FUEL REPROCESSING, A UNIQUE OPPORTUNITY TO MAXIMIZE THE AVAILABILITY OF TRANSURANICS

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ABSTRACT AND SUMMARY

The processing of spent commercial nuclear fission reactor fuels to simplify fission product disposal makes available many useful by-product isotopes in addition to fissile plutonium. Moreover, processing makes available intermediary isotopes that can be utilized to produce additional amounts and types of transuranics and removes most of them from waste streams requiring disposal. Such intermediary isotopes are available in the uranium and plutonium as well as in other streams. The extent to which the purposeful production of selected transuranics is enhanced will be driven by perceived economic conditions, both current and future. Technologically, production enhancement can be substantial (a factor of 2 or 3 for production chains involving longer lived isotopes and 10 to 100 for production chains involving shorter lived isotopes). Theoretically some 180 kilowatts per year per 1200 MW a reactor is possible (9 to 18000 kw/yr for 50 to 100 reactors). For incidental production, possible revenues to defray processing and waste disposal costs are estimated to be 3 to 4 mills/kwhr with recycle of intermediaries. In addition, waste disposal of TRU's should be simplified as the longer-lived TRU isotopes, such as Np^{237} will have an economic value when removed and transmuted to 89 year half-life Pu^{238}. System studies completed in the early Sixties are reviewed so as to provide a "start" for enhancing both incidental and purposeful production of the transuranics by adjusting the fuel cycle, especially fuel exposure, and by avoiding isotopic dilution of recycled uranium and plutonium streams. In addition, production could be increased by appropriately "managing" irradiation of intermediaries that include two or more isotopes of the same element. Attention is also directed towards "indifference" pricing philosophies to result in a stable enterprise even though a sequence of processing and irradiation involving different fiscal enterprises were involved. The "indifference" pricing system avoids both losses and windfalls to the participants. Examination also indicates that a significant reduction could result in the costs of electrical power if a substantial market develops in decay isotopes.

The diversion of neutrons from production of fissile fuel to special isotopes appears relatively minor since the major neutron investments are now being made in these isotopes as neutron parasites. Moreover, in view of the long-term development of the breeder and the possible near saturation of the demands for energy, an excess of neutrons will be available beyond the needs to breed fissile fuel. Thus, decay isotopes can be bred and "conversion ratios" for such isotopes are developed. In addition, the technological approaches and pricing methodologies reviewed here can be applied for the extra long-term view involving the production of additional transuranic isotopes from thorium irradiation, both thorium-232 and thorium-230 (Ionium).

INTRODUCTION

This review paper will not be addressed to the most important transuranics that will be made available through processing. I speak, of course, of reactor grade plutonium and uranium-233; with their recycle in a breeder system, the uranium and thorium in average earth makes a ton of earth equivalent in heat content to 80 tons of coal. The value of plutonium and U^{233} as nuclear fuels has been characterized through analysis as well as demonstration. In thermal neutron reactors the value of fissile plutonium is proportional to the price of fully enriched uranium, which is substantially more than the equivalent burnout cost of U^{235} in slightly enriched uranium. This apparent anomaly is due to the chemical separability of plutonium from uranium and from thorium. Similarly, when mixed with thorium, U^{233}'s value in thermal reactors is proportional to the price of fully enriched uranium but not so if U^{233} is mixed with U^{238}. The true value of both the fissile plutonium chain and fissile uranium chain is similarly detracted by growth of respective non fissile, non fertile isotopes Pu^{242} and U^{236}, each of which with neutron absorption forms higher transuranic isotopes. Potential positive values of these higher isotopes constitutes the major thrust of this review paper. Their potential positive value will alter the heretofore developed value functions for the bred fuels (Pu^{239}-Pu^{241} and U^{233}) from the generic form A + B - C to A + B + C where, in the instance of plutonium, A and B are the concentration weighted values of Pu^{239} and Pu^{241} and C is the concentration weighted value (+ or -) of Pu^{242}. In the instance of uranium fuels, A represents U^{233} (if present), B is U^{235}, and C (+ or -) the value of U^{236}. Most notably, U^{236} is the precursor of Pu^{238}, and Pu^{242} is the presursor of Cm^{244}.

POTENTIAL ROLE OF ADVANCED ISOTOPE SEPARATION

However, before proceeding, I wish to note two major factors not present twenty years ago when most of the studies I am reviewing were conducted, namely, advanced isotope separations, and the possibility of fuel processing to simplify waste disposal. Of greatest technical impact are the possibilities of selective enrichment schemes such as laser methods which hold the promise of being able to selectively "snatch out" specific isotopes. This immediately brings to mind selective removal of U^{236} and Pu^{242} from their respective mixed isotope series. If this can be done in practice, it allows the otherwise parasitic role of U^{236} and Pu^{242} to be removed from recycled mixtures of plutonium and uranium as fuel. Studies have shown this will substantially increase the value of bred plutonium and previously irradiated

uranium as fuels but, in addition, will greatly facilitate the purposeful production of higher transuranics series such as Pu^{238} and Cm^{244}. Such isotopic separations techniques would have further benefits. For example, Am^{241} could be separated from Am^{243} which would grossly simplify production of Cm^{242}-Pu^{238} on the one hand and Cm^{244} on the other. Also very possibly Pu^{236} could be removed as an impurity in Pu^{238} wherever small amounts of Pu^{236} lead to obnoxious gamma ray levels due to thallium-208, a decay product of Pu^{236}. Since the Pu^{236} levels in Pu^{238} must be below 10-100 ppm for some applications, even laser enrichment may be overtaxed. However, laser enrichment could remove Pu^{238} from reactor grade plutonium where it may be as high as 4% in spent S.E.U. fuel at 50,000 MWD/ton, 2% at 40,000, and 1% at 30,000 MWD/ton. By removing U^{236} from bred U^{233} mixtures, some U^{233} systems could indeed breed at just a "tad" over 1, something that has been difficult to show to date. Similarly, removal of Pu^{242} could almost do the same for plutonium in high specific power machines at low fuel exposures. Or even if advanced enrichment systems are limited to producing highly enriched U^{235}, this material could be used to re-enrich spent S.E.U. to allow recycle of U^{236} to enhance Pu^{238} production. Thus, a viable laser type isotope separation scheme could not only greatly increase the neutronic value of recycled bred fuels in thermal neutron reactors, but will greatly simplify production of Pu^{238}, Cm^{244} and other transuranics for special applications. This review will reinforce the promise of laser type isotope separation to simplify production of transuranics because chemical isolation and special fuel configuration schemes described in this review could be obviated by advanced isotope separation. Even so, the methods reviewed, while cumbersome, indicates that there are several practical schemes capable of substantial production which lead to positive values for U^{236} and Pu^{242} without isotope separation.

to complicate waste storage unless substantially removed. Possibly Np^{237} with its two million year plus half-life is a case in point. From a waste disposal point of view, a two million year half-life is "too short to be long and too long to be short." A longer half-life such as that of U^{238} allows disposal with dispersion; plants, animals, and man cannot accumulate enough U^{238} to be harmful. And a shorter half-life such as cesium and strontium assures benignness in a few hundred years. Not so Np^{237}, so let's use it up to make useful isotopes such as Pu^{238}.

Perhaps the "zeal" of our collective radioactive waste programs has jaded our view as to what should be relegated as waste. The term "waste" means of no current value and implies of such little future value as to not be worth saving. To some, the term "fission product" carries the heavier implication of being so dangerous as to require banishment.

In the instance of transuranics, while plutonium concentration involving less than 100 nanocuries per gram may pose no hazard to man, it may yet to be valuable. This is variously summarized in Table I for a range of plutonium concentrations. Note that in terms of fissile concentrations, Pu^{239} at 100 nanocuries/gram is equivalent to natural uranium at 224 ppm, a uranium concentration considered marginally minable today. The heat content of 100 nanocuries plutonium concentration is equally impressive, namely being equivalent to 6 to 10 tons of coal if the Pu^{239} were fissioned.

The figures in Table I also indicate that special precautions are warranted to remove transuranics from high-level wastes. If 99% of the transuranics are removed from HLW at 30% HLW loading of glass, each kilograms of vitrified waste contains 100 to 1000 ppm of fissile plutonium or heat equivalent of 40 to 400

Table I

SOME EQUIVALENT FIGURES BETWEEN NANOCURIES OF PLUTONIUM, GRAMS HEAT, AND NATURAL URANIUM

Nanocuries of ^{239}Pu/gm of Earth	Grams of ^{239}Pu Per Ton of Earth	^{239}Pu Heat Equivalent In Tons of Coal	p.p.m. or gms/ton Of Natural Uranium Having Same Fissile Equivalent
1	.016	$6 - 10 \times 10^{-2}$	2.24
10	.16	0.6 - 1.0	22.4
100	1.6	6 - 10	224.0
1,000	16.0	60 - 100	2,240.0
10,000	160.0	600 - 1000	22,400.0

FUEL PROCESSING FOR FISSION PRODUCT DISPOSAL

Of great economic impact today on utilization of transuranics is the potential appeal of the chemical processing of spent reactor fuels to simplify waste disposal. If utilization of transuranics must carry the startup costs and uncertainty of licensing, then nothing may happen yet again. Certainly with all the troubles and high capital costs of nuclear power, there is little value perceived by reactor operators to support chemical processing and utilization of transuranics--especially recycle of plutonium which will involve licensing alternative fuels. While having chemical processing in place to simplify waste handling may not sound a charge to recycle plutonium, for many years plutonium may well be placed in bonded-like storage. However, processing could make available the incidentally produced Np^{237} and Am^{241} and Am^{243} for production of Pu^{238} and Cm^{244}. In fact, some of the transuranics leading to Pu^{238} and Cm^{244} production tend

tons of coal.

In assessing the future of long-term technological enterprise such as nuclear fission energy, estimated future costs are often heavily emphasized; and on many occasions, I have indulged in such exercises. However, on this occasion I believe it would be more useful to review technical prospects tempered with general concerns of cost rather than assessing the future on the basis of differences in estimated future costs. Estimates of such cost differences are more appropriate for making specific investment decisions.

In addition and perhaps even more important, the examination of isolated costs of energy in view of general inflation and other concerns can be misleading, especially when taken out of context. To

avoid such aberrations one should insist upon examining relative prices throughout all facets of the energy industry and the economy. This is a large task beyond our concern here. However, worth noting is that the prospects for nuclear fission energy should be consistently good because there is no technological reason for the components of nuclear fission power reactors and the associated nuclear fuel processing to experience an ever increasing escalation of costs in terms of relative prices.

Moreover, spent fuel processing has more flexibility than often perceived. The fuel's separations plant really consists of three major steps: 1) the head-end where the irradiated fuel meat is put into solution, 2) the partitioning steps where thorium, uranium, plutonium, and other transuranics and fission products are separated to any desired degree, and 3) the final packaging steps, in particular, wherein fission products are prepared for storage. In support of a given number of reactor megawatts, Step 3 (fission product storage) is essentially the same regardless of the reactor fuel exposure; and Step 2 (partitioning) is nearly the same except for the throughput of uranium and thorium which is greater at lower exposures but the concentration of fissile isotopes may be less. Step 1 (the head-end) is the most variable, the amount of dissolution varying inversely with the fuel exposure. Thus, separations plants could be operated and priced so as to reflect a fixed and variable cost situation.

Moreover, funding and operation of fuel processing plants should possibly be more like the franchised utilities to be served rather than like one of several separately owned chemical plants producing the same product and struggling for market share. The gross load on a spent nuclear fuel plant is determined by the commitment to handle radioactive wastes and by the reactor builds which are known years ahead.

TRANSURANICS

Some transuranics have the appropriate combination of half-life, specific power and radioactivity (predominately active alpha particles) to serve as good heat sources. But securing these isotopes from irradiated fuel and, more particularly, arranging fuel cycles in order to increase their production are undeveloped technologies. For this reason, the current production costs of heat-source transuranics are high, and only relatively exotic applications, as in space programs, can currently justify their use.

In this review no effort has been made to estimate the cost of the special fuel-element encapsulation or separations processes required to produce and recover heat-source transuranics; the reason being that the future scale of the industry and the technology to be selected are unknown. It is believed, however, that the relative reactor burnup costs can be computed with some relevance, and this is the task to which the present review paper is mainly addressed.

More specifically, this review was undertaken 1) to compute the rate at which Pu^{238} and Cm^{244} (as well as their preceding isotopes Np^{237}, Cm^{242}, Am^{241}, and Am^{243}) are incidentally produced by power reactors, 2) to compute the increase in the production rate and purity of Pu^{238} and Cm^{244} when normal power reactor fueling methods have been altered, 3) to estimate equitable price relationships between the product isotopes Pu^{238} and Cm^{244} and all of their preceding man-made isotopes, 4) to compute the effects of various transuranium isotope credits on the fuel-cycle costs for normal fueling methods, and 5) to compute the effect of various transuranium isotope credits on the fuel-cycle costs for special fueling methods that increase the power reactor production of transuranium heat sources.

TRANSURANIUM HEAT SOURCES AND STUDY SCOPE

$Curium^{242}$ and Am^{241}, while decay heat isotopes in their own right, are not the primary isotopes of interest in this study. They enter into the investigation because they are intermediate isotopes to Pu^{238} and, to a less extent, Cm^{244}. Figure 1 illustrates the reason for the special interest in Pu^{238} and Cm^{244}: during the first 5-year period, the heat generated by Pu^{238} drops only 3% as against 16% for Cm^{244} and 99% for Cm^{242}; and although the heat produced by Am^{241} remains nearly constant, this constant level is low. It is interesting to note, however, that the high initial heat output of Cm^{242} (See Table II) makes it easily comparable to Po^{210} in short-period applications (6 months to a year).

Table II

CHARACTERISTICS OF SUBJECT RADIOISOTOPIC HEAT SOURCES

	$Plutonium^{238}$	$Americium^{241}$	$Curium^{242}$	$Curium^{244}$
Specific Power, Watt/g	0.56	0.11	120	2.8
Half-Life, Yrs.	89	458	0.45	18
Isotopic Purity, %	80	90	90	90
Compound Form	PuO_2	Metal	Cm_2O_3	Cm_2O_3
Density of Compound g/cm^3	10	11.7	11.75	11.75
Specific power of Compound, Watt/g	0.39	0.1	98	2.3
Power Density, $Watt/cm^3$ of Compound	3.9	1.17	1150	27
Volume for 2 kW Heat, cm^3	513	1710	1.74	74

In common with other isotopic heat sources, the four transuranium isotopes Pu^{238}, Cm^{244}, Cm^{242}, and Am^{241} generate valuable amounts of heat for useful periods of time. Also, their reliability as a power source is unique. Table II indicates the major properties of these four transuranium isotopes.

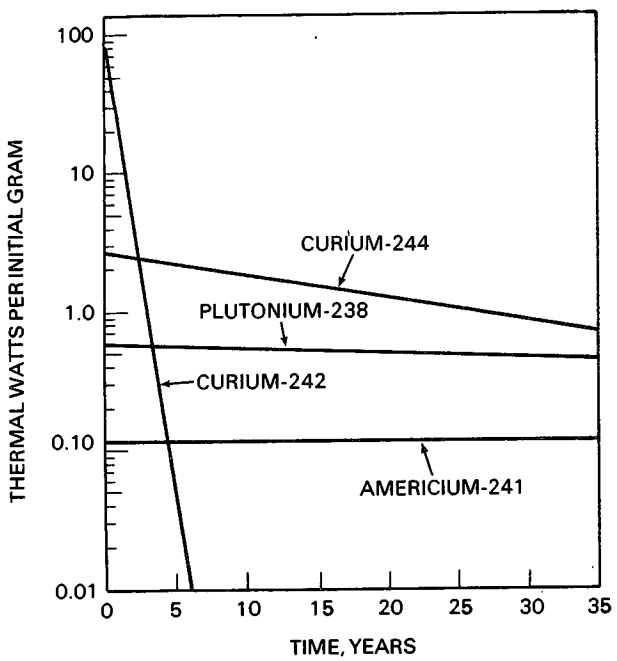

Fig. 1. Heat Rate of Four Transuranium Isotopes

Plutonim^{238} and Cm^{244} are particularly suitable for use in space or in other types of remote operation: they both have half-lives that are more than several months but less than 100 years, their specific heat is not less than 0.1 watt per gram, and the chemical separation of the product and target is economically feasible. As for radiation, Cm^{244} spontaneously emits 50 times as many neutrons as Pu^{238}, and this may detract from the usefulness of Cm^{244} as a power source near operating personnel; pure Pu^{238} does not have this possible drawback because its radiation is limited mainly to alpha emission, which is easily shielded. Pu^{238} contaminated with Pu^{236}, which eventually forms Thallium^{208} can lead to a gamma radiation problem. Accordingly, special Pu^{238} synthesis techniques may be necessary for Pu^{238} application such as heat sources for heart pacemakers and implanted hearts. As regards toxicity, Cm^{244} ranks 20th, whereas Pu^{238} ranks 6th. Also, Pu^{238} has a greater critical mass problem than Cm^{244}: at a density of 10 g/cm^3 the critical mass of Pu^{238} as an unreflected sphere is about 32 kg, or 18 kW; that of Cm^{244} is about 58 kg, or 162 kW.

The major sources of Pu^{238} and Cm^{244} are from U^{236} and Pu^{242} respectively. These in turn result from neutron absorption in fissile species (U^{235} and Pu^{239}-Pu^{241}) that do not lead to immediately fissioning species. Thus, to increase the production of Pu^{238} and Cm^{244} one would consider irradiation under circumstances to increase the capture to fission ratios (σ_c/σ_f) of U^{235}, Pu^{239} and Pu^{241}. Thus, if there were a market for U^{236} and Pu^{242} as precursors to Pu^{238} and Cm^{244}, respectively, it might be economical to alter the reactor fueling system so as to obtain a higher value of alpha for U^{235}, Pu^{239}, and Pu^{241}. Furthermore, if there were a sudden market for transuranium heat-source isotopes, a short-term windfall situation would be created for those holding U^{236} and Pu^{242}, which at present have no market price.

Although a market for a transuranium heat-source isotope would necessarily create a market price for its precursor isotopes, or target material, this price must be known before there can be an incentive to deliberately irradiate target material in power reactors. This report reviews how such a price, which is the equitable price of target material, can be approximated. Briefly, this equitable price is defined as the target indifference price--that is, the amount of money that a reactor operator pays for a target whose product yields an income sufficient to make the fuel-cycle cost of slightly enriched uranium with the target equal to the fuel-cycle cost of the same fuel without the target. The target indifference price is also called the target break-even price, because with either type of fueling method a reactor operator breaks even on his fuel-cycle costs.

This review then includes how various market prices for Pu^{238} and Cm^{244} products would change target indifference prices. The effect of processing costs on the net indifference price of a special target like Np^{237} is a matter of subtracting.

FORMATION OF TRANSURANIUM ISOTOPES

Plutonium^{238}, Cm^{244}, Cm^{242}, and Am^{241} are formed by a combination of neutron captures, beta decays, and alpha decays. Figure 2 shows the major sequences in the production of these isotopes. Deleted are intermediate isotopes whose half-lives are not long enough to appreciably affect the power reactor production of transuranium heat-source isotopes. A typical example is that of Np^{238}. Its elimination from the computer code increases the power reactor production rate of Pu^{238} less than 1% at a normal power reactor flux of 1 x 10^{13} neutrons cm^{-2} sec^{-1}. At a flux of 1 x 10^{15} neutrons cm^{-2} sec^{-1}, the error would be 34%.

Also omitted in this sequence are some reactions that significantly affect the production of impurities. The Np^{237} n-2n reaction is one example. This reaction produces Pu^{236} (2.85 year half-life) alpha-decaying into U^{232} which, after several alpha decays, forms the hard gamma emitter Thallium^{208}. Thus for long missions (10 years), the gamma activity of Pu^{238}-Pu^{236} mixture would be increased from almost nothing to a significant amount. In theory, calculations should include all those reactions--n-2n's, n-3n's, γ-n's etc.,--that significantly affect product purity. But in practice, such calculations cannot always be provided, either because too much computer calendar time would be required to account for all the important reactions in the isotopic mishmash of each element under consideration or because the relevant cross section and spatial flux detail is lacking. In practice, therefore, the contamination levels of transuranium isotopes cannot be accurately determined except by chemical analysis of discharged fuel and targets under anticipated operating conditions.

In spite of its "busyness", Fig. 2 shows that Pu^{238} can be formed in two ways--namely, via Np^{237} or Am^{241}. In the sequence shown in Fig. 3 there are three steps: U^{235} captures a neutron to form U^{236}; U^{236} captures a neutron to form Np^{237} and Np^{237} captures a neutron to form reasonably pure (85%) Pu^{238} (Np^{237} is also formed by the U^{238} n-2n reaction and decay of Am^{241}). A major Pu^{238} contaminating step is neutron capture in Pu^{238} to form Pu^{239} during the irradiation of the remaining Np^{237}. In the above sequence, however, there is one assumption, namely, Np^{237} is irradiated only after it is isolated from the U^{238} in the slightly enriched uranium fuel; otherwise, the U^{238} will form so much Pu^{239} that the specific heat of the Pu^{238}-Pu^{239} mixture will be tremendously reduced (about 99%) from that of reasonably pure Pu^{238}. (About 65% pure Pu^{238} can be produced by irradiating 90% enriched uranium.)

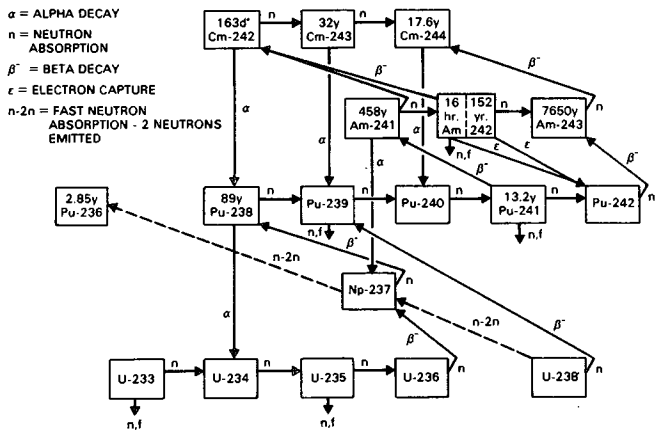

Fig. 2. Principal Nuclear Reactions for Transuranic Isotopes at Typical Power-Reactor Flux Levels

Shown in Fig. 4 is a way that essentially isotopically pure Pu^{238} can be formed requiring two steps (but only one neutron), namely, Am^{241} captures a neutron to form Cm^{242}, and Cm^{242} alpha-decays into Pu^{238}. To obtain Am^{241}, one can allow Pu^{241} to decay either in or out of the reactor. Plutonium^{241} that is separated and allowed to decay outside the reactor will form pure Am^{241}, whereas Pu^{241} decay inside the reactor during irradiation will form a mixture of Am^{241} and Am^{243}, and this mixture will contain less Am^{241} because of the neutron captures by Am^{241}. It should be noted that the rate at which Pu^{238} is formed from Am^{241} will be limited both by the availability of plutonium with a high content of Pu^{241} and by the Pu^{241} half-life of 13 years.

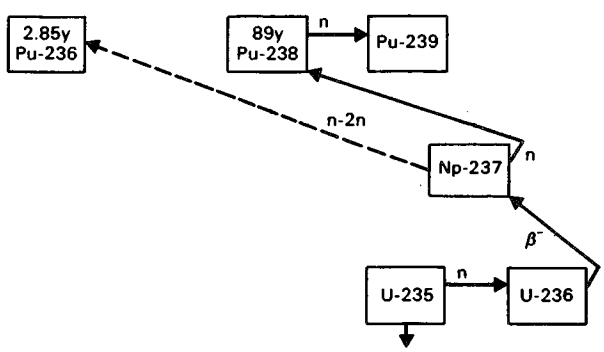

Fig. 3. Principal Nuclear Reaction For Pu^{238} Production From Np^{237} at Typical Power Reactor Flux Levels

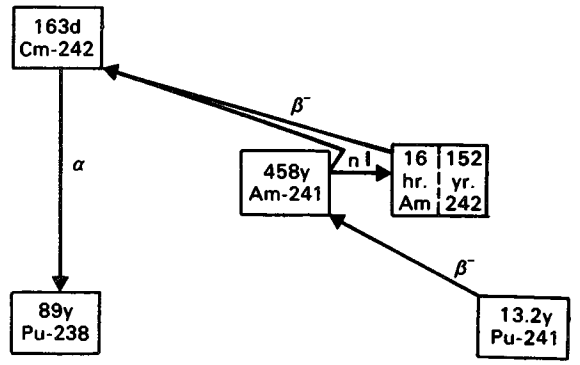

Fig. 4. Principal Nuclear Reactions for Pu^{238} Production From Am^{241} at Typical Power Reactor Flux Levels

In contrast to Pu^{238}, there is essentially only one way to form Cm^{244}, namely, by neutron captures in Am^{243}, which is formed by neutron captures in Pu^{242} and to a less extent, in Am^{242} as shown in Fig. 5.

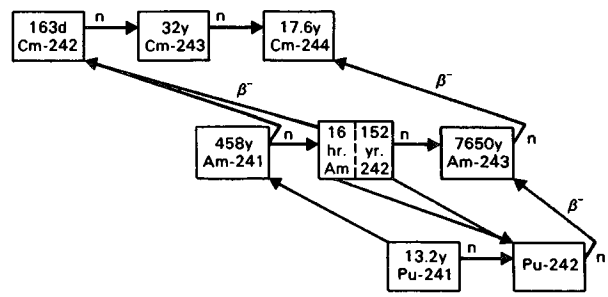

Fig. 5. Principal Nuclear Reactions for Cm^{244} Production at Typical Power-Reactor Flux Levels

Curium^{244} also has a purity problem. For when plutonium (a mixture of Pu^{239}, Pu^{240}, Pu^{241}, and Pu^{242}) is irradiated, both Am^{241} and Am^{243} are formed. Consequently, Cm^{242} is always produced with the Cm^{244}. And since Cm^{242} has an initial heat output 50 times greater than that of Cm^{244}, the heat output of Cm^{244}, which is relatively stable, will be temporarily masked if there is much Cm^{242} in the curium mixture. Moreover, the thermal energy of Cm^{242} decay can pose an obnoxious cooling and handling problem of the irradiation capsules. The purity problem, however, diminishes with time because of the decay of shorter half-life of Cm^{242} as shown in Table III. Thus, after 695 days, Cm^{244} is 99% pure and is considered usable, though the heat output of the mixture will drop about 10% in the first year of use and 5% the next.

PRODUCTION OF TRANSURANICS

It should also be noted that the *amounts* of Pu^{238} and Cm^{244} that can be ultimately produced depends on 1) the alpha (σ_c/σ_f) of U^{235}, Pu^{239}, Pu^{241}, and, to a less extent, U^{233}, and 2) also to a less extent, the

n-2n reaction of U^{238}. Furthermore, theoretically, the value of alpha, particularly in the case of U^{235} and Pu^{239}, is sensitive to the energy spectrum of the incident neutrons. Because a higher alpha means less fissioning and more production of transuranium isotopes, there is a possible trade-off between the two, depending on the market prices for the heat from fission in the reactor and for the heat from decay out of the reactor.

in hard LWR spectrums. The higher cross sections speed up reaction rates and increase product concentration in targets as well as the production per year for a given reactor. However, the harder spectrums do not increase the ultimate production of transuranics that are theoretically possible except for decreasing intermediary decay losses (of great importance if higher isotopes such as Cf^{252} are desired). Even so, the ultimate production capability of transuranics in power

Table III

RELATIVE POWER SUCCESSIVELY GENERATED BY A CURIUM MIXTURE INITIALLY CONTAINING 86% WEIGHT CURIUM-244

Days After Separation	Curium^{244} Purity(a)	Relative Power From Mixture			Number of Cm^{242} Half-Lives After Separation
		Cm^{242}	Cm^{244}	Pu^{238}	
0	86.12	142	21	0.00	0
43	88.12	119	21	0.11	0.4
206	93.58	59	20	0.38	1.4
369	96.62	30	20	0.52	2.4
532	98.25	15	20	0.59	3.4
695	99.10	7	19	0.62	4.4
858	99.54	4	19	0.63	5.4
1021	99.77	2	18	0.64	6.4

(a) $100\% - \%Cm^{244} = \%Cm^{242}$

However, because of self and cross resonance shielding encountered in power reactor fuel embodiments, there is not as much variation in alpha for the fissile isotopes as may be expected on the basis of the dilute cross section. For example, in soft spectrum LWRs, the effective alpha of U^{235} and Pu^{239} are approximately .22 and .52 respectively whereas in hard spectrum LWRs the respective alpha's are .29 and .55. Nonetheless, the effective cross section of some transuranics underlined in Table IV can be doubled or tripled

reactors is large on the basis of the neutron capture without fission that can be achieved by the intrinsic conversion of U^{235} to U^{236} and Pu^{239} to Pu^{242}. Producing Pu^{238} from all of the U^{236} formed in LWR's would provide about 82 kW per 1000 MWe reactor year to which 1.5 kW should be added for Pu^{238} from the Np^{237} formed by the n-2n reaction U^{238}. For the 50 to 100 reactors that should be operating for the next 30 years this amounts to 4000 to 8000 kilowatts of heat per operating year. At 1000 dollars per watt, this represents 4 to 8

Table IV

SOME ISOTOPE PROPERTIES(a)

Isotope	Effective Cross Sections(b)		Alpha of Fissile Isotopes	
	Soft Spectrum	Hard Spectrum	Soft Spectrum	Hard Spectrum
Th^{232}	8.4	11.5		
U^{235}	602	597	0.22	0.25
U^{236}	23	11.8		
U^{238}	5.0	14.4		
Np^{237}	180	260		
Pu^{238}	453	397		
Pu^{239}	1565	1914	0.50	0.55
Pu^{240}	559	1918		
Pu^{241}	1602	1573	0.37	0.39
Pu^{242}	88	497		
Am^{241}	874	1605		
Am^{242}	7220	7140		
Am^{243}	146	553		
Cm^{242}	18	18		
Cm^{244}	14	13		

(a) Includes resonance data.
(b) The effective cross sections include the effect of thermal flux depression which increases with an increasing spectral hardness. The enrichment for each spectrum was sufficient to attain a fuel exposure of 25 MWd/kg.

billion dollars/year. Of course, this yield cannot be achieved; but even if only 1/4 of it were, the numbers are still impressive and would help defray processing costs for waste disposal. To this figure should be added the potential Pu^{238}, Cm^{242} and Cm^{244} production that stems from the in situ Pu^{239} and Pu^{241} destruction occurring in the fuel cycle which accounts for about 40% of the heat energy (55% U^{235} and 5% U^{238}) in an LWR. If the unfissioned Pu^{241} is all converted to Cm^{244} one can add 106 kW/year of heat/1000 MWe to the 83.5 kW/year energy from Pu^{238} via U^{236} and n-2n on U^{238}. For 50 to 100 reactors then a total of 9000 to 18,000 kW/year of decay heat capacity is theoretically possible (9 to 18 billion dollars at $1000 per watt). Again, this cannot be realized because of transmutation and fission of precursors (especially for Cm^{244}).

It may be useful to measure the potential production of Pu^{238}, Cm^{242}-Pu^{238}, and Cm^{244} with a theoretical "instantaneous" conversion in much the same way we looked at potential bred fuel production before the proliferation of large computers. Using the values from Table IV, the maximum instantaneous conversion ratio for Pu^{238} from U^{235} is 0.2, for Cm^{242}-Pu^{238}, .355, and for Cm^{244} from Pu^{239}, 0.1. Of course, from a watts viewpoint (not energy) the potential decay heat conversion for Pu^{239} to Cm^{244} is greater than U^{235} to Pu^{238} by $\frac{.1 \times 2.8 \text{ watts/gm}}{.2 \times .56 \text{ watts/gm}} = 2.5$.

Many studies* and seminars to examine ways to economically convert U^{236} to U^{238} and Pu^{242} to Cm^{244} were completed in the early 1960s. Also included in these studies was the collection of Am^{241} from the decay of Pu^{241}. As indicated, Am^{241} is used as an ionizing, α-n and decay heat source in its own right. However, the major investigation here involves neutron absorption in Am^{241} to form Cm^{242} (163 day half-life) which emits an alpha particle to form Pu^{238} (Cm^{242} is a 120 W/gram heat source). Rather obviously, trade-off studies were performed of forming extremely pure Pu^{238} by storing Pu^{241} on the one hand and forming Cm^{244} by irradiating Pu^{241} forming Pu^{242}-Am^{243} on the other. Providing even a comprehensive synopsis of these studies leads to a sizable report, thus, only their major attributes will be reviewed here.

As indicated, we can only highlight the results of the many studies of higher transuranic production. The many combinations of beginning isotope composition, effective neutron spectrums, and recycle strategies leads to a never ending number of cases. While many alternatives to study may warm the hearts of fuel cycle analysts, they can, and yet are, a "pain" to the decision maker. Nonetheless, we shall not be overwhelmed by the analysts nor daunted by potential decision makers. To begin, Fig. 5a indicates how the time rate of Np^{237} production can be increased (from lowest curve to higher curves) by hardening the neutron spectrum and by recycling U^{236} containing U^{235}-U^{238} from a single irradiation of slightly enriched uranium (S.E.U.). (Fully enriched U^{235} was mixed with the spent S.E.U.)

Next attention is drawn to Fig. 6 relating Np^{237} irradiation time and the decrease in net Pu^{238} production as regards both quantity/yr and quality (compromised by neutron absorption in Pu^{238} to form Pu^{239}/. Nonetheless, Pu^{238} production from U^{236} is a relatively straightforward operation involving irradiation of Np^{237} when once isolated. In turn, Np^{237} production can be increased and concentrated by arranging to recycle U^{236} in spent U^{238}-U^{238} fuels without deleting the U^{236} by supplying it as feed to the diffusion cascade or even centrifuge plants geared to enrich U^{235} in U^{238}.

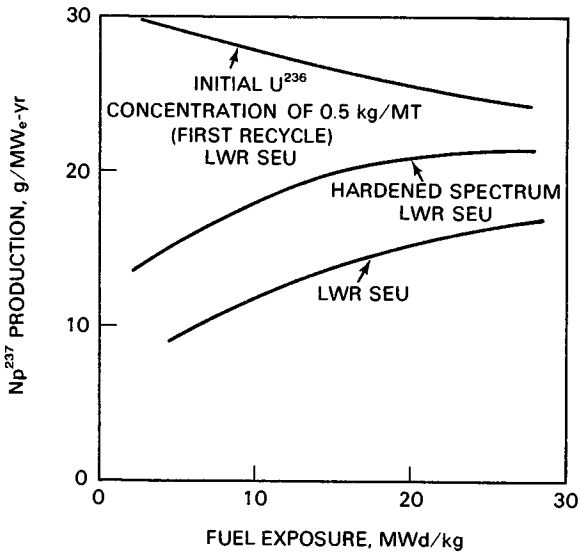

Fig. 5a. Np^{237} Production for Various Conditions

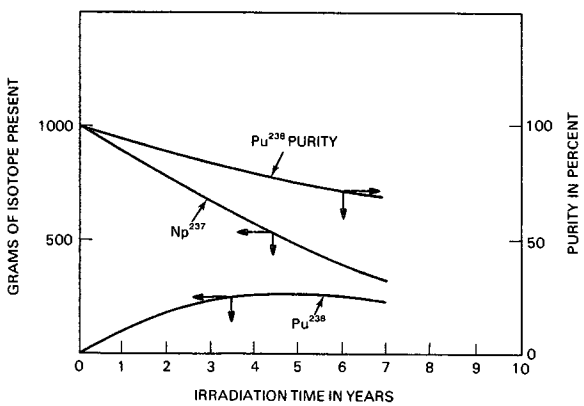

Fig. 6. Np^{237} Target Depletion and Pu^{238} Product Formation (Simulated LWR)

As shown in Fig. 7 and as is obvious, Cm^{244} production per MWe-yr can be increased by using high Pu^{242} composite feedstocks for enriching uranium. For sake of orientation, the uppermost curve is for plutonium from a cumulative S.E.U. fuel exposure of about 50,000 MWD/ton and the intermediate curve is for plutonium from a cumulative S.E.U. fuel exposure of about 35,000 MWD/ton. Not shown, of course, is the Am^{234} production which is about 3 times greater and can be used as feed for relatively direct Cm^{244} production.

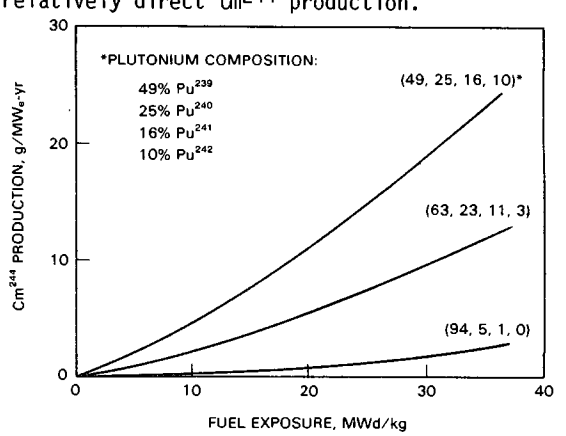

Fig. 7. Cm^{244} Production in an LWR for Various Plutonium Compositions in Natural Uranium

Shown in Fig. 8 is the influence of neutron spectra on the *rate of* Cm^{244} production from the plutonium composition utilized to prepare the middle curve of Fig. 7 (plutonium from S.E.U. fuel irradiated to about 35,000 MWD/ton. The difference between the soft spectrum LWR and the other is worth noting because soft spectrum (more open lattice) LWR's are normally thought of to maximize the value of plutonium recycle from the standpoint of maximizing fission energy rather than maximizing the value of neutrons in the larger sense occasioned by a market for decay heat sources in addition to central station power.

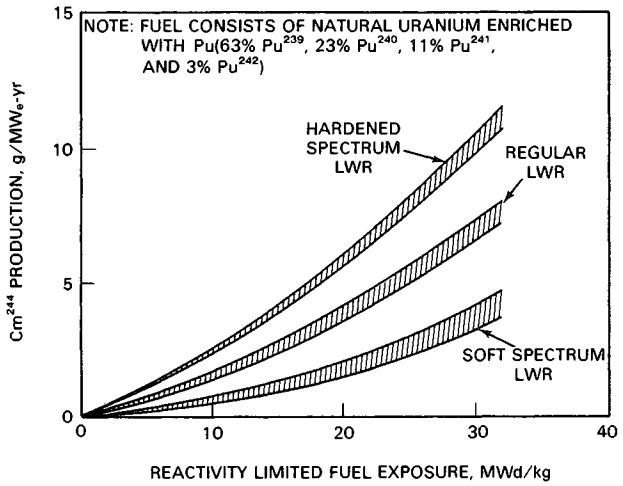

Fig. 8. Cm^{244} Production for Various LWR Neutron Spectra

Some of the more exotic systems studies are shown in Fig. 9. Of even additional complexity is to add the Np^{232} and U^{236} produced with the U^{235} driven cases. Such cases may be of greater interest today if, indeed, advanced isotope enrichment systems can selectively remove Pu^{238} from plutonium mixtures. It then becomes interesting to simply remove fission products and recycle spent fully enriched uranium, plutonium, thorium, or uranium mixtures and not rely on special capsules to keep products isotopically separated.

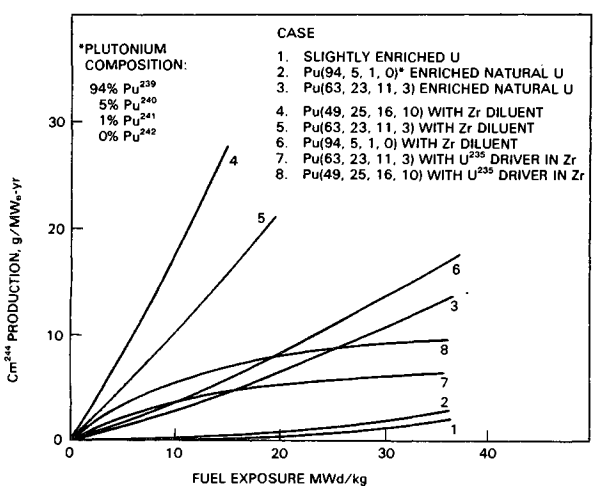

Fig. 9. Cm^{244} Production From Various Fueling Schemes in a Simulated LWR

INDIFFERENCE PRICES FOR TARGET ISOTOPES

It was observed that when the initial plutonium composition contains a higher percentage of Pu^{242}, the Cm^{244} production rate will be greater. And, as was pointed out, the production rate would have been even greater if americium from the previous cycle had been included. Consequently, if a price is assigned to Cm^{244}, a value will also exist for the americium and plutonium isotopes, particularly Pu^{242}; and this value will reflect both the value (plus or minus) they already have as thermal reactor fuels and that due to them as Cm^{244} producers. However, that part of the value due to them as Cm^{244} producers will depend both on their nearness to Cm^{244} in the production sequence and on the difficulty with which they are moved through the sequence.

The equitable price relationship between a product isotope price and the value of a target isotope is adequately expressed (as a first approximation) by the target indifference price. This price is the amount that a reactor operator pays for a target whose product yields a return sufficient to make the fuel-cycle cost (the sale of targets or product material formed from slightly enriched uranium must also be included when calculating the fuel-cycle cost) of slightly enriched uranium fuel with the target equal to the fuel-cycle cost of the same fuel without the target. When a target has such a price, a reactor operator is "indifferent" costwise to the two types of fuel since both fuels have the same fuel-cycle cost; or at such a target price, a reactor operator breaks even with either type of fuel.

Figure 10 shows the target indifference price of Np^{237} for a given Pu^{238} market price. If the market price of Np^{237} is less than the Np^{237} indifference price, a reactor operator could lower his fuel-cycle cost by purchasing the target, irradiating it, and selling the product Pu^{238}. The target indifference price, therefore, is the equilibrium point (equitable price) toward which the market price of a target would soon gravitate, eliminating windfall situations. But if the product (Pu^{238}) market price changes, so will the equilibrium point, the intersection of the two curves.

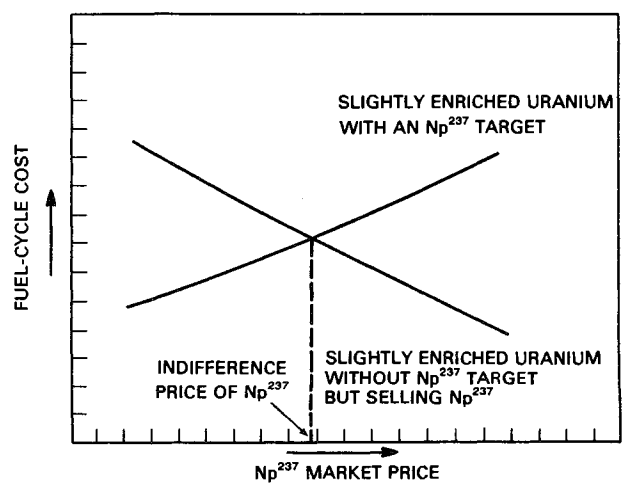

Fig. 10. Indifference Price of Np^{237} for a Given Pu^{238} Price

A calculation was performed to determine what effect various Cm^{244} market prices would have on the indifference prices of Cm^{244} precursor isotopes produced in a water reactor. Twenty cases were selected on the basis of the statistical requirements necessary

to obtain a meaningful set of indifference prices from a minimum number of cases. This explains why some cases in Table V have concentrations of pure isotopes and some cases have combinations of isotopes that do not normally occur during irradiation. Furthermore, for any given Cm^{244} price in the calculation, the price of each target at each of its locations in the production sequence was adjusted by an iterative procedure until the fuel-cycle costs of all 20 cases were equal to within 0.01 mill/kWh_e of the fuel-cycle cost of slightly enriched uranium without a target but selling transuranics to the systems. The results of the calculations are shown in Fig. 11. Notice in the figure that the indifference price of Pu^{239} remains the most constant, slowly rising from \$30/g, which is equal to its fuel value in a thermal reactor based upon present uranium and separative duty costs. However, Pu^{242}, which is closer than Pu^{239} to Cm^{244} in the production sequence, acquires a credit sufficiently high to increase greatly the value of plutonium fuel, in which it is normally a parasite that reduces the plutonium fuel value, as shown in the equation: Plutonium value = A - B (% Pu^{242} in plutonium). (With sales of higher transuranic isotopes the value equation starting with Pu^{239} is A + B + C + D because both Pu^{240}(B) and Pu^{242}(D) have values.

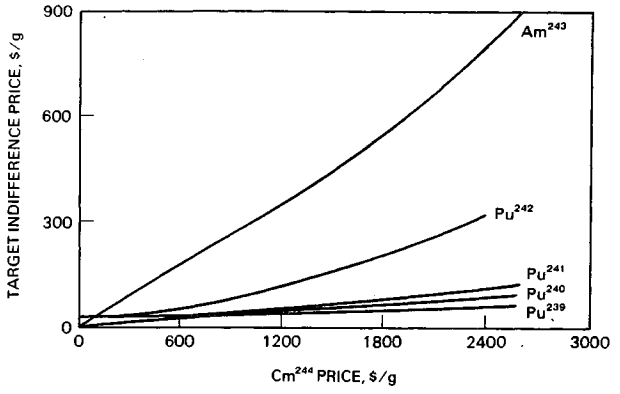

Fig. 11. Target Indifference Prices as a Function of Cm^{244} Credit (Simulated LWR)

Table V*

TARGETS AND TARGET CONCENTRATIONS USED FOR DETERMINING THE INDIFFERENCE PRICES OF ISOTOPES LEADING TO CURIUM^{244}

Case No.	Isotope Target Concentrations, g/cc				
	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}	Am^{243}
1	0.1				
2	0.5				
3		0.1			
4		0.5			
5			0.1		
6			0.5		
7				0.1	
8				0.5	
9					0.1
10					0.5
11	0.05	0.05			
12	0.025	0.025			
13	0.05	0.05	0.05		
14	0.025	0.025	0.025		
15	0.05	0.05	0.05	0.05	
16	0.025	0.025	0.025	0.025	
17	0.05	0.05	0.05	0.05	0.05
18	0.025	0.025	0.025	0.025	0.025
19	0.064	0.022	0.011	0.003	
20	0.128	0.044	0.022	0.006	

*This table is based on the statistical requirements for obtaining a meaningful set of indifference prices from a minimum number of cases. This explains why some cases have concentrations of pure isotopes and some cases have isotope combinations that do not normally occur during irradiation.

Another calculation, based on the same method, was performed to determine the effect that different Pu^{238} credits would have on the indifference prices of Np^{237} and U^{236}. But this time, because of the shorter production sequence, only 8 cases were involved. The results of the calculation (see Fig. 12) show that Np^{237} is worth from one-third to one-half the product price and that U^{236}, normally a parasite, is worth from \$4/g to \$10/g, depending on the price assigned to Pu^{238}.

Figure 13 shows the various indifference prices of Am^{241}, another isotope leading to Pu^{238}. This curve is based on the assumptions that Am^{241} is irradiated as a target to form Cm^{242} and that all the Cm^{242} existing in the capsule at discharge decays into Pu^{238}. For the sake of simplicity, the indifference prices represented by the curve in Fig. 13 do not include any credits for Am^{243} and Cm^{244} formed during the irradiation of Am^{241}. Had these credits been included, more calculations involving interactions would have been necessary in order to relate the indifference price of Am^{241} to the market prices of Am^{243} and Cm^{244} as well.

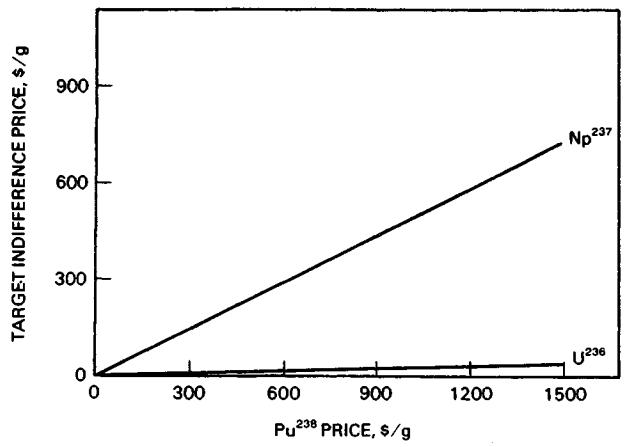

Fig. 12. Target indifference Price as a Function of Pu^{238} Credit (Simulated LWR)

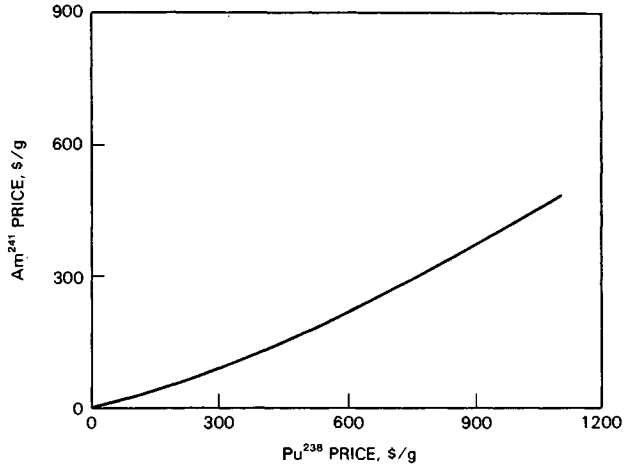

Fig. 13. Indifference Price of Am^{241} as a Function of Pu^{238} Credit

Since this report is mainly concerned with Pu^{238} and Cm^{244}, the indifference prices presented thus far have not included various market prices for Cm^{242} as a power source but rather we have valued Cm^{242} the same as Pu^{238} to which it decays in a short time. Including various market prices for this isotope would have increased the indifference prices of those isotopes that are also in the Cm^{242} production sequence.

THE TOTAL COST OF PRODUCING TRANSURANIC ISOTOPES FROM TARGET MATERIALS

Whether power reactors or special reactors should be used to produce transuranium isotopes is a question beyond the scope of this report. It may be that maximum production would require a combination of the two types. In any event, before a power reactor operator can afford to buy and irradiate a target, the target market price must be equal to the target indifference price. And since the total cost of producing transuranium isotopes from target materials includes the processing costs (fabrication, separations, etc.), the effect of these costs on the target indifference price will now be considered. According to Fig. 14, a reactor operator can afford to buy Np^{237} at \$100/g only if the Pu^{238} credit is at least \$215 and the Np^{237} processing costs are \$0. (The Np^{237} curve for \$0 *processing costs in Fig.* 14 is the same as the Np^{237} curve in Fig. 10.) But if the processing costs are \$10/g, then Np^{237} at \$100/g must have a Pu^{238} credit of at least \$250/g. On the other hand, if the Pu^{238} credit is \$267/g and the processing costs are \$50/g, then the highest price a reactor operator would be willing to pay for Np^{237} is \$100/g, the indifference price of Np^{237}. And if the Pu^{238} credit is \$324/g and the processing costs are \$100/g, the indifference price of Np^{237} is again \$100/g.

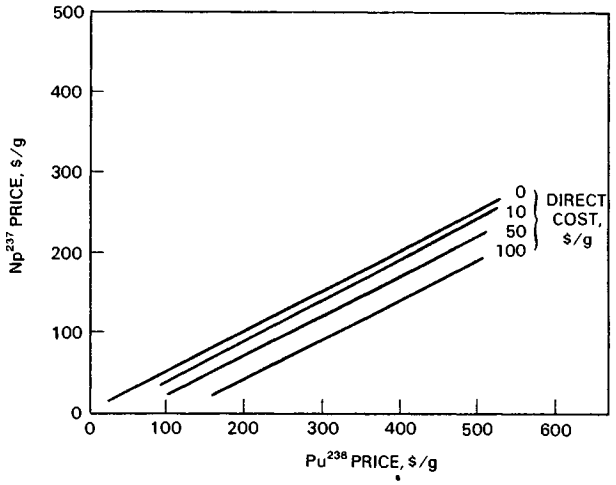

Fig. 14. The Effect of Processing Costs on the Indifference Price of Np^{237} Produced in a Simulated LWR

The total cost of producing transuranium isotopes from target materials can, therefore, be subdivided into the target indifference price and the processing costs; but the effect of processing costs on the target indifference price must be known before a reactor operator can determine whether he can afford to produce transuranium isotopes from target materials. Apart from the processing costs, the factors that go into determining the target indifference price are 1) the cost of target depletion, 2) a product purity correction, 3) the irradiation cost, and 4) the inventory costs of target and product. The processing costs, on the other hand, are made up of 1) the encapsulation cost, 2) fuel displacement cost, 3) shipping and storage costs, 4) recovery costs, 5) out-of-reactor decay costs and 6) interest on the working capital required. Depletion, irradiation, inventory, and working capital costs have been captured in these initial computations of indifference prices. The impact of the other costs can be estimated as shown for processing costs in Fig. 14.

MAJOR CONCLUSIONS

An example of the overall value of transuranics is shown in Table VI for S.E.U. fuel enjoying a consistent set of indifference prices as described in the foregoing paragraphs. While a figure in terms of mills/kwhr may not be compelling (even 3.6 mills/kwhr) in today's beleaguered nuclear enterprise, this figure does amount to twenty-five million dollars per year to a 1200 MWe reactor at .67 load factor. For 50 reactors, this amounts to \$1.25 billion per year, which could go a long way to help defray processing costs to simplify fission product disposal. As indicated, the \$1.25 billion is not a net figure, the net may only be 1/2 or 1/3 of this figure. Even so, it is impressive, especially when it involves removing major quantities of the longer-lived transuranics such as Np^{237} from waste streams in addition to yielding revenues for interim plutonium storage (through a

market for Am^{241}-Cm^{242}-Pu^{238} derived from Pu^{241} decay (13.5 year half-life).

Table VI

POTENTIAL VALUE TRANSURANIC CREDITS
COMPUTED 40,000 MWD/TON[a] SPENT LWR FUEL S.E.U.

Isotope	gms/ton	gms/MW_e-yr	Indifference Value $/gm	Value mills/kWe
U^{236}	6,670	137	20	.39
Pu^{239}	6,880	141	30	.60
Pu^{240}	2,610	53.7	35	.26
Pu^{241}	2,080	42.8	40	.25
Pu^{242}	820	16.8	80	.19
Np^{237}	736	20.3	115	.35
Am^{241}	69.6	1.92	385	.11
Am^{243}	120	3.31	270	.13
Am^{242}	1.1	.032	-	-
Cm^{242}	23.9	.658	900[b]	.08
Cm^{244}	32.9	.908	900	.13
Pu^{238}	226	6.24	900	1.14
TOTAL POTENTIAL CREDIT				3.63

25.5 x $\$10^6$ Total Potential Credit 1200 MWe .67 Load Factor.

(a) 24.2 x $\$10^6$ Yearly Credit With Fuel at 25,000 MWD/Ton.
29.8 x $\$10^6$ Yearly Credit With Fuel at 54,000 MWD/Ton.
(b) Cm^{242} Valued as Pu^{238}.

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