to maximum α, spectrum hardening and flux modification by increasing enrichment or change in fuel element density. However, also without introducing design changes the operation scheme and in particular the fuel cycle management of power reactors may be altered in such a way that very important gains may be obtained. In this connection especially fuel and target burn-up and recycle must be considered. In addition, the choice of which combination of reactors for the production of base material and for its final irradiation is the most attractive requires careful study. Both aspects will be discussed in this chapter.

Again it must be stressed that often considerations regarding the quality of the desired product may play an important role. For instance, if a ^{238}Pu quality of 65% is considered acceptable, it is possible to obtain it directly from the reprocessing of 93% enriched uranium fuel without intermediate recovery of the neptunium. On the other hand, ^{238}Pu contained in the plutonium obtained from reprocessing of less enriched uranium or plutonium fuel, has no other value than that as precursor for ^{239}Pu, or simply a negative value if regarded as an undesirable diluent of ^{239}Pu. Similar considerations are valid for the case of ^{238}Pu obtained from decay of ^{242}Cm, which may be contaminated with ^{240}Pu. The case of ^{244}Cm dilution by ^{242}Cm has been discussed before.

From this it will be clear that the future development of the market for isotopic generator source material may influence the choice of the fuel management scheme finally adopted by a particular reactor

operator. For the purposes of this discussion, however, it is assumed that the aim should be the production of maximum possible quantities of the highest attainable quality.

Of direct interest are also questions of time, closely related with fueling schemes. First of all it may be pointed out that in general the product material will become available only three years after the start of the first irradiation. This may put an additional investment load on the reactor operator which should be matched by later profits. Another, although similar, situation is found in the possible hold up of plutonium outside of the reactor, to allow for the decay of ^{241}Pu in view of ^{241}Am recovery. Time is also one of the major aspects of concern in questions of purity of starting material for curium production, since the $^{241}Am/^{243}Am$ ratio is very time dependent at a given flux. It will also be evident that production of ^{242}Cm from ^{241}Am will in general not be compatible with normal power reactor operations, since it would require loading or even refueling operations at much too short intervals.

The same argument applies in general to the irradiation of ^{237}Np, ^{241}Am and ^{243}Am if only yield and flux are considered. A very illuminating example has been derived from one of the studies on the optimalization of the production of α-emitting nuclides. Figure 5, constructed from data given by Lang[2], shows the total production of ^{238}Pu, ^{242}Cm and ^{244}Cm in 4 years resulting from the irradiation of a mixture of 600 grams of ^{241}Am and 400 grams of ^{243}Am in a PWR as a function of the length of the irradiation cycle and for different flux values. After each irradiation a four months cooling period was allowed prior to separation and target refabrication.

First of all, as demonstrated in the same report, the harder spectrum of a PWR produces in short irradiations more α-emitting nuclides than were produced in the softer BWR spectrum for equivalent or long irradiation times. Secondly, it is clear from Fig. 5 that yields of ^{242}Cm, ^{238}Pu and ^{244}Cm at the lowest flux drop sharply after a very short period of irradiation (incompatible with normal power reactor operation). In the case of ^{244}Cm, which constitutes the effective endpoint of this irradiation, time dependence is less marked after the first half-year at high fluxes. The most interesting point to note, however, is the fact that ^{244}Cm production is by far the highest at a flux of 2.10^{14} $n/sec^{-1}cm^{-2}$ with irradiation periods of from 6 to 12 months.

FIG. 4

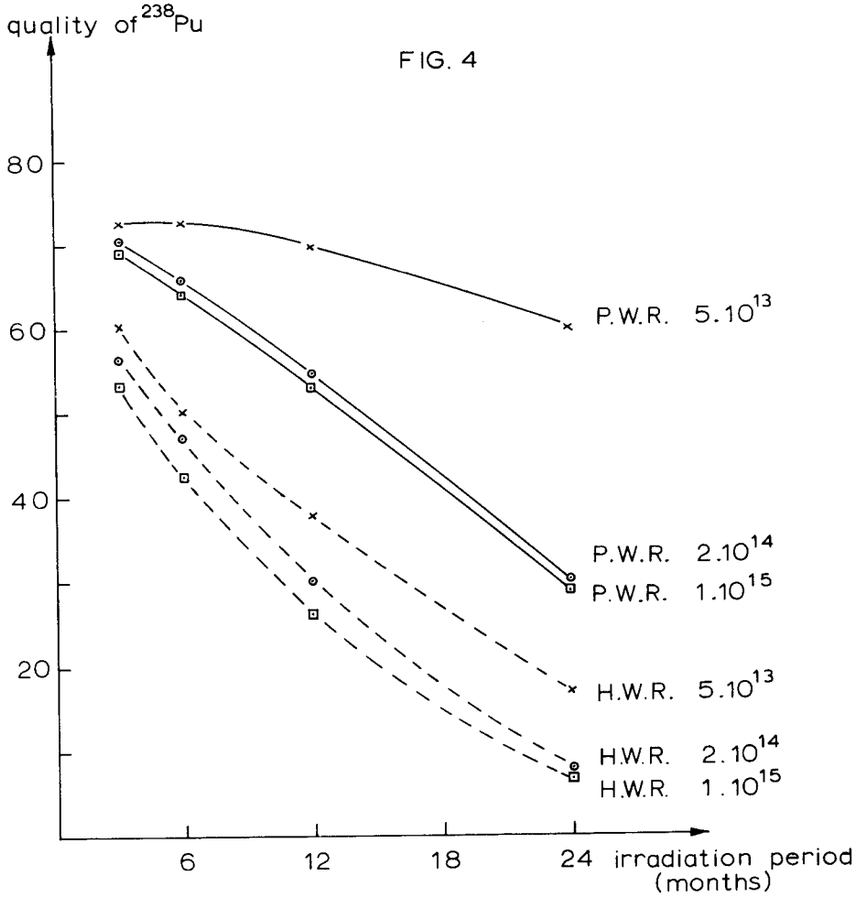
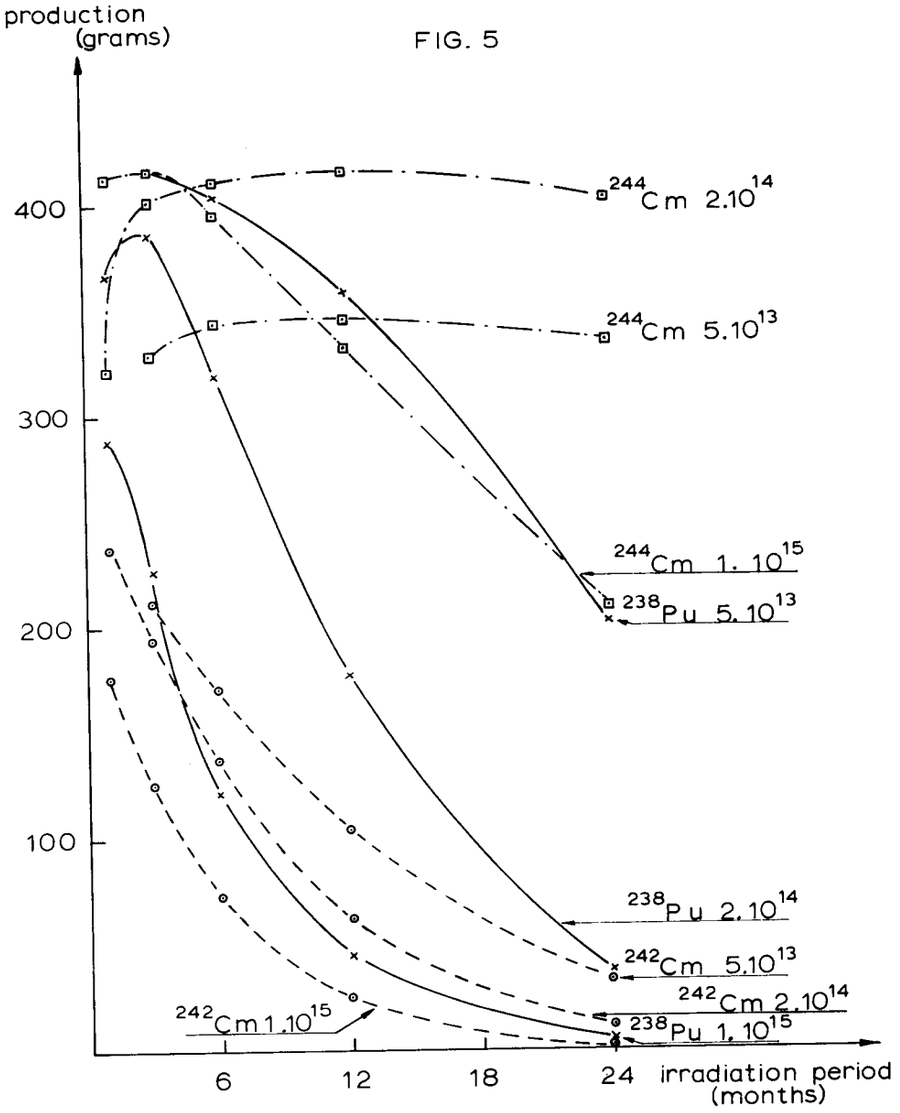

quality of ^{238}Pu
80
60
40
20
6
12
18
24
irradiation period (months)
P.W.R. 5.10^{13}
P.W.R. 2.10^{14}
P.W.R. 1.10^{15}
H.W.R. 5.10^{13}
H.W.R. 2.10^{14}
H.W.R. 1.10^{15}

FIG. 5


production (grams)
400
300
200
100
6
12
18
24
irradiation period (months)
^{244}Cm 2.10^{14}
^{244}Cm 5.10^{13}
^{244}Cm 1.10^{15}
^{238}Pu 5.10^{13}
^{238}Pu 2.10^{14}
^{242}Cm 5.10^{13}
^{242}Cm 2.10^{14}
^{238}Pu 1.10^{15}
^{242}Cm 1.10^{15}

As a second example of the influence of time and flux Figure 6, based on data given by Meichle and Owsley[15], presents the total amounts of ^{238}Pu produced from ^{237}Np as a function of neutron flux for different campaign-times. Starting with one kilogram of ^{237}Np and adding at the beginning of each year one kilogram of fresh ^{237}Np, the total quantity of ^{238}Pu produced has been calculated for total periods of 5, 10, 15 and 20 years. Curves are given for 2 conversion efficiencies. Figure 6 also demonstrates, for the case of ^{238}Pu production from ^{237}Np, that the optimum flux for this conversion is in the range of from 1 to 3 times 10^{14} n/sec cm^{-2}. The optimum becomes more pronounced with higher depletion of the base material.

These two figures illustrate very well the conclusion reached in many instances that in order to achieve optimum production the conversion of the base material, obtained from power reactor operation, should be carried out in production or experimental reactors. These offer the right flux and have the (flexibility in) fuel cycle management schemes to permit the optimum irradiation time to be used. Moreover, these reactors usually offer many more possibilities for adjustment of neutron spectrum and (temporary) variations in flux level, which may be required.

Although an increase in burn-up or depletion of target material which corresponds with longer fuel cycles may have only minor effects on the yield of the product per unit quantity target material, economic considerations may be important. Less frequent recharging operations, smaller material throughputs and hold-up and less frequent reprocessing cycles may contribute to make the production of α-emitting isotopes more easily feasible. In addition, larger depletion may make the desired product earlier available and permit to reach equilibrium conditions at an earlier moment. However, any beneficial effect of higher burn-ups has to be balanced against their effect on the product quality. Longer exposure will, e.g. in the irradiation of ^{237}Np, tend to produce more of the higher plutonium isotopes. Figure 7, prepared from data of Meichle and Owsley[15], shows the influence of ^{237}Np depletion on the quality of ^{238}Pu at different flux levels.

Usually studies on fuel recycle only concern its influence on the optimum use of fissile nuclides. In regard to the production of α-active nuclides for isotopic power generators fuel recycle plays

FIG. 6

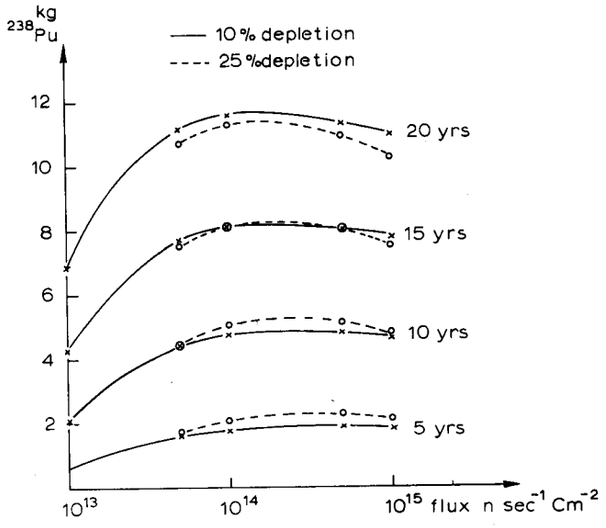


FIG 7

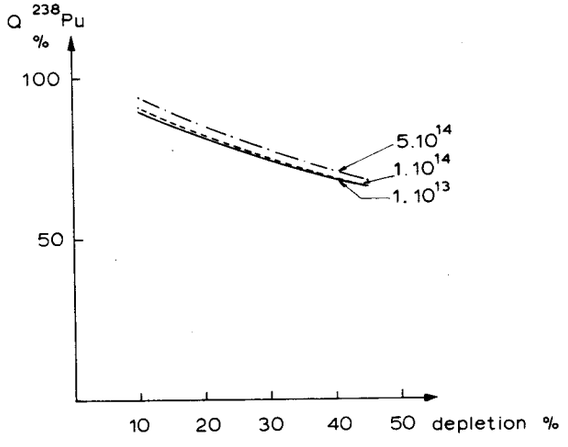
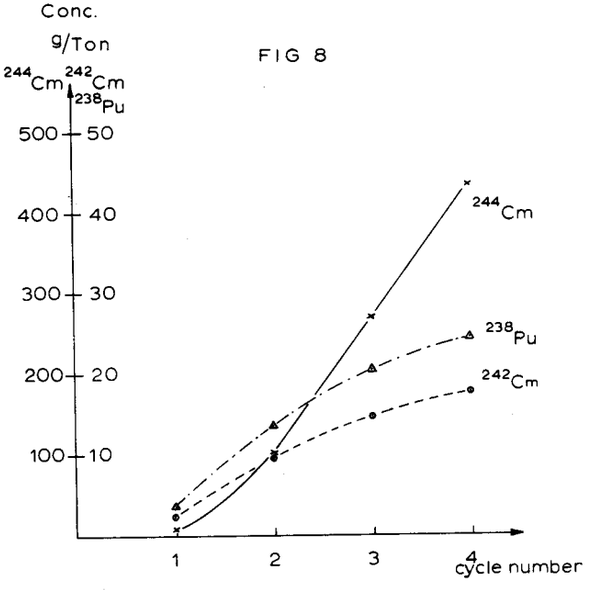


FIG 8



a very important role indeed. It is evident that the content of ^{236}U (formed in about 15% of the neutron interactions with ^{235}U) in socalled depleted fuel and the content of isotopes heavier than 239 in plutonium are indeed a most valuable asset. The invested neutrons have a very real value, as will be discussed in chapter VII.

Tables IV and V, taken from Deonighi and Eschbach[1] illustrate this point very well. Figure 8 graphically presents the data from table IV for the product isotopes, Figure 9 those for the precursor isotopes. In addition, it will be clear without further discussion that e.g. the quantity of ^{244}Cm produced at a given fuel exposure of plutonium, will be higher with higher contents of heavier isotopes, and especially with higher ^{242}Pu content. Figure 10, also taken from reference [1], shows the ^{244}Cm production for various plutonium compositions in natural uranium in a PWR type reactor. Plutonium composition is indicated by the percentages of isotopes 239, 240, 241 and 242, resp.

Table IV

Recycle of plutonium and americium in a PWR using slightly enriched uranium (SEU) as supplementary enrichment. 24.000 MWD/ton per cycle.

Cycle No.	Initial fuel	Enrichment weight %	Isotopic concentration (g/T) Pu 239	Pu 240	Pu 241	Pu 242	Am 241	Am 243[a]	After discharge Cm 242	Cm 244	Pu[b] 238	% 238Pu
1.	SEU	2.92	5518	1882	995	255	88	45	2.6	8.4	3.7	95
2.	SEU+recycle Pu+Am cycle 1	3.27	6579	2831	1773	778	256	258	9.6	102.6	13.5	85
3.	SEU+recycle Pu+Am cycle 2	3.57	7136	3223	2249	1217	301	537	14.5	270.1	20.3	77
4.	SEU+recycle Pu+Am cycle 3	3.82	7514	3413	2498	1533	359	797	17.5	433.9	24.4	71

a) includes ^{241}Am formed after separation

b) ^{238}Pu only from ^{242}Cm decay after separation (120 days cooling).

FIG. 9

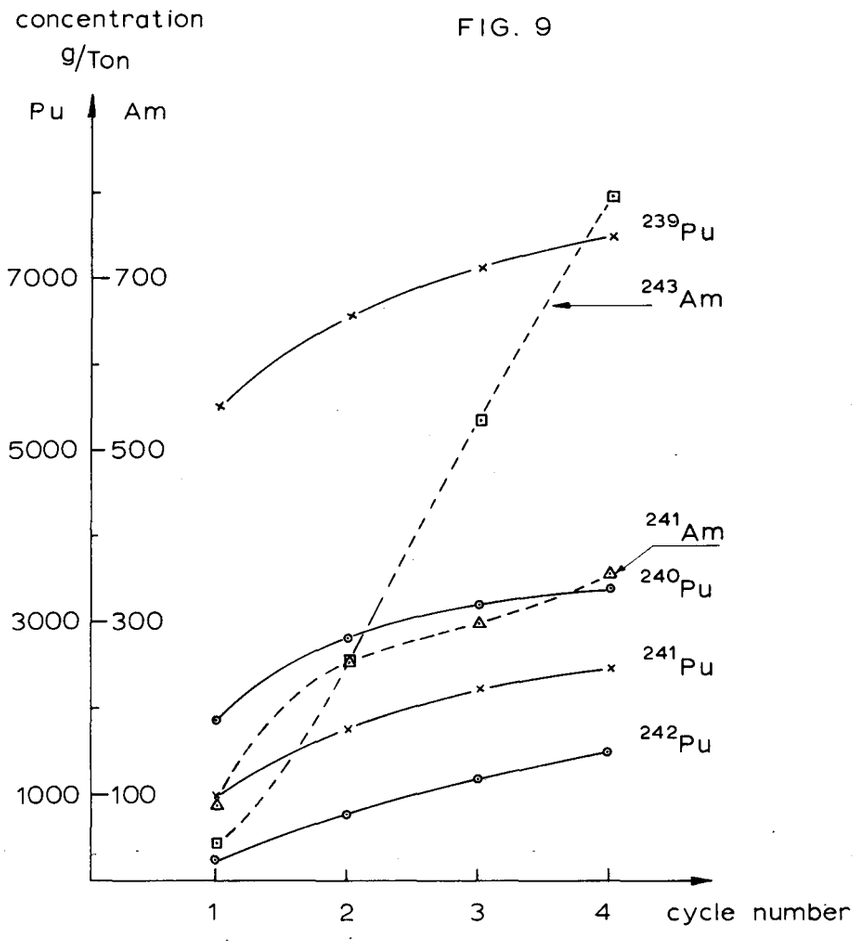

concentration
g/Ton
Pu
Am
7000
700
5000
500
3000
300
1000
100
239Pu
243Am
241Am
240Pu
241Pu
242Pu
1
2
3
4
cycle number

FIG. 10

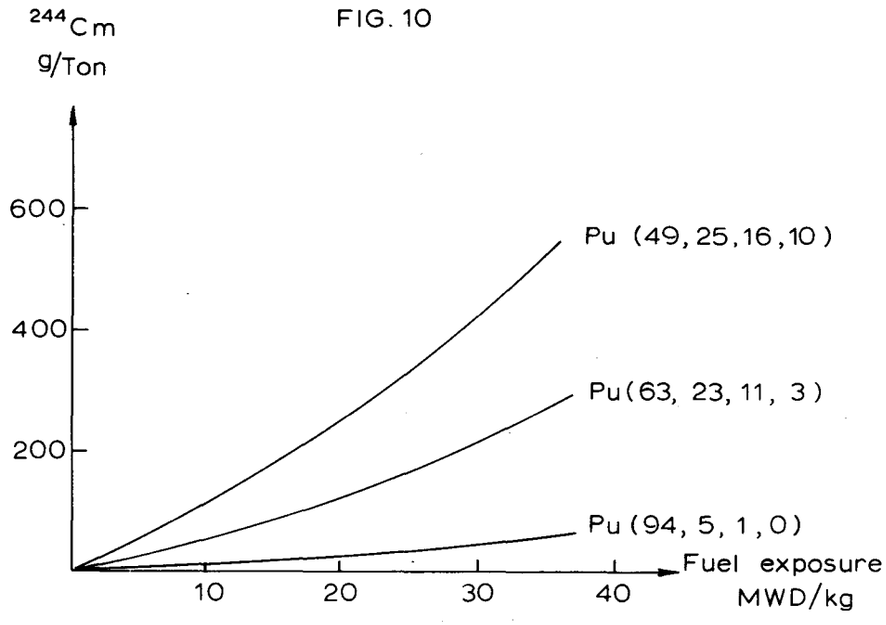

244Cm
g/Ton
600
400
200
Pu (49, 25, 16, 10)
Pu(63, 23, 11, 3)
Pu (94, 5, 1, 0)
10
20
30
40
Fuel exposure
MWD/kg

Table V

Recycle of ^{236}U and ^{237}Np in PWR
24.000 MWD/ton.

Slightly Enriched Uranium

Cycle	Fuel enrichment weight %	^{236}U Isotope Concentration, g/T. Initial	^{236}U Discharged	^{237}Np Discharged	^{237}Np Target in	^{237}Np Target out	^{238}Pu Via ^{237}Np [a]	^{238}Pu via ^{242}Cm [b]
1.	2.92	0	3288	407	0	0	0	6.2
2.	3.06	3187	6079	665	394	315	79	5.5
3.	3.16	5896	8488	806	950	760	190	4.8
Fully Enriched Uranium								
1.	3.16	0	3863	1150	0	0	559 [c]	0 [a]
2.	3.21	3747	6701	1643	1115	892	777	223
3.	3.26	6500	8970	1953	2485	1988	902	497

a) Q = 85%
b) Q = 90%
c) Q = 65%

Simultaneously, the $Q^{244}Cm$ will, of course, increase with ^{242}Pu-content for equal fuel exposure. Finally, attention may be drawn to the fact that also variations in cooling times and time required for reprocessing and (re-)-fabrication may influence the overall production figures. These, relatively minor, factors will not be further discussed here.

Chapter VI

PRODUCTION CAPACITIES

It is, of course, very important to have a reasonable estimate of the quantities of ^{238}Pu, ^{242}Cm and ^{244}Cm which could become availabl in the ten to fifteen years to come. This will be necessary for getting an idea of to which extent the demands may be met and whether any priorities will have to be assigned to specific applications. Secondly, it will also help to evaluate the (future) interest of reactor operators, fuel processors and prospective manufacturers, since prices and profits will also depend on the extent of imbalance between demand and supply. Also decisions on whether, when and where specific processing and/or target fabrication facilities will have to be constructed depend on assumptions regarding future production capacities.

It seems reasonable to assume that sufficient reactor capacity will be available in the $1 - 3 \times 10^{14} nsec^{-1} cm^{-2}$ flux range for irradiating the base materials neptunium and americium produced by the power reactors. It is also clear that the special irradiation programs like those at Oak Ridge and Savannah River which are carried out within the framework of transplutonium elements research programs or for shorter special production campaigns will produce quantities which are small or even very small compared to those originating from the nuclear power industry.

As discussed earlier and shown below the production of the nuclides under discussion is dependent on the reactor type chosen. Reactors of the PWR-type produce for the same fuel burn-up more of them than BWR-type reactors. Also fast reactors are capable of producing large quantities of these nuclides. ^{235}U-fueled reactors will produce in particular ^{238}Pu and its precursors, while plutonium fueled reactors will lead to considerable quantities of curium isotopes. By analogy, fuel enrichment by plutonium instead of by ^{235}U would shift the production more to the curium side. Although fast breeder reactors for power production still seem to be a decade or so away, plutonium fuel enrichment is near. These developments will undoubtedly influence the total production figures and should be taken into consideration in any detailed analysis.

More important factors in the near future will be, as shown in the previous chapter, the possible optimizing of reactor design respective to production of α-emitting nuclides and the extent to which recycling will be applied. It has been reported by Wilson and Showalter[16] that recycling for enhancing this production is already being applied in the U.S.A. It may safely be assumed that by making full use of these possibilities in a systematic way the overall production may be stepped up by a factor of between 2 and 4 over that estimated on the basis of a once-through fueling system of the projected nuclear power production.

Nevertheless, the only figures available for arriving at a, necessarily crude, estimate of the total production capacity are those on the contents of base material, precursor nuclides and products in power reactor fuel for a few specific reactor types. Only in one case experimental analytical data have been reported. These confirm very well the calculated figures, which, therefore, may be considered with confidence.

Also an estimate of the total nuclear power capacity to be installed in the next one or one and a half decade is very difficult. The sudden 1966 expansion in nuclear plant orders in the United States proves this, while also latest developments in the Euratom countries show large deviations from very recent forecasts. Therefore, it is clear that the best estimate, which can be made today on the basis of figures concerning once-through cycles and todays installed nuclear power capacity forecasts, must be a very low one indeed. It might be surpassed by a factor varying even from two to five.

The figures of table VI are taken from one of the most detailed reports available[1]. They concern the yields of some of the product and target materials in different power reactors, calculated for a fuel exposure of 24.000 MWd/ton. The load factor has been taken as 80%, the thermal to electrical conversion efficiency as 33%, the time between fuel discharge and product availability as one year and processing losses as 5%.

Table VI

Nuclide production in typical power reactors
(grams/ton)

Fuel	^{237}Np	^{238}Pu a)	^{243}Am	^{244}Cm
1. slightly enriched uranium	375	4.0	50	9.6
2. as 1)+Pu recycle from 1)	318	12.1	250	85
3. Pu b) in natural uranium	122	27	540	165
4. as 1) Pu,Am, ^{236}U recycle from 1)	600	14	265	108
5. Pu b) in zirconium	8.8	44	1440	1130
6. fully enriched uranium in zirconium	1070	530 c)	37	51
7. as 6)+recycle ^{236}U from 6)	1500	720 d)	38	44

a) ≈ 80%, from ^{242}Cm-decay
b) Pu composition: 238: 1.1%; 239: 62%; 240: 22.8%; 241: 11%; 242: 3.1%.
c) Q = 65%
d) Q = 67%

Table VII presents the data, given by Roberts and Van Tuyl[17] on typical PWR- and BWR-fuel as determined by chemical analysis.

Table VII

Analyses of Yankee and Vallecitos BWR fuel.
(grams/ton)
(one year after end of irradiation)

Reactor	Burnup LWd/ton	^{236}U	^{237}Np	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{242}Cm	^{244}Cm
Yankee	31.800	3690	473	113	6350	2160	1300	390	139	1.6	16
"	24.300	3650	455	90	7490	1700	1110	200	119	1.0	7.9
" a)	17.700	2230	205	32	5620	1240	599	96	85	0.03	0.89
Vallecitos	17.400	2890	140	17.5	3330	880	329	51	45	0.02	0.55

a) 3.49% ^{235}U initially

These and other data available from other sources all lead to similar results, although variations occur.

Although in recent years and months many forecasts have been published on the installed nuclear capacity in, e.g. 1980 for Western Europe and the United States, all estimates have proved to be wrong.

Table VIII gives forecasts which were generally accepted and widely publicized. Also listed are the most recent figures on installed or committed capacity, and the ratio's between forecast and capacities that will exist in the early 1970's. Assuming that special factors have contributed to the order-boom in 1966, these factors have been arbitrarily reduced to 1.5 for the Euratom countries and 2.5 for the U.S.A. in order to arrive at a very rough corrected estimate for the installed capacity in 1980 based on the earlier official estimates (1965/66).

Table VIII

Predicted and estimated nuclear power
Capacity in LWe

Date	EURATOM			UNITED STATES		
	1965/6	1970	1980	1966	1970	1980
Forecast 1964[18]	1000	3.800				
" 1965[19]		3.700	40.000		7.000	75.000
" 1966[20]				12.300	20.000	120.000
Known 1966[21]	1750	8.000[a]			>30.000[b]	
ratio forecast '65/ known '66		2.1			4.3	
corrected estimate			60.000			185.000

a) Total in operation, under construction and under disign by December 1966.

b) Total in operation, under construction and ordered by December 1966.

In these estimates all other countries, some with large nuclear power industries in existence or under development like Great Britain and Russia have been left out of consideration.

Finally, Table IX gives an estimated annual overall production of the three nuclides under consideration, based on the data of Tables VI to VIII, assuming a 3 to 2 ratio of PWR to BWR types of reactors, an overall conversion of 65% for ^{237}Np to ^{238}Pu and of 33% to ^{238}Pu, 15% to ^{242}Cm and 40% to ^{244}Cm for the mixed americium starting material and neglecting effects of plutonium enrichment, existence of fast breeder reactors and of e.g. recycle operating. These figures certainly may not be considered as being more than an estimate of the order of magnitude of annual production of ^{238}Pu, ^{242}Cm and ^{244}Cm at the dates given.

Table IX

Estimated annual production
in kilograms

Nuclide	Euratom		United States	
	1970	1980	1970	1980
^{238}Pu	18	400	140	1250
^{242}Cm	1	25	12	90
^{244}Cm	4	80	30	250

In calculating these figures it has been assumed for the United States and the Euratom 1980 figures that 3 years will elapse before products become available after reactor loading and 4 years for the Euratom 1970 figures.

It may, therefore, be concluded that if the nuclear power capacities develop according to the corrected extrapolated optimistic trend of last year's figures U.S. space requirements as estimated today may be met by 1980. In addition, many different other applications will be possible. In any case, both in the United States and the Euratom countries very important amounts of these valuable nuclides will be available for a great variety of applications.

Chapter VII

ECONOMIC ASPECTS

From the preceeding discussion it will be evident that in any case industrial non-fuel applications of α-emitting nuclides will develop in the near future, due to the properties of these materials, the technically attractive possibilities and their inherent value. It has recently also become clear that the isolation and production of these nuclides will offer substantial financial profits to the nuclear power industry. In this chapter an attempt will be made to outline the economic importance of the subject.

At present no real price exists for the transuranium elements. A price has been set only for plutonium-239 and plutonium-241 in respect to their value as a nuclear fuel. Both are considered as having a price of $ 10 per gram. The only other prices are $ 1500 per gram for ^{241}Am and $ 500 per gram for ^{237}Np. It is, however, only possible to acquire under special conditions one to ten gram quantities of these nuclides. These prices, therefore, have to be considered more as a processing and handling charge than as a real price.

Several attempts have been made to determine a more realistic price of a number of the nuclides considered and even to obtain an indication of future prices which could be expected in a well developed nuclear power industry. Such prices, the number of dollars per gram to be paid by a customer, include the costs of the original starting material, the costs of production (target manufacturing, irradiation, processing, transportation) and investment and profits for the producer of the nuclide from its precursor or base material.

Rohrmann[22] has derived future prices for the nuclides of interest, based on certain assumptions, the most important of which are:

1) There will be an unlimited market for ^{238}Pu and ^{244}Cm at the projected prices of $ 500 (Q = 80%) and $ 1000 per gram, respectively.
2) A competitive situation exists for reactor fuel processing in combination with isotope recovery systems.
3) Formation and concentration of the products will be established in typical, low enrichment, power reactor fuel (25.000 MWd/ton).

4) The cost of fuel-processing is $ 27.000/ton of uranium. Half of this cost ($ 2/gram) will be charged to plutonium and the other half to the recovered uranium.

5) ^{237}Np and ^{242}Cm are only valued as precursors of ^{238}Pu; ^{243}Am only as precursor of ^{244}Cm.

Regarding these assumptions, the following comments may be made. The prices quoted under 1) seem to be most reasonable extrapolations under present circumstances. In case the expected shortage may be overcome, the quantities will be so much larger, that it seems likely to assume, that the overall profits will be essentially the same. Assumption 2) is now being fulfilled for the United States and one might expect the same to be valid for Europe within a couple of years, or at least at the time when these nuclides will be handled in the quantities expected. Assumptions 3) and 4) are most reasonable and conservative, while assumption 5) underestimates the value of ^{242}Cm which itself may be used as isotopic fuel before yielding the product ^{238}Pu.

Kohrmann also calculates the value of ^{236}U and the additional value of the other plutonium isotopes which is due to the value of the invested neutrons in view of their role as precursor material for the production of isotopic fuel. This calculation is based on a neutron price of $ 2500 per gram, derived from the price of the additional ^{235}U enrichment required for producing 1 gram of extra neutrons available in a power reactor. In other words, the values quoted in Table X for the different plutonium isotopes indicate the total value to be realistically ascribed to specific isotopes. Some of these were up to now considered as having no or negative value.

Table X also lists the assumed future production costs and the value of the nuclides in the fuel, to be credited to the reactor operator. Future production costs are primarily determined by the processing costs which are in some cases shared equally by different isotopes.

Table X

Estimated values of nuclides in fuel
in dollars/gram

Nuclide	Current Price	Future Price	Assumed Production Cost	Estimated Value in Fuel
^{236}U	-	11	0	11
^{237}Np	500	100	80	20
^{241}Am	1500	100	50	50
^{243}Am	-	500	50	450
^{242}Cm	(20.000)[a]	300	150	150
^{244}Cm	-	1000	100	900
^{238}Pu	(880)	500	350	150
^{239}Pu	10	11[b]	2	9
^{240}Pu	-	7	2	5
^{241}Pu	10	19[b]	2	17
^{242}Pu	-	41	2	39

a) based on estimated present value as isotopic source material
b) inclusive of value as fuel ($10/gram)

A first indication as to the validity of these prices may be obtained from two sources. In a 1964 report Williams[23] points out that the present ^{241}Am price of $ 1500/gram may already be lowered very considerably by processing larger amounts at a new facility which is under construction. With a design capacity of 3600 grams per year (which could be increased to 5000 grams per year) processing costs would come down to $ 140/gram. Further improvements are certainly possible, in particular since the americium may be processed together with other valuable by-products. Secondly, the U.K.A.E.A. recently announced[24] that neptunium recovery at Windscale will be stepped up in view of the rising demand. NpO_2 prices would range from $ 252/gram for orders of 5 kgs to $ 75.60 per gram for orders of 20 kilograms.

It therefore seems fully justified to use the prices of Table X as a basis for evaluating possible profits for the nuclear power industry, especially as higher prices for certain isotopes will tend to increase these profits.

In order to approximately evaluate the profits to be gained by the nuclear power industry, several attempts have been made to interpret the existing data.

Rohrmann simply calculates, on the basis of his in-fuel value and calculated contents of 25.000 MWd/ton slightly enriched uranium fuel, total values per ton and per electrical kWh. As his estimated contents are for some nuclides higher, for some others lower than those of Tables VI and VII, his conclusions are, in Table XI, compared with those based on the 24.300 MWd/ton Yankee fuel and those of case 1) of Table VI making approximations for nuclides not listed in Table VI.

Table XI

Nuclide values in spent fuel
$/ton

Nuclide	Rohrmann[22]	Table VI Case 1.	Table VII
^{236}U	36.070	39.600	40.150
^{237}Np	6.320	7.500	9.100
^{241}Am	2.800	10.800*	5.950
^{243}Am	39.120	22.500	36.000*
^{242}Cm	3.450	-	150
^{244}Cm	19.800	8.640	7.110
^{238}Pu	- a)	1.200c)	- a)
^{239}Pu	49.100b)	10.000*	8.240b)
^{240}Pu	7.585	10.500*	8.500
^{241}Pu	24.000b)	23.000*	18.870b)
^{242}Pu	15.050	9.400*	7.800

* estimated
a) ^{238}Pu in fuel considered as of no value
b) inclusive of $10/gram as nuclear fuel
c) Q > 80% from total ^{242}Cm decay

Although the estimated figures (indicated with *) are very rough, the total credit per ton of spent fuel due to the non-fuel value of the nuclides listed amounts for the three cases to the very similar figures of approximately $ 145.000, $ 127.000 and $ 128.000 respectively for 25.000 MWd/ton fuel. This is equivalent to an average saving of 0.68 mill. per kWhe.

Another approach has been taken by Deonighi and Eschbach[1], who try to arrive at reasonable price settings for the base materials which will be valid in a free market. In that case the reactor operator will be willing to pay a target material price which will

at least assure him of breaking even in his fuel cycle price whether he performs these production irradiations or not.

In their study the influence on the target prices and final profits is considered of a large number of factors as e.g. reprocessing costs, product prices, target composition and recycle. Some of their results are presented in Table XII in which the fuel cycle cost reduction for four different reactors for normal fueling methods are given as a function of different values, credited to specified nuclides. For comparison the credit values given by Rohrmann are also listed.

As may be seen, the values quoted by Rohrmann which may be considered reasonable, would give results in between cases 9 and 10, probable closer to case 10 than to case 9. For the four reactor types listed, fuel cycle reduction costs would than be typically reduced by from 15 to 75 percent. Again, it is found that PWR-reactors allow greater profits to be made than BWR-reactors.

As another example Deonighi and Eschbach point out that in case 2 - only neptunium valued at $ 100/gram - an annual saving of about $ 750.000 would be obtained for a 1000MWe power reactor.

Finally, the report by Nilson and Showalter[16] may be quoted, to illustrate the financial gains which could be realized by a fuel cycle system of a PWR-reactor in which only the ^{236}U is consequently recycled. As pointed out above, uranium fuel recycle is a very simple way of enhancing ^{237}Np production. Figure 11 graphically presents the accumulative investment and return over an entire reactor lifetime for a 1000 MWe PWR-reactor. The basis for the calculation has been an estimated value of $ 300 per gram of ^{238}Pu, which is much lower than the value assumed by Rohrmann. Even so profits are very considerable even compared with the cost of the reactor itself, although the reactor operator will have to be willing to make additional investments in the first years of reactor operation.

Table XII

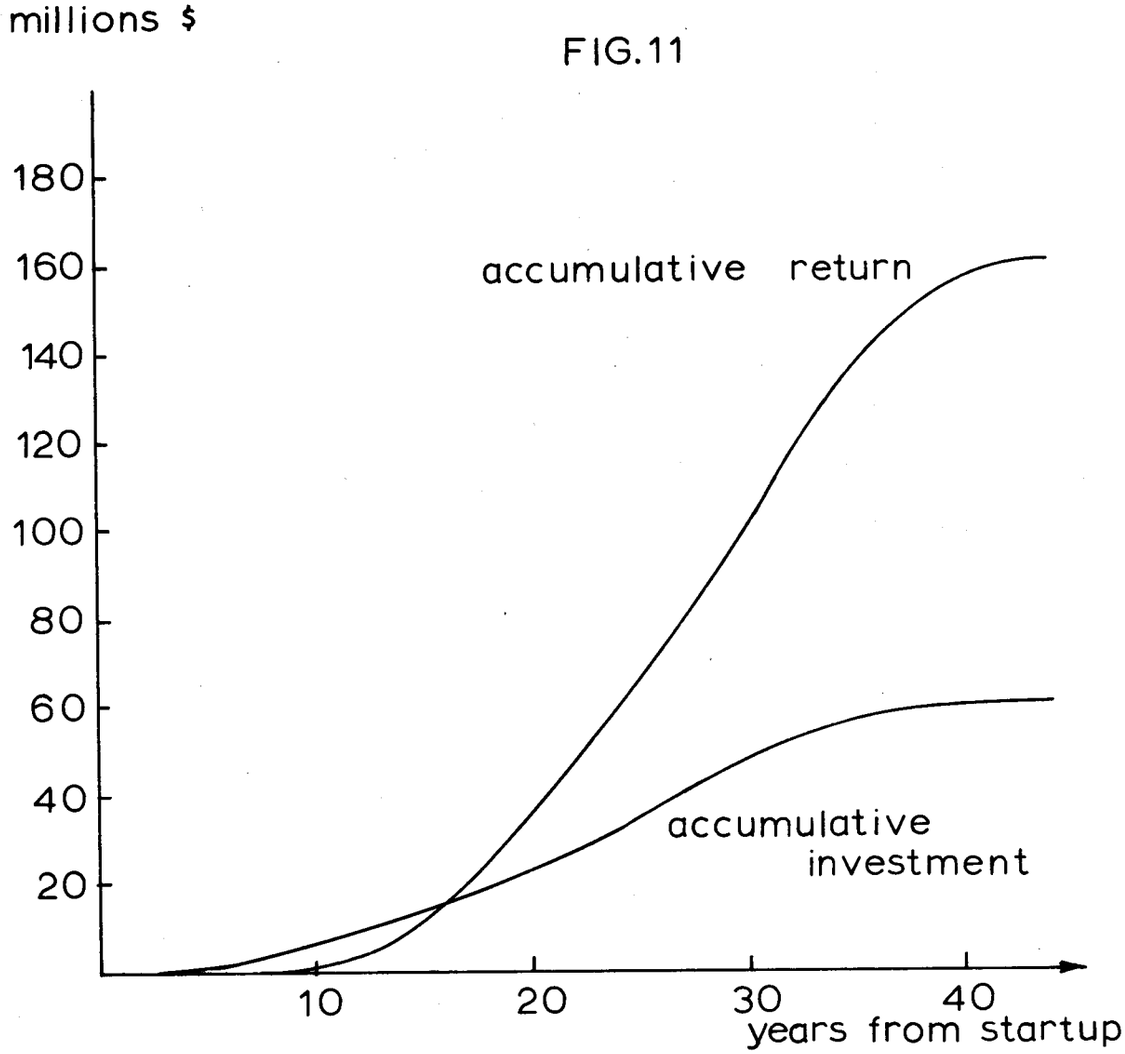
Case No.	Credit ($/gr) Nuclide ^{236}U	^{237}Np	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{243}Am	^{242}Cm	^{244}Cm	Fuel Cycle Cost Differences[a] (mills/kWhe) BWR[b]	PWR[b]	PWR (Pu)[c]	PWR (Pu nat.)[d]
no credit											2.165[e]	1.885[e]	1.885[e]	1.885[e]
1	-	-	10	-	10	-	-	-	-	-	-0.000	-0.000	-0.000	-0.000
2	-	100	10	-	10	-	-	-	-	-	-0.098	-0.141	-0.161	-0.095
3	-	150	10	-	10	-	-	-	-	-	-0.147	-0.212	-0.187	-0.144
4	-	-	10	-	10	-	50	-	200	-	-0.016	-0.019	-0.038	-0.124
5	-	-	10	-	10	-	-	300	-	1000	-0.110	-0.217	-0.656	-1.497
6	-	-	10	-	10	-	-	100	-	500	-0.043	-0.106	-0.285	-0.687
7	-	150	10	-	10	-	150	300	400	1000	-0.295	-0.486	-0.980	-1.886
8	-	50	10	-	10	-	50	100	200	500	-0.108	-0.174	-0.400	-0.845
9	-	150	10	-	20	100	150	300	400	1000	-0.428	-0.658	-1.197	-1.777
10	-	50	10	-	15	50	50	100	200	500	-0.172	-0.251	-0.505	-0.793
Rohrmann Values	11	100	11	7	19	41	100	500	300	1000				

a) fuel exposure has been optimized to specific conditions
b) fueled with slightly enriched uranium
c) recycling plutonium from previous U-cycle (Pu 63,23,11,3)
d) fueled with plutonium enriched uranium
e) mills/kWhe, fuel cycle cost case 1

A point of major importance to be stressed which results from all detailed studies is the fact that production costs of the three nuclides of interest are almost entirely processing costs (target fabrication, chemical processing, transportation, etc.) and not irradiation costs. One report[25] even indicates that for the nuclides ^{238}Pu and ^{242}Cm only <1% of the production costs is due to the irradiation and in the case of ^{244}Cm 27%.

From the foregoing it will be clear that both reactor operators and fuel reprocessors will profit from the production of these nuclides and that major financial incentives do exist towards developing satisfactory techniques.

FIG.11



Chapter VIII

DEVELOPMENT REQUIREMENTS

In the preceeding chapters a survey has been given of presently existing typical data on the production and application of the three α-active nuclides ^{238}Pu, ^{242}Cm and ^{244}Cm which will become industrially available with the coming of age of nuclear power. It has also been attempted to make a first estimate of the importance, both technically and financially of these developments in the near future. Although it is very evident from the data that large profits can be gained, it still has to be determined how large these profits may be in given cases and which are the best ways and means to obtain them.

In the first place it must be pointed out that both from a theoretical and an experimental point of view present data are far from complete. Although many aspects have been studied separately - and often for similar reactor types - no complete and detailed evaluation has yet been published for an assumed actual condition of the nuclear power market in for instance 1970 or 1980, taking into account the extent to which different reactors will be available. For example, no data were found on the influence on costs and profits of irradiating target materials in production or testreactors instead of in powerreactors. Also, no complete study has been made as to the relative attractiveness (technically and financially) for reactor operators and fuel processors of the (simultaneous) optimalization of a variety of factors. The balancing of profits against technical complexity may be difficult. Therefore, it must be most rewarding to evaluate optimum conditions for a given set of conditions expected for the nuclear power market, e.g. in the Euratom countries.

Secondly it must be stressed that in fact only very limited experimental data exist. Only in one case analyses have been performed on the contents of four nuclides in spent fuel. Almost no reliable experimental data exist on a number of nuclear data of major interest. Cross-sections for e.g. capture, fission and (n,2n)-reactions as a function of non-thermal neutron energy are almost completely lacking, as are, for example also data on self-shielding. The effects of neutron flux and energy on changing composition during burn-up, of intermittant irradiation and processing cycles, etc. are not known from experiment.

Above all, therefore experimental irradiations should be carried out under typical but well-known conditions - similar to those expected later in the larger programs - in order to privide chemical analytical data. These should be used, first of all to check the calculations, secondly to find out the best combination of irradiation and processing cycles. These experiments do not seem to require large experimental facilities which, moreover, do exist. Financial requirements will be extremely modest.

The next point to be mentioned is the fact that today no satisfactory technology exists for target fabrication. For example, questions as to the most suitable form and optimum concentration in view of nuclear behaviour and cooling are largely unanswered.

Furthermore, no detailed knowledge and experience exists on the best "integrated" fuel and target processing schemes. Since, as mentioned in the foregoing chapter, the processing costs are substantial and price determining, huge rewards may be expected from even small improvements. More in particular, present knowledge of isolation and separation of americium and curium is far from being satisfactory, from a fundamental point of view and regarding to technological development. Particularly interesting seems to be the question whether some reprocessing systems (e.g. aqueous, non-aqueous liquid, and volatility processes) are inherently better adaptable than others. The outcome of studies like these will also indicate whether the desire to reprocess and to refabricate targets at the site of the conversion reactors can be realized.

One of the points of major interest, still to be studied extensively, seems to be a critical evaluation of the quality requirements to be met for specific applications of α-emitting nuclides. These results will be of great significance for any study concerning the optimalization of reactor design, reactor operation and fuel treatment. As early as possible criteria should be formulated regarding permissible power output variations, irradiation protection and health physics-aspects, and, in general, isotopic fuel source construction and behaviour.

It is evident that the development itself of isotopic power applications plays an essential role. This, however, seems to be getting well underway. Contributions from all these and still other areas are required in order to arrive at realistic value assessments of the nuclides under consideration.

Chapter IX

SPECIAL ISOTOPES

Very briefly some uses will be mentioned of α-emitting nuclides not considered in the previous chapters.

First of all the californium isotope of mass 252 may be mentioned. This shows spontaneous fission activity ($T_{\frac{1}{2}}$ = 66 years) and α-particle emission ($T_{\frac{1}{2}}$ = 2.55 years), the first decay mode being most interesting. Apart from its uses as standard fission neutron source, it is ideally suited for all applications where point sources of high neutron intensities are required. An excellent review of the possibilities was very recently given by Stetson[26] who also discussed other sponteneously fissioning nuclides as ^{244}Cm, ^{254}Cf and ^{256}Fm. Stetson draws in particular attention to those aspects which make these neutron sources preferable above others, like neutron generators, (α,n) sources and nuclear reactors. A wide field of interesting experiments is being opened up.

Also certain space applications (light weight sources for activation analysis in space) and medical uses call for higher production rates. For example the use of ^{252}Cf for cancer treatment looks promising since the irradiation can be extremely well focused, thus reducing high irradiation doses on environmental tissue[27]. In vivo activation analysis may be amongst other near future applications. Without doubt a commercial demand for these isotopes will arise as soon as there will be a supply possibility.

^{245}Cm has been proposed as being very suitable for making compact reactors due to its low critical mass.
A few words may be said about the possible uses of ^{210}Po and ^{227}Ac as isotopic power source material. ^{210}Po, having roughly the same application characteristics as ^{242}Cm, is difficult to produce and will be several orders of magnitude more expensive than ^{242}Cm. ^{227}Ac, obtained by irradiating radium, has favorable characteristics. The quantities obtainable will, however, be extremely small compared to those of the nuclides discussed in this report, while also the irradiation of substantial amounts of radium in a reactor does not seem to be very attractive.

Finally, it may be pointed out that there is widespread interest in producing the longest lived isotopes of the transplutonium elements in general, since these will permit to carry out detailed chemical and physical studies which are impossible with the shorter lived isotopes. ^{244}Pu, ^{247}Bk, ^{251}Cf, ^{254}Es and ^{257}Fm may be mentioned.

Chapter X

CONCLUSIONS

Within a timespan of only a few years the outlook as regards the possiblity and interest of producing α-emitting nuclides for non-fuel applications has drastically changed. Still only two years ago it was emphasized that under all circumstances power production would and should be the only legitimate purpose of nuclear power reactors and that the production and isolation of simultaneously produced isotopes would be feasible only if it would not interfere, technically or financially, with what was called normal reactor operation. Interesting applications of these nuclides had been found and were under development, but the essential questions were how to obtain them and at what (meaning often any) price.

Nowadays technical developments, theoretical studies and some experimental evidence have demonstrated that a large demand will exist for these nuclides and that profits to the nuclear power industry will be so substantial that drastic changes in reactor operation and reactor design seem to be justified. These changes could largely contribute to arriving more readily at a well balanced, integrated nuclear power market. These developments are also the logical consequences of the emergence of plutonium as a nuclear fuel.

Will it be possible to effectuate the necessary modifications and innovations at the right time, then much experimental work and theoretical studies will have to be carried out. The experimental work will include not only laboratory studies, but also a substantial effort in process development. Since the need for isotopic power begins to be urgent and, on the other hand, nuclear power is playing a fastly increasing role in our economy work will have to be started now, where it has not yet got underway taking into account the complex nature of the problems.

It seems to be reasonable to state that for Euratom in about five years the pilot plant stage must start. On the basis of our present experience and in view of the existence of some specialized groups within Euratom, it seems that this could be realized by fully exploiting the possibilities offered by todays reactors. In particular the first experimental work, which will require only a very modest amount of money, could be immediately started.

As indicated above, these experiments should concern the irradiation of available target material in well defined neutron spectra in fluxes of the order of $10^{14} nsec^{-1} cm^{-2}$. They will not only provide valuable data on product yields under different irradiation conditions and irradiation cycles but also contribute to the development of target design and fabrication and, in particular, of reprocessing schemes. On the basis of these results supplemented by theoretical evaluation and reconnaissance a more detailed program of wider scope could soon be drawn up.

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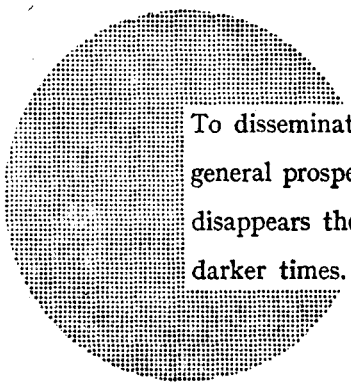

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Alfred Nobel

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