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VALUES IN SPENT FUEL FROM POWER REACTORS

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VALUES IN SPENT FUEL FROM POWER REACTORS

by

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TABLE OF CONTENTS

VALUES IN SPENT FUEL FROM POWER REACTORS

INTRODUCTION

Recent predictions indicate far greater nuclear power generation than that forecast only about 2 yr ago.[1] A renewed inquiry into the worth of the many useful and potentially exploitable elements present in discharged reactor fuels is called for. The economical recovery of these materials has a broad area of interest, including:

- The reactor designer who may wish to capitalize on design concepts that favor formation of certain products in highest yields
- The reactor operator who may expect credit for neutrons invested in the higher isotopes that are unavoidably produced
- The fuel processor who may profit from his efforts to recover these materials
- The power consumer who can expect to share in the credits to the reactor operator by obtaining cheaper power
- The developer of new enterprises through practical application of some of these materials that are available from no other sources, that may be competitive with conventional sources, or that have unique properties to fill new demands.

The results of an earlier study of this subject appeared in Reference (2).

OBJECTIVE

It is the objective of this study to appraise the magnitude of future amounts of by-products available from discharged reactor fuels, to establish the candidate materials which are most likely to have value, to utilize some simplified ground rules for appraising value and recovery cost, to estimate in a preliminary way the value of candidate materials in unprocessed fuels, and define the chemistry and technology of economical recovery processes. It is not intended that these estimates be precise but only that they have reasonable bases. Such information is considered sufficient to determine whether further interest or study is justified.

ASSUMPTIONS

1. There is an unlimited market for Pu^{238} and Cm^{244} at the projected prices of \$500 and \$1000/g. (3)*
2. A competitive situation exists for reactor fuel processing in combination with isotope recovery systems (more than one fuel processor in business).
3. The products will be available as most appropriate pure solid compounds or aqueous solutions except that the rare gases will be marketed as gases and the noble metals as metals.
4. Fission product radioisotopic heat sources (Sr^{90}, Cs^{137}, Ce^{144}, Pm^{147}) are assumed to be marketed as bulk products at near the "HIP" cost (for encapsulated sources) as established in Reference (4).
5. Formation and concentration of the products will be as established in References (5) and (6) for typical, low enrichment, power reactor fuel.
6. No isotopic separation processes are employed for any of the products.
7. Where credit for neutrons absorbed is employed to establish value, neutrons shall be valued at \$2500/g. (This is a minimum incremental cost.)
8. All fuels are assumed to have been irradiated to 25,000 megawatt days per short ton (MWd/t).
9. The cost of fuel processing is \$27,000/ton of uranium with one-half of this cost charged to plutonium (\$2/g for 25,000 MWd/t) and the other half charged to recovered uranium.
10. Both Np^{237} and Cm^{242} are considered as precursors of Pu^{238} and are valued accordingly.
11. Am^{243} is the precursor of Cm^{244} and is valued accordingly.
12. Other products formed by neutron addition have values determined by the value of the neutrons absorbed adjusted for the yield of such neutrons in progressing to the ultimate products.

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*Glenn T. Seaborg in "Nuclear Energy in the Next Decades," Vol. 1 Proceedings of the 1964 Annual Conference, Atomic Industrial Forum, San Francisco California, p. 17 states: "---Huge requirements, difficult to comprehend by todays standards, will exist for Pu^{238}, Cm^{244} and other transuranium isotopes."

13. The estimate of the cost of producing the materials is based on judgments involving the following considerations:
    a. Process complexity.
    b. Degree of integration with other companion products.
    c. Value of ultimate product: for example, Am^{243} value is high because it is the precursor of Cm^{244}, an extremely valuable product.
    d. The incremental value of neutrons (see Assumption 6).
    e. The relationship between the price of conventional raw materials and price of products. In a number of products this ratio is close to 10 = $\left(\frac{\text{price of product}}{\text{price of raw material}}\right)$.
14. Future prices for products other than for Pu^{238}, Cm^{244} and fission product heat sources are based on judgments of known costs and selling prices and estimates of prices which appear to be reasonable for the market.
15. The products are those materials which are either stable or of sufficiently long half-life and yields sufficient to assure multigram amounts being available per ton of fuel at the time of processing (6 mo after reactor discharge). In addition, some reasonable expectation for use must be visualized. Unless an isotope which also occurs in nature is of high value (about $1/g or more) it is not included (see Table VII, p 39 for current prices of some costly commercial elements). Materials of low fission yield or low value are thus excluded.

SUMMARY

The results of this study are shown in Table I. In general it is concluded that there is expectation of only modest credit to the reactor operator or fuel owner for fission products. However, the fuel processor may find items of profitability among these. This conclusion may be somewhat biased unfavorably by the high production cost assumed for some of these materials.

At the level of credit for absorbed neutrons utilized in this study, very high returns to the fuel owner or reactor operator are indicated

TABLE I

ESTIMATED VALUES OF SOME ELEMENTS AND ISOTOPES IN FUEL AS DISCHARGED FROM POWER REACTORS

Element or Isotope	Potential Use	Half-Life, Years	Specific Power, W/g	Current Prices: Cost, $	Current Prices: Unit	Current Prices: Calculated to $/g	Assumed Future Price,[a] $/g	Assumed Production Cost, $/g	Estimated Value in Fuel, $/g	Approximate g/ton 25,000 MWd/t[b]	Estimated Value, $/ton Spent Fuel	
Kr^{85}	Special Light and Radiation Source	10.4	0.54	7.50	curie	3 045	300	200	100	17	1 700	
Sr^{90}	Heat and Beta Source	28	0.95	0.75	curie[c]	108	20	18	2	411	822	
Tc^{99}	Corrosion Inhibitor, Alloying Agent	2.1×10^5	--	90.00	gram[d]	90	11	10	1	628	628	
Rh	Industrial, Electrical, Decorative	Stable	--	6.00	gram	6.00	8	6	2	337	674	
Ru[e]	Industrial and Electrical	Stable (+1 yr Ru^{106})	--	1.85	gram	1.85	2	1.50	0.50	1 707	354	Fission Products $7 660
Pd[e]	Industrial, Electrical, Decorative	Stable	--	1.00	gram	1.00	1.25	1.00	0.25	976	244	
Xe[e]	Special Light Source	Stable	--	35.00	liter[f]	6	2	1.70	0.30	3 987	1 196	
Cs^{137}	Heat and Radiation Source	30	0.42	0.50	curie[c]	49	10	9	1	950	950	
Ce^{144}	Heat and Beta Source	0.78	25.6	0.30	curie	954	25	22.50	2.50	241	260[g]	
Pm^{147}	Heat and Radiation Source	2.6	0.33	1 630.00	watt[d]	670	30	25	5	133	335[h]	
U^{238}	Fertile Material	4.5×10^9	--	--	--	--	6.75/lb	6.75/lb	0	8.9×10^5	0	
U^{236}	Target for Np^{237}	2.4×10^7	--	--	--	--	11	0	11[i]	3 277	36 070	
Np^{237}	Target for Pu^{238}	2.2×10^6	--	500.00	gram[d]	500	100	80	20	316	6 320	
Am^{241}	Heat Source (and target for Cm^{242})	458	0.1	1 500.00	gram[d]	1 500	100	50	50	56	2 800	The Higher Isotopes $104 110
Cm^{242}	Heat Source (decays to Pu^{238})	163 days	120	167	watt	20 000	300	150	150	23	3 450[k]	
Am^{243}	Target for Cm^{244}	7 650	--	--	--	--	500	50	450	87	39 120	
Cm^{244}	Heat Source	17.6	2.8	--	--	--	1 000	100	900	22	19 800	
Pu^{238}	Heat Source	90	0.56	1 600.00	watt[d]	880	500	350	150	--	3 450	
Pu^{239}	Fissionable	2.4×10^4	--	10	gram	10	11	2	9[i]	5 453	49 100	Plutonium Isotopes $99 185
Pu^{240}	Fertile (and source of Pu^{241})	6.8×10^3	--	--	--	--	7[j]	2	5[i]	1 517	7 585	
Pu^{241}	Fissionable (and source of Am^{241})	13	--	10	gram	10	19[j]	2	17[i]	1 412	24 000	
Pu^{242}	Target of Am^{243}	3.8×10^5	--	--	--	--	41[j]	2	39[i]	386	15 050	
									Value: per short ton		$210 955	
									per pound		105.50	
									per kilogram		232.50	
									per kWH_e		1.055 mills[l]	

(a) Pure isotope or element in bulk form.
(b) These numbers vary with reactor spectrum and the figures here are approximate. (Values are from documents HW-78140 and HW-78141-1.)
(c) Nucleonics Week, April 4, 1962, AEC prices per curie.
(d) Atomic Industrial Reporter, March 7, 1962, AEC prices; also Nucleonics, April, 1961, page 70, for Pm^{147} and Pu^{238}.
(e) Would be mixed isotopes, essentially stable (except Ru); some have high fission yields, especially from Pu^{239}.
(f) Price for by-product material from liquid air manufacturing.
(g) At 1 yr after discharge.
(h) At 1 yr half-life.
(i) These values are based on credit for absorbed neutrons (at $2500/g neutrons effective in the production of higher isotopes).
(j) These assumed future prices are substantially greater than those now generally accepted as the fuel value of plutonium isotopes.
(k) As Pu^{238}, value included with Pu isotopes.
(l) At 33 1/3% efficiency thermal to electrical.

which could significantly lower the cost of power. This credit appears so large for certain products formed by neutron absorption that a more critical analysis of the value of neutrons or value of products is suggested. The very large credits for Am^{243} and Cm^{244} that are based on the extremely high costs of today's and projected future methods of producing these isotopes may not be consistent (still too high) when compared with anticipated costs based on neutron credits and expected simplified recovery technology.

Laboratory investigation of integrated processes for fuel and isotope processing is recommended, with particular emphasis on the technology of americium, curium, the noble metals, and the rare gases.

Credit for the fissionable plutonium isotopes at the published \$10/g figure[7] assures the soundness of the economics of fuel processing.

DISCUSSION

THE SITUATION

Recent forecasts and estimates of the growth of nuclear power are far more optimistic than earlier projections. The publicized low or competitive costs associated with the power plants recently proposed for the middle-Atlantic area account for these more liberal predictions. These statements indicate the justification for a second fuel processing plant before 1970 with a rapidly increasing fuel processing load in the following 10 yr. A nuclear power industry exceeding 80,000 Mw_e has been forecast by 1980 (Figure 1). Under such circumstances the amount of many elements and isotopes in fuel processing waste may reach such levels and values that, if they could be marketed, the fuel cycle economics could be appreciably influenced. In addition, there could be a great impact on existing marketing of the "natural" elements which are also produced in fission, such as rhodium, palladium, and xenon.

For example, rhodium warrants some special comment. Rhodium, a member of the platinum group of so-called noble metals, is by far the most expensive of all stable solid elements. Yet, even though it is rare and costly, it has such outstanding properties of high temperature durability and corrosion resistance that it is a fairly common, important, and useful

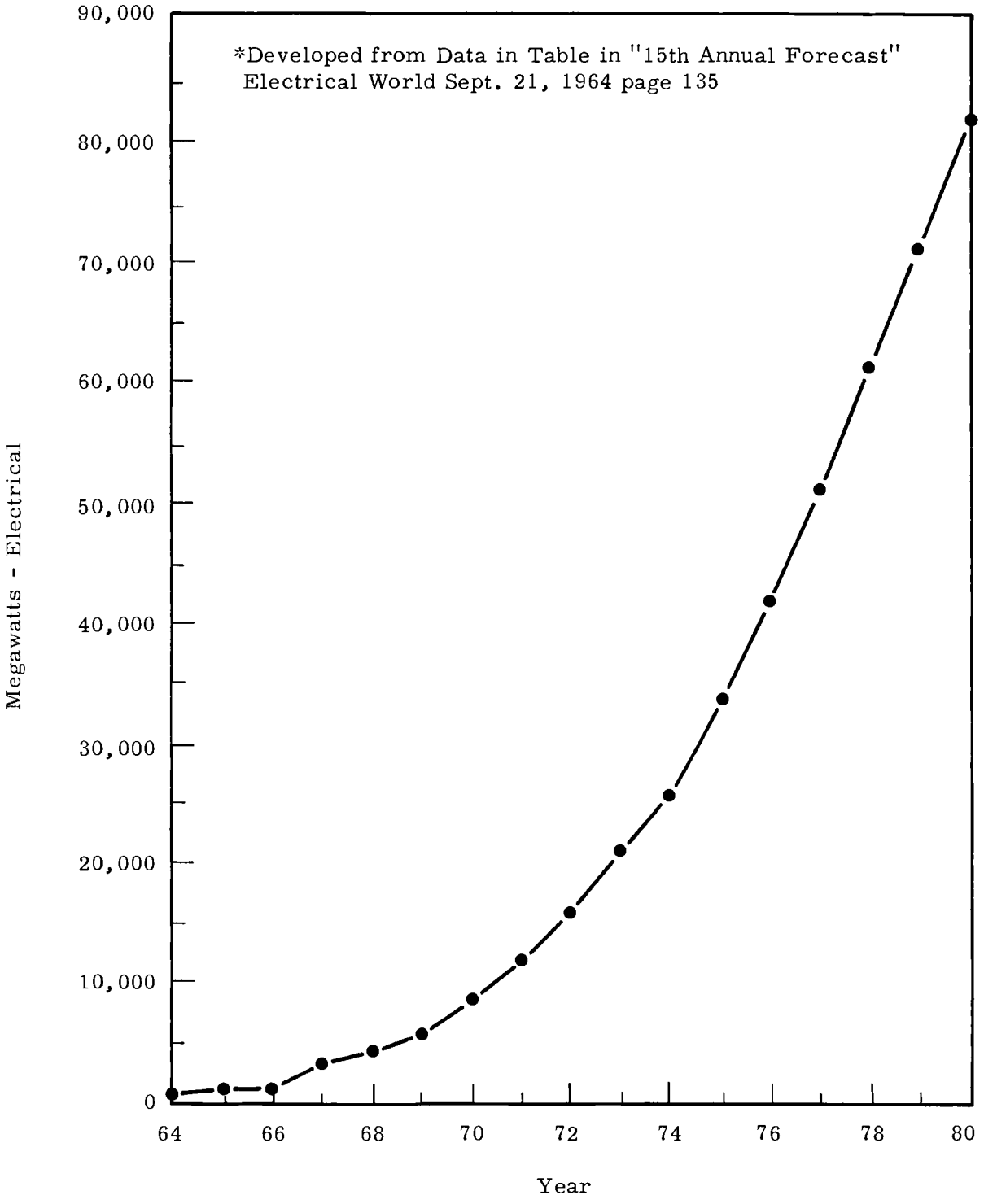

FIGURE 1

Estimated Growth of Nuclear Power in the United States*
(End of Year Basis)

article of commerce. (For example, a common brand of kitchen gas ranges uses a small coil of platinum-rhodium alloy wire to serve as the electric pilot light for the gas broiler.*) In nature it occurs as an element with only a single isotope. The short half-lives of most of its artificially produced isotopes assure that fission product rhodium will be essentially stable and the same as that found in nature.** As a fission product it exists among the group of lighter isotopes which form in highest fission yield and is only slightly less prevalent than Sr^{90}. Furthermore, from plutonium fission it appears in even greater yield than does Sr^{90} (Figure 2). Unless some practical recovery technique is found for this valuable element in the fuel processing wastes, more rhodium will be delivered to underground confinement annually by 1980 than was mined in 1962 worldwide and marketed in the U. S. ! Similarly, palladium resulting from fission would also comprise at that time a significant fraction of the palladium mined and marketed today.

Xenon, a rare and costly by-product of a few liquid air manufacturing plants, will be discharged in large quantities. The annual discharge of xenon from reactor fuel processing at about 1980 will be equivalent to that existing in about 5.5 cubic miles of air. Depending on the fuel dissolution process utilized, this xenon could be evolved in very high concentrations in the off gases. The three elements mentioned above are examples of fission products that are stable elemtns*** for which expanding markets already exist but which, for rhodium and palladium, most certainly have shrinking world reserves.

The situation with the radioactive elements and isotopes is even more exciting (or alarming), since these can only be derived from atomic energy processes and to date have not been generally available. Under the

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* "Industrial Research", November 1964, p. 9.

** An n, 2n reaction may produce active, 206 day Rh^{102}. However, this has not been established and thus appears to be only a potential problem.

***At least 20% of the palladium should, however, comprise Pd^{107}. This is a radioactive isotope of such long half-life and of such low pure beta energy that it is expected to be utilized in all normal commercial applications without need for control. No formation of Ag^{107} is anticipated.

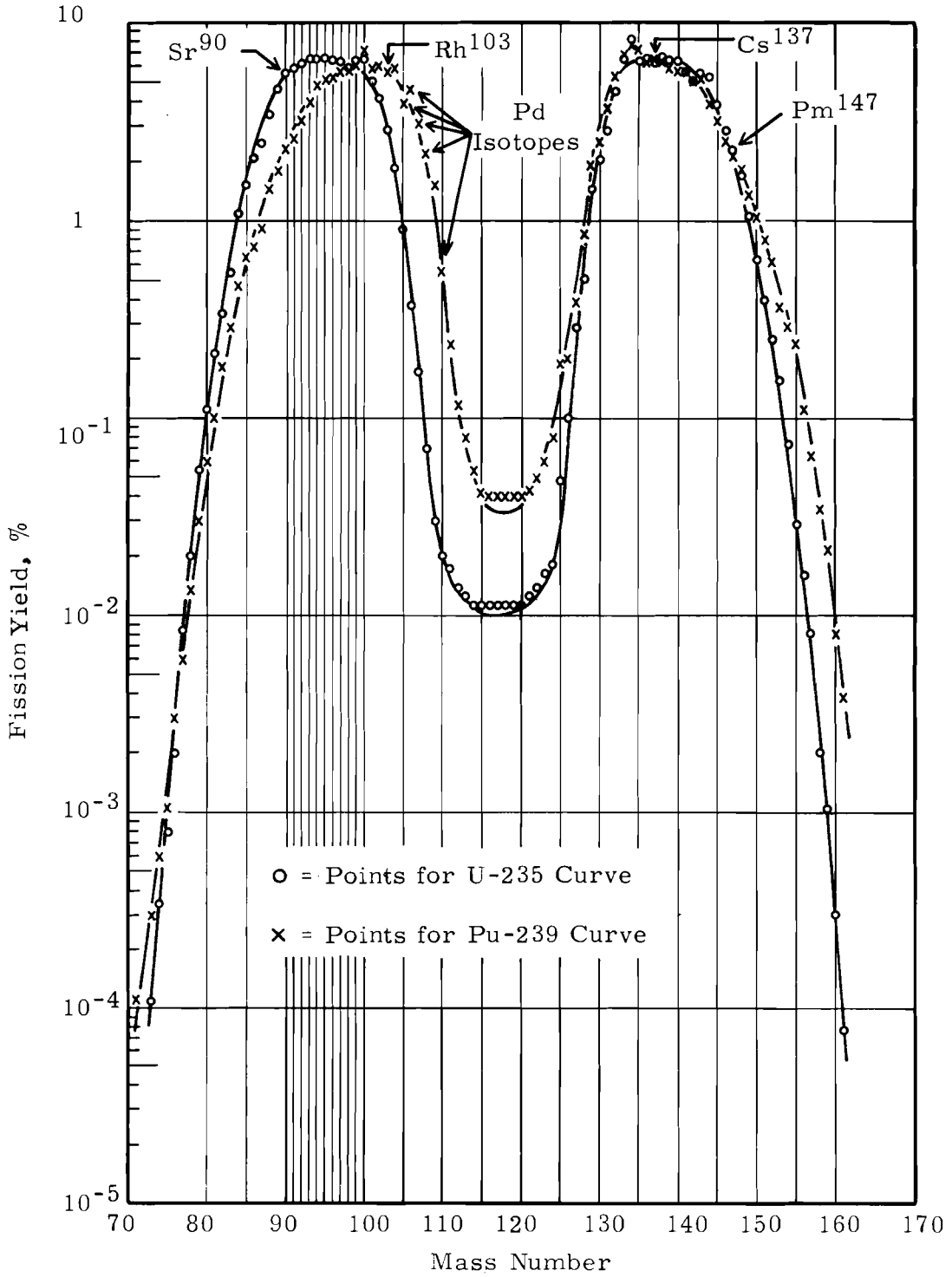

FIGURE 2

Yields from Thermal Fission of U^{235} and Pu^{239} from:
Katcoff, Nucleonics 18, no. 11, p. 201. 1960

projected conditions, tons of such materials will not only be available but will have to be safely disposed of. Hopefully, there will be developed economical, safe, and highly beneficial uses for most of these unique materials.

In addition, there will exist significant amounts of the higher isotopes. These are not produced by fission but by neutron capture in the nonfissioning fraction of the fuels. Among such isotopes is curium. Whereas today a major effort is going on at the Savannah River Plant to produce 3 kg of Cm^{244} by 1967, by 1970 an equivalent quantity may have been discharged in only about 140 tons of certain conventional power reactor fuels of high exposure. By 1980 nearly twenty times that amount could exist in the annual output of fuel processing wastes. Furthermore, it is conceivable that it will be extractable as a by-product at relatively low cost. The recycle of plutonium in thermal reactors would also contribute major increases in curium production.

Concurrently, the precursors of valuable isotopes such as Pu^{238} and Cm^{244} will also be at hand. The recovery and recycle of these precursors will further enhance the availability of these materials. Included among these are Am^{241}, Am^{243}, U^{236}, Np^{237}, Cm^{242}, Pu^{242}, Pu^{241}, and Pu^{240}. All of these should have values either based on their utility in producing the Pu^{238} and Cm^{244} or at least based on the neutron investments involved.

A greatly changed situation will exist when essentially all of the plutonium produced in power reactors is recycled. This will involve a further enhancement of the availability of the higher isotopes. In addition, this is the case not only for the higher isotopes but also for some of the potentially valuable fission products which are produced in higher fission yield from plutonium than from U^{235}. Rhodium and palladium are both among these.

This study is concerned essentially with those elements or isotopes which will be available annually in kilogram quantities from a plant that is processing normal power reactor fuels of low enrichment. Although many useful isotopes can also be produced by special target irradiations, the production of such materials is not a part of this review. Special and possibly useful isotopes whose fission yield is small and the higher isotopes,

beyond Cm^{244}, which would be produced in milligram amounts are also not included.

THE VALUE OF NEUTRONS

The intriguing prospects of using neutrons to produce unique materials encourages efforts to consider neutrons as raw material or essential material in conventional ways for arriving at values and manufacturing costs by the usual chemical processing industry methods. However, the variety of reactors and the special features of each make the appraisal of neutron cost or value a subject so clouded by the wide differences in bases and situations that there is an apparent reluctance to display any cost figures on the subject. It may have been concluded that neutrons are so expensive that their intentional use for the production of materials other than for defense (plutonium, tritium, etc.) is almost unthinkable.

However, some relatively uncomplicated situations can certainly be assumed to provide a basis for putting a dollar value on neutrons. There are also certain isotopes that have such high value when compared with costly yet conventional materials that the neutron cost involved in their manufacture could be actually almost insignificant. For example, for certain purposes, a most convenient means of producing a source of neutrons for research purposes is by preparing a mixture of Ra^{226} and beryllium. The alpha activity of the radium acts on the beryllium to produce a substantial output of neutrons (about 10^7 neutrons/sec per gram of radium present in such a device). Over the years radium has been available at about $15,000/g. Any other energetic alpha emitter could also be used in the same way. Furthermore, several of such emitters can be produced by neutron irradiation of certain target materials in reactors. Among these could be Th^{228} which has more than about 1000 times the alpha activity of Ra^{226}. Then, to be competitive with radium for use in these special neutron producers, Th^{228} would have to be available at a figure not in excess of a seemingly ridiculously high cost of $15,000,000 per gram! Technology is actually at hand to assure product costs far below this level.[8]

Th^{228} can be produced by irradiation of Th^{230}. Two grams of neutrons added to Th^{230} produces U^{232} which will decay to yield 228 g of

Th^{228}. These neutrons then should be worth $\frac{228 \times \$15,000,000}{2}$ or about $\$1,700,000,000/g$!

A reactor can be visualized in which nearly 1 g of net neutrons (neutrons available for plutonium or other isotope production) could be made available for each gram mole of U^{235} fissioned. With fully enriched U^{235} available at about \$12/g (July 1, 1962, AEC U^{235} price schedule), a net gram of neutrons should thus cost as little as about \$2800. This calculation is obviously unrealistic since it does not include such costs as fuel and target preparation, reprocessing, and use charges. On the other hand, costly, fully enriched uranium is obviously not the most economical source of neutrons, particularly if the reactor in which the irradiation is taking place is fueled with low enrichment or natural uranium. Assuming a figure of \$12/lb for natural uranium metal (thus containing U^{235} at \$3.75/g) neutrons may cost as little as about \$900/g on the same unrealistic basis! However, on such a basis the cost of neutrons needed to produce an isotope with an atomic weight of about 225 need thus be only about \$4/g of the element or isotope. Many isotopes can be mentioned that are valued at much higher figures: for example, Cm^{242} at \$20,000/g.[3] The conclusion can thus be reached that, in reality, for certain valuable elements or isotopes, the neutron cost component can really be minor.

Test or experimental reactors, of course, cannot be considered as the most economical or practical means of producing multigram quantities of any isotope or element, since in such small reactors too large a fraction of the total useful reactor volume and corresponding operating and fixed costs must be charged to any isotope production program involved. With such viewpoints, neutrons in large reactors appear as rather logical "raw materials" for the manufacture of useful products. Similarly, any reactor operation that produces recoverable and valuable elements or isotopes either intentionally, inadvertently, or as by-products could logically request credit at least for the value of the neutrons invested in these materials. Although this may not apply to fission products it could be argued to apply to all of the higher isotopes that can be shown to have value.

The following bases are regarded as reasonable to establish a likely minimum incremental value for neutrons.

Assume:

1. That a power reactor normally uses fuel of 2% enrichment under conditions of private ownership.
2. An increase of enrichment to 2.02% is needed to provide neutrons for target irradiation to produce special isotopes, with the fuel burned to the same final U^{235} content.
3. The reactor operates so that each gram mole of fissioned U^{235} yields one net gram of neutrons that is used for special isotope production.
4. In this operation no extra costs are involved in fabricating the 2.02% fuel or in its final chemical processing.
5. The cost of uranium of 2% enrichment is \$146.50/kg and for 2.02% fuel is \$148.60.

1 kg of 2.02% fuel contains 20.2 g of U^{235}
1 kg of 2.00% fuel contains 20.0 g of U^{235}

$\therefore$ For each kilogram of 2.02% fuel used, 0.2 g of U^{235} are consumed specifically for isotope production.

$\therefore \frac{235}{0.2}$ = 1175 kg of 2.02% fuel required for 1 g mole of U^{235} for 1 net gram of neutrons.

1175 (\$148.60 - \$146.50) = 1175(2.10) = \$2468

The incremental cost of a gram of neutrons under these conditions is therefore \$2468, or rounded off, \$2500.

Although such an estimate involves many simplifications it is realistic, since it probably represents the lowest range of neutron value. Only relatively minor changes in value occur at different enrichment levels, e.g., 1% or 3% instead of the 2%. The assumption of 1 g of net neutrons per gram mole of fissioned U^{235} is also optimistic; lower yields are probably certain with corresponding increase in actual neutron value. However, from the conservatively low \$2500 figure, minimum credits for higher isotopes are realized. This is regarded as a suitable approach for this study.

THE PRODUCTS

The products considered appropriate for a study such as this comprise those elements and isotopes whose annual output from a reactor fuel processing plant would be measured in not less than kilograms. These products may be highly radioactive ones, such as Pm^{147}; slightly radioactive ones, such as U^{236}; or even essentially stable ones such as rhodium or xenon. They would include both the fission products and those isotopes produced by neutron absorption in the elements of the fuel. Not included in this study are those isotopes which can be produced in reactor by neutron irradiation of special target materials. However, some reference is made to this class of materials in a paragraph at the end of this report.

Since a large portion of these products will be useful because of their intense radioactivity, it is considered worthwhile to emphasize the magnitude of their prospective future outputs. Since the AEC began distributing radioisotopes in 1946, the Oak Ridge National Laboratory has shipped a total cumulative amount of about 2, 350, 000 Ci of all isotopes including 360, 000 Ci of Sr^{90}, one of the commonest radioisotopes produced in highest yield in fission of U^{235}.[9] The Hanford Isotopes Plant[4] is proposed to produce 10, 000, 000 Ci/yr of Sr^{90} along with 140, 000, 000 Ci of three other isotopes. The annual availability of only these same four isotopes from reactor fuel in 1980 will amount to about 2, 000, 000, 000 Ci. These data point the need for the development of uses which can consume these large quantities of radioactive material. Certainly even the well-known and important uses in industry (thickness gages), medicine (radioiodine), and biological research (radiosulfur, phosphorus, nitrogen, etc.), use such small quantities that adequate expansion in these areas cannot be relied upon to consume even an infinitisimal fraction of either present or future outputs. For example, some of these applications involve quantities of isotopes as represented by the following:

- Snow gages–These generally employ Co^{60} as the source amounting to only 30 to 80 mCi per device.
- Neutron probes–Those which employ Ra^{226} require only about 5 mCi per device.

The products which appear to be most reasonable as useful materials are listed in Tables II, III, and IV.

TABLE II

FISSION PRODUCTS

Element or Isotope	Half-Life, yr	Isotopic Purity, %	Current Prices	Potential Uses
Kr^{85}	10.4	5	$7.50/Ci[a]	Special light source radiation source (phosphors)
Sr^{90}	28	50	$0.75/Ci[a]	Heat source, beta source
Tc^{99}	2.1×10^5	100	$90.00/g[a]	Corrosion inhibitor, alloying agent, semiconductor
Rhodium	Stable	100	$6.00/g[b]	Industrial, electrical, decorative
Ruthenium	Stable mixture +1 yr Ru^{106}	--	$1.85/g[b]	Industrial, electrical, and heat source
Palladium	Stable mixture[c]	--	$1.00/g[b]	Industrial, electrical, decorative
Xenon	Stable mixture	--	$35.00/liter (STP)[d]	Special light source
Cs^{137}	30	35	$0.50/Ci[a]	Heat and gamma sources
Ce^{144}	0.78	18[e]	$0.30/Ci[f]	Heat and beta sources
Pm^{147}	2.6	100	$0.75/Ci[g]	Heat, beta, and X-ray sources

(a) "Radioisotopes - Special Materials and Services," ORNL Catalogue and Price List. Price changes as of 2/15/62.
(b) Current prices (calculated from prices per troy ounce), Engineering & Mining Journal, February, 1965. p. 28.
(c) But includes very long-lived, very low beta energy Pd^{107}, assumed to be completely nonhazardous and nonobjectionable in all uses.
(d) Price of by-product material from liquid air manufacturing.
(e) For low exposures; for long exposures (20,000 MWd/t) and a 1 yr decay time before processing the composition is less than 3%.
(f) Estimated, based on HW-77770, data on p. 8, Reference (4).
(g) Nucleonics, April 1961, p. 70.

TABLE III

HIGHER ISOTOPES

Isotope	Half-Life, yr	Isotopic Purity	Current Prices	Uses
U^{236}	2.4×10^7	A few tenths of a percent	-----(a)	Target for Np^{237} formation in normal fuel
Np^{237}	2.2×10^6	Essentially 100%	$500/g(b)	Target for Pu^{239} formation
Am^{241}	458	May be 100% but depends on source(c)	$1500/g(b)	Heat source and target for Cm^{242} (Pu^{238})
Am^{243}	7650	May be 100% but depends on source(c)	-----	Target for Cm^{244} and other higher isotopes
Cm^{242}	0.45	May be 100% but depends on source(c)	$20,000/g(d)	Heat source and precursor of Pu^{238}
Cm^{244}	17.6	Fairly pure; may contain some Cm^{245}	$1000/g(d)	Heat source and target for other higher isotopes

(a) Not separated; stays with U^{238} or enriched uranium.
(b) Atomic Industrial Reporter, March 7, 1962, AEC prices.
(c) Pure Am^{241} may be recovered from aged plutonium (from Pu^{241} decay); however, from reactor fuel processing it will be mixed with Am^{243} and even some Am^{242} (152 yr); the composition of such mixtures depends on the exposure. At 20,000 MWd/t exposure the americium is essentially 50-50 Am^{241} and Am^{243}. Such mixtures also assure that separated curium will also consist of both Cm^{242} and Cm^{244}, but on ageing fairly pure Cm^{244} should result along with pure Pu^{238} from the aged Cm^{242}.
(d) Nucleonics, p. 63, March 1963 (projected future prices).

TABLE IV

PLUTONIUM ISOTOPES

Isotope	Half-Life, yr	Isotopic Purity	Current Prices	Uses
Pu^{238}	90	80%	$500/g[a]	Heat and alpha sources
Pu^{239}	24,000	Depends on exposure; can be very high (> 95%)	$10/g[b]	Fissionable material and source of higher isotopes
Pu^{240}	6,800	Depends on exposure; 5 to > 40%	0	Fertile material and source of higher isotopes
Pu^{241}	13	Depends on exposure; 1 to > 20%	$10/g[b]	Fissionable material and source of higher isotopes
Pu^{242}	3.8×10^5	Depends on exposure; 0.5 to near 100%	0	Target for Am^{243} formation and source of other higher isotopes

(a) From Reference (3).
(b) $10/g is the estimated fuel value of Pu^{239} and Pu^{241}. (7)

In view of the very large number of elements and isotopes formed via the fission process, the relatively small number selected for consideration in this study calls for further explanation. When one contemplates the typical fission yield curves (Figure 2), the very small yield (less than 1%) for a large fraction of all of the fission product elements is impressive. This group includes isotopes of elements such as zinc, arsenic, selenium, indium, cadmium, silver, antimony, tin, gadolinium, terbium, and dysprosium. It is reasonably clear that expectations for large-scale recovery (kilogram) of such materials is improbable. This situation coupled with the practical expectations for producing, and, more importantly, finding uses for isotopes of short half-life, even those of high fission yield, also eliminates a very large number of isotopes (such as Rb^{86}, Ba^{140}, Pr^{143}, and the like). In addition many isotopes and elements produced in fission are entirely stable and thus are essentially identical with elements plentiful in nature; included among these would be rubidium, yttrium, lanthanum, and praseodymium. It is illogical to consider these as economical products in view

of the costly processing necessary for their recovery from the complex mixture of highly radioactive materials present. These factors combine to emphasize the probable usefulness and practicality of recovering on a large scale only those few fission product elements and isotopes as listed in Table II.

The potential availability of higher isotopes is similar but even more restrictive. This is so because of the few elements at hand in fuel which can serve as target materials–simply uranium and plutonium. It is improbable that elements higher than curium will exist in sufficient concentration to justify their recovery from normal fuels. Feasible but costly methods such as further irradiation of target material such as recovered curium would, however, be a more practical means of producing those elements beyond curium. It is conceivable that power reactor operators may even find circumstances under which they could profitably charge for such irradiation services. Such possibilities are, however, beyond the scope of this present study. Table III lists the higher isotopes which are certain to be of interest and which will be present in appreciable amounts. The plutonium isotopes which would also be valuable precursors of costly americium and curium are listed in Table IV.

PRICES, VALUES, AND PRODUCTION COSTS

Although several of the products listed in the above tables have been recovered on a fairly large scale from fuel processing wastes, the concept of an integrated facility to recover each of them routinely and economically has received little attention. The problem of establishing values, costs, and prices is therefore one of guessing or preferably one of estimating on the basis of reasoning and judgment utilizing whatever information that most closely approximates the situation at hand.

No production cost was assumed for U^{236} since it accompanies massive amounts of U^{238} and would not be separated. The plutonium isotopes were assumed to bear an equal share of the rather modest processing cost which was apportioned on a dollars-per-gram basis. The processing of Sr^{90}, Cs^{137}, Ce^{144}, and Pm^{147} as presented in the Hanford Isotopes Plant Study[4] gave a fairly good basis for costs from which other fission

product production costs could be estimated, based on throughput (grams per ton of fuel) and anticipated processing complexity. The costs of the HIP products are, however, assumed to be for bulk products, not yet encapsulated. It is assumed that the production cost (including profit) for many of these would be 90% of the future bulk price; with 10% of this figure then being the amount creditable to the value of the unprocessed fuel. The latter ratio can be supported by a review of several common industrial chemical processing situations; for example, market-price*-to-raw concentrate costs show the following ratios:

	Ratio
Barite to precipitate $BaCO_3$	7.85
Beryllium ore to BeO	8
Chromite to CrO_3	10.75
Fluorspar to HF (aq.)	9.6
Phosphate rock to ammonium phosphate (feed grade)	9.8
Monazite to rare earth oxides (CeO_2)	10
Zircon to ZrO_2 (optical grade)	9.9

For those intermediates or precursors in which the only investment comprises the value of neutrons absorbed, such neutrons are charged in at $2500/g, which is regarded as a conservatively low incremental cost of a "typical" gram of neutrons. This procedure was modified in the case of Pu^{240}, since in this case a portion of the invested neutrons go to produce an isotope which is largely consumed in fission (Pu^{241}) instead of progressing "up the scale" to Pu^{242}.

For isotopes like Kr^{85} the establishment of a basis for a future price is very uncertain. Kr^{85} at one time was expected to be marketable at $15.00/Ci. The intended uses which supported such expectations have obviously not developed even with the present price of $7.50/Ci, which has prevailed for 2 1/2 yr. The future price, for the purposes of this study, was then assumed to be $0.75/Ci (or about $300/g) which is 10% of the present price. It was thought that such a drop in price may encourage sufficient uses under future conditions.

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*From current market quotation in "Oil Paint and Drug Reports," and "Engineering and Mining Journal."

For the noble metals, essentially present prices were assumed for the future.

In view of the very high prices for xenon today, a much lower price was also assumed for the future. The very large availability of the fission product source and potentially convenient and rich concentrations in the off-gases from fuel processing supports expectations for relatively low production costs.

For the higher isotopes which are one step away from the valuable ultimate products (Am^{243}, Cm^{244}, and Am^{241} for Cm^{242} and Pu^{238}) the value of the ultimate products and a reasonable processing cost was taken into consideration in arriving at the value of the intermediates.

COMMENTS, COSTS, AND PRICING FOR EACH PRODUCT

THE HEAT SOURCE FISSION PRODUCTS

The fission products for which the major use relates to their application as heat or power sources are Sr^{90}, Cs^{137}, Ce^{144}, and Pm^{147}. Each of these was discussed in Reference (4), the "HIP Report." The future prices for bulk products are assumed to be essentially the total cost as reported for encapsulated products in that reference including feed cost. Although no feed cost is expected,* it is retained and assumed to cover instead amortization, taxes, and profit. The assumed future production cost is then estimated to be about 90% of the future prices as mentioned above. Because of the greater potential value of promethium, in view of its low shielding requirements which make it competitive with such costly isotopes as Pu^{238}, a somewhat greater spread between future price and production cost has been assumed.

Power reactor fuels will tend toward very long exposures. Furthermore, there will probably be no great incentive to assure a short cooling time between reactor discharge and chemical processing. These two factors introduce variables which greatly influence the quality of certain by-product

*It is even conceivable that fission product recovery might receive credit for waste management expenses which would be avoided through sale of of these materials.

elements and isotopes. In some cases higher yields will result from prolonged ageing; in others lower yields and lower concentrations will be obtained. Ce^{144} is a prominent example of the latter situation. Its rather short half-life leads to saturation concentrations at fairly low exposures. In addition, the concentration of the stable isotopes of cerium continue to increase throughout the exposure period, leading to lower and lower isotopic concentrations of Ce^{144}. Finally, at the conclusion of the exposure period and during the ageing, cooling, or storage period between discharge and chemical processing, the Ce^{144} decreases via beta decay leading to a further major reduction in isotopic concentration. Thus, to achieve highest Ce^{144} yields and isotopic concentrations, the fuel exposure should not exceed the minimum level at which saturation is achieved, and the ageing period should be as short as possible.

Neither of these conditions is compatible with power reactor fuel cycle economics. Although Ce^{144} is an outstanding example of this problem, Ru^{106} presents very much the same situation. For Ru^{106}, however, a significant advantage exists because of the very much higher Ru^{106} fission yields obtainable from situations which emphasize the burning of plutonium. In addition, the exposure at which saturation occurs is much higher than for Ce^{144}. Also the longer half-life slightly favors the buildup of Ru^{106}. However, the buildup of inert ruthenium isotopes is also very significant. From data of Reference (5), at 20,000 MWd/t for low enrichment, U^{235} fuels, it can be shown that at 1 yr after reactor discharge the cerium will contain somewhat less than 3% Ce^{144} and the ruthenium will contain slightly more than 3% Ru^{106}.

OTHER FISSION PRODUCTS

Krypton-85

Although on an annual basis, kilogram quantities of Kr^{85} should be available, the amount in each ton of fuel is the smallest of any of the fission products. The unique application (phosphors) which originally was expected to support a high price may still be contemplated and may be significantly encourage by a much lower price. Even at prices one-tenth that of current prices, and with rather substantial processing costs, a sufficient margin

remains to assure a significant value in unprocessed fuel. The total krypton will be about 258 g/ton for 25,000 MWd/t fuel. This would be extracted along with the massive amounts of xenon, a situation of mutual benefit in sharing the recovery costs.

Xenon

Xenon will be discharged as a mixture of the stable isotopes. The potential contaminating radioactive xenon isotopes have such short half-lives that by the time fuel is processed they should be essentially absent. The availability of xenon in grams-per-ton of fuel is far higher than that for any of the other potentially useful fission products--3987 g/ton of 25,000 MWd/t fuel. This, plus the feasibility of a fuel process which could release xenon along with krypton in fairly high concentrations encourages the consideration of a recovery cost and selling price below that existing today for the liquid air by-product.

Technetium-99

Technetium, an element not found in nature, is produced via fission in very high yield, 50% greater than for Sr^{90} from low enrichment power reactor fuels. Its isotopes are generally of very short or very long half-lives. The long lived ones are adjacent. The principal isotope produced in fission is Tc^{99} with a half-life of 210,000 yr. The Tc^{98} which has an even longer half-life (1.5×10^6 yr) is not produced in yield high enough to be of interest. The other fission-produced technetium isotopes have half-lifes measured in minutes and less. Tc^{99} would therefore be obtained in high isotopic purity. Its recovery by either solvent extraction or ion exchange is simple and has been demonstrated on a kilogram scale. The ion exchange process used and the yields are so similar to those for Cs^{137} that the costs have been assumed to be about the same per gram.

Uses for technetium have not been fully developed. Its existence as a new element, the trend toward lower costs, and some of its properties, particularly in the areas of metallurgy and its property of imparting corrosion resistance to steel exposed to aqueous solutions containing small amounts of technetium, indicate future uses. Its use as an alloying agent

to improve the metallurgical properties of tungsten is also being studied, as are superconductivity characteristics. Its long half-life and low energy pure beta emissions (free of gamma activity) indicate that it may be used with a minimum of control.

The Noble Metals

As a group the similar noble metals, ruthenium, rhodium, and palladium, have the next the highest availability (after xenon), among all potentially useful fission products and as a group might be easily recovered. This possibility would appear to be greatly enhanced in an integrated fuel processing and multiple isotope recovery facility. The cost per gram may then be sufficiently low to make recovery economical in competition with conventional ore sources. The recovery cost, therefore, is assumed to be only slightly below the expected selling prices.

In precious metal mining today the existence of accessible and large hard-rock ore bodies containing about one-half of an ounce of gold ($17) to the ton justifies mining and processing. For large placer deposits, gold values at far less than 1 g ($1) per ton justify mining. In expected power reactor fuels, the rhodium value alone based on the present prices will be about $2000/ton. Palladium values would add about another $1000 per ton of fuel. Compared with the concentrations in natural ores, such levels of the noble metals in spent fuels would be regarded by miners and economic geologists as nothing less than fantastic. The acknowledged existence of such concentrations in extensive and available geological formations would certainly initiate a "gold rush" of wild intensity! However, the reality of the high cost of processing intensely radioactive materials plus the rather modest quantities of such material which can be anticipated in the near future (within the next 5 to 10 yr) requires a temperate and more deliberate approach to such recovery. Although such recovery apparently has not been demonstrated on a significant scale even experimentally, the possibilities and eventual practicality of a large scale approach seems reasonable as well as probable.

Rhodium-103

Stable fission product Rh^{103} is the daughter of 40 day Ru^{103}. For normal fuels of U^{235} or Pu^{239} its yield is about 2.9 or 5.8%, respectively (Figure 2). The fairly long lived Rh^{102} may be present in very small amounts as a result of (n, 2n) reactions on Rh^{103}. Fuels of very long exposure may thus have sufficient Rh^{102} to impair the uncontrolled utility of the stable Rh^{103}. In contrast to this possibility the cross section for this particular (n, 2n) reaction may be so low, or the absorption cross section for the resulting Rh^{102} so high that no problem with Rh^{102} will arise. Until this unknown is resolved by precise analyses of samples of fuels exposed for long burnup, no problem is assumed in the use of rhodium recovered as a fission product.* For fuels in which Pu^{239} comprises a large fraction of the fissions, the yield of rhodium will be highest. For conventional fuels at 25,000 MWd/t exposures, the yield is about 337 g/ton (about 80% of that for Sr^{90}). Rhodium from conventional mineral sources today sells for about $6.00/g.** In the rapidly expanding nuclear power industry rhodium will, therefore, become increasingly significant as a potentially recoverable and valued by-product. By about 1980 the magnitude of fission product rhodium will exceed the quantity marketed in the U. S. in 1962 from conventional mineral sources. In other words, unless this fission product is recovered, more rhodium will be returned to the earth in the form of reactor fuel processing wastes than was recovered from minerals mined from the earth in 1962!

Little is known of the exact state and fate of rhodium in the nuclear fuel separation processes. A modest analytical effort would be necessary to obtain such information. The technology of recovering gross quantities of rhodium economically in high yield and purity from conventional fuel processing wastes has also yet to be developed. However, some special processes have been published in the form of patents for such recovery of rhodium.[10] One of these patents relates to the separation of Ru^{103} which

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*These analyses are in process at the present time, but results are not yet available.

**It is of interest to note that this value comes close to that for plutonium from high exposure power reactor fuels (with the plutonium valued only at its fuel value at $10/g for the Pu^{239} and Pu^{241}). Such plutonium can be about 70% Pu^{239} plus Pu^{241}.

is aged and then processed for pure Rh^{103} recovery; in the other patent the nitric acid waste solutions are converted to chloride solutions from which the rhodium is recovered via sulfide precipitation.

About 26,000 troy ounces of rhodium were marketed in the U. S. in 1962. Its principal uses were in the chemical, glass, electrical, and jewelry (and decorative) industries. As an alloy with platinum it has very useful and distinctive properties in applications involving very high temperatures in air: thermocouples, heating elements, crucibles, etc.*

Palladium

In nature palladium is the most abundant of the noble metals, being about twenty times that of rhodium but less than twice that of platinum. Its selling price today is about $1.00/g. It has widespread use in chemical (catalyst), electrical, glass, dental, and jewelry (decorative) applications. In fission the combined yield of the four principal isotopes is over twice that of either rhodium or Sr^{90}. As with rhodium, the highest yields are obtained via plutonium fission. Its price is, however, borderline as an element of significant value in spent nuclear fuels. If its recovery cost were low (a possibility with an integrated recovery process for all of the noble metals), it would possibly be worth recovering. Among its isotopes there appear to be none which could impair the possibility of uncontrolled use. Radioactive Pd^{107}, which comprises a large fraction of the isotopes (about 20%), has a long half-life (7,000,000 yr) with no gamma emissions and with a beta emission of such low energy (0.035 MeV) that it may be regarded as totally without hazard for any of the major uses, even including dental.

As in the case of rhodium, little is known about palladium in the fuel recovery process. Also as with rhodium analysis of typical high exposure power reactor fuel samples is in progress to establish the concentration of palladium. Similarly, the technology needs development before economical recovery can be assured. The future magnitude of the nuclear fuel processing industry could provide amounts of palladium comprising a significant fraction of today's needs.

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*The "Minerals Yearbook" for 1937, p. 752, makes the following statement: "Because of its brilliance and durability, rhodium has been called "the diamond of the metals." As rhodium is one of the whitest and hardest of all metals and never tarnishes, rhodium plating is being used more and more---."

Ruthenium

Unfortunately, ruthenium has an active isotope, Ru^{106}, of fairly long half-life (1 yr). The realization of conventional uses for such ruthenium is therefore questionable. Ruthenium is the most abundant solid element of potential value in spent fuel. Ruthenium from conventional mineral sources today sells for about $1.85/g. It is assumed to be readily recoverable from spent fuel and to eventually have some uses. If it could be recovered at the indicated costs, it may have potential as a powerful (high power density) radioisotopic heat or power source. As such, ruthenium may be worth far more than as a noble metal: possibly 50 to 100 times as much. The extremely energetic beta decay energies of its daughter, Rh^{106}, indicates potential applications. The fission yield of Ru^{106} is, however, quite low from U^{235} (0.38), but from Pu^{239} fission is over a factor of thirteen greater.

Xenon and the noble metals rank among the most expensive of the conventional elements. However, their prices are low when compared with the possible costs of processed radioactive materials. Thus, to be economical for recovery, the processing cost must also be low. Since on a weight basis xenon and the noble metals comprise by far the greatest of the potentially useful products, fairly low recovery costs have been assumed; however, it will be noted on inspection of Table I that the processing cost is only slightly below the assumed future value of these materials. The credit which can be expected is thus also modest.

THE HIGHER ISOTOPES

The higher isotopes are formed by neutron absorption in the basic fuel materials, uranium and plutonium. Most are mixtures of isotopes not considered to be separable. They include U^{236}; Np^{237}; two americium isotopes, Am^{241} and Am^{243}; two curium isotopes, Cm^{242} and Cm^{244}; and five plutonium isotopes, Pu^{238}, Pu^{239}, Pu^{240}, Pu^{241}, and Pu^{242}. For the purpose of this review the plutonium isotopes are discussed as a group and are covered in detail in a following section. The special situation between the competing neutron reactions of absorption and fission, particularly for Pu^{241}, indicates that special consideration be given to the individual discussion of these materials.

Uranium-236

U^{236} is formed by neutron absorption in U^{235}. About 15% (one-sixth) of the neutrons taken up by U^{235} form U^{236}; the balance result in fission. The U^{236} thus formed remains with the processed uranium. As with essentially all such neutron reactors the actual cross sections depend greatly on neutron energy and reactor conditions and to accept the 15% absorption figure is for illustration only.

U^{236} is a precursor of Np^{237}, the latter element being the target material used in the production of valuable and costly Pu^{238}. U^{236} is therefore valuable in this production chain. Since it is not fissionable by thermal neutrons, essentially all of it is useful for Np^{237} formation. Since its content in recovered uranium is so low (about 0.2%) it is assumed not to share in the uranium processing cost–"It just goes along for the ride." Realistically though, its share of any processing cost, figured on a dollar-per-gram basis, would be so small (1.5 ¢) compared with its value that it can be neglected. However, since one neutron is invested in its formation it is assumed to carry an appropriate value, this being the incremental cost of this neutron. With incremental neutrons valued at \$2500/g and each gram of neutrons thus distributed over 236 g of U^{236}, the value of the U^{236} in the unprocessed fuels is thus set at \$10.60/g (\$2500 ÷ 236 = \$10.60). This value, rounded to \$11, is assumed then to be creditable to the reactor operator for the U^{236} which he has inadvertently produced.

Neptunium-237

Np^{237} originates from two sources: neutron absorption in U^{236} and the (n, 2n) reaction on U^{238}. For power reactor fuels of higher exposure the contribution from the (n, 2n) reaction is the smaller. Np^{237} is useful primarily as the target material for the production of valuable Pu^{238}. The value of this precursor of Pu^{238} should therefore reflect not only the investment in neutrons required but also the value of the final product, Pu^{238}; the conversion efficiency; and the cost of the chemical processing required to recover the Np^{237}. The present price of Np^{237} in limited quantities (grams) is \$500/g (Table III).

Neptunium is an alpha emitter and would be most conveniently handled as such, with a minimum of shielding. The purification is therefore important. In view of the relatively small quantities recoverable from fuels it would be expected to be more costly than the gamma emitters as well as the plentiful alpha emitters such as plutonium which shares its costs with the uranium fuel processing costs. A figure of $80/g was regarded as appropriate for the future production cost of Np^{237}.

Based on the projected price for Pu^{238} of $500/g; the rather poor yield in going from Np^{237} to Pu^{238} (50 to 60%); and the special target preparation, irradiation, and processing for Pu^{238} and recovery of unconverted Np^{237}, a figure of $100/g was considered as suitable for the future price of Np^{237}. Allowing a rather high figure of $80/g as the production cost, this leaves $20/g or its value in the unprocessed fuel; this is essentially equal to the value of the neutrons required for its formation.

Americium

The two principal americium isotopes, Am^{241} and Am^{243}, are derived exclusively from successive neutron absorption in Pu^{239}. Am^{241} is subsequently formed by the beta decay of 13 yr Pu^{241}. Am^{243} comes from neutron absorption in Pu^{242}. Both americium isotopes are long lived alpha emitters. Am^{241} has a sufficiently short life (458 yr) that it could be used as a heat source, if pure enough and cheap enough. Conceivably both purity and low cost could be obtained by simplest reprocessing of pure aged plutonium. The americium isotopic composition obtained from direct processing of power reactor fuel depends on age and exposure. At 20,000 MWd/t, the isotopic ratio is essentially 50-50. It is worth noting that at such exposures the total yield of americium is substantially greater than that for promethium and other more familiar fission products such as Ce^{144} and Kr^{85} (Table I).

The irradiation of the target americium mixture would produce both Cm^{242} and Cm^{244}. On ageing of this mixture of curium isotopes for a few years, essentially all of the Cm^{242} would be converted to Pu^{238}. Thus, Am^{241} like Np^{237} can be considered as a source of Pu^{238}. There may be

definite advantages in such a route in yield and purity of the Pu^{238} thus produced. Am^{243}, on the other hand, is useful only as the immediate precursor of valuable and costly Cm^{244}.

For use as a competitive heat source, Am^{241} would have to have a value of about one-fifth of that for Pu^{238}, or \$100/g. Its recovery and purity as a by-product of operation of the chromatographic ion exchange system for promethium manufacture should provide americium of required purity at low cost. The direct accumulation in the promethium system from which it must be removed thus obviously indicates recovery with little effort. This encourages consideration of a production cost figure lower than Np^{237}. If it is to share the cost of operating the ion exchange system it should not be cheaper than the per gram cost of promethium. Therefore, \$50/g seems reasonable for the production cost of Am^{241}. Since Am^{243} would be recovered concurrently with Am^{241}, its production cost is also assumed to be \$50/g.

Curium

The two principal curium isotopes to be found in fuel are Cm^{242} and Cm^{244}. Because of the relatively short half-life of Cm^{242} (163 days), the ageing time before processing is important in establishing the ratio of these two isotopes. Both are derived from neutron captures in the two americium isotopes. As mentioned above, Cm^{242} could be recovered as a step in the production of Pu^{238}.

For the Cm^{244} to be useful as a heat source it would have to be aged sufficiently long so that the Cm^{242} content would be so low that the heat output was predominantly from long-lived Cm^{244}. Otherwise, the heat source would show the rapid decline in heat output characteristic of Cm^{242}. The ageing time required would be dependent on the concentration of the Cm^{242}; however, for anticipated concentrations, a period of 2 to 4 yr would be needed to bring the Cm^{242} content to less than 0.25% so that initially less than about 10% of the total heat would be coming from the Cm^{242}.

Such an ageing step would, of course, involve some loss of Cm^{244} (probably less than 25%). However, such losses are much smaller than those resulting from the required ageing of heat sources such as promethium.

The concentrations of curium and the relatively high-fission-yield by-product, promethium, when compared on a weight basis, shows the curium concentration to be about one-sixth that of promethium. However, the significance of the Cm^{244} concentration is more impressive when the comparison is made on the basis of potential availability of the emitted energy: about 2.8 W/g for Cm^{244} and only 0.33 W/g for Pm^{147}. Furthermore, the Pm^{147} would have to be aged for about one half-life to remove the offensive (gamma emitting) Pm^{148}. Therefore, effectively, only one-half of the concentration of Pm^{147} in the fuel can be considered useful as a heat source. This situation coupled with the inherent low specific power gives a completely different and more favorable picture of the comparative significance of the Cm^{244} concentration. From this standpoint then, Cm^{244} will yield twice as much useful heat as Pm^{147}:

Cm^{244} = 22 g/ton of fuel.
At 75% recovery at 2.8 W/g = 46.2 W/ton of fuel.

Pm^{147} = 133 g/ton of fuel.
At 50% recovery at 0.33 W/g = 22.2 W/ton of fuel.

A similar comparison for Pu^{238} based on its production from the neptunium recovered from the same fuel (Table VI) shows the following:

316 g Np^{237} per ton of fuel; 90% recovery; 50% conversion efficiency to mixed plutonium isotopes containing 80% Pu^{238}; a specific power of 0.56 W/g of Pu^{238}:

$$(316)(0.50)(0.9)(0.56) = 79.5 \text{ W/ton of fuel.}$$

Thus, the heat available from Pu^{238} which is produced by a much more circuitous and time consuming route is less than double that obtainable directly from by-product Cm^{244}. Furthermore, this route for Pu^{238} takes time since the neptunium must be irradiated in cycles which only convert a fraction of the neptunium to Pu^{238}. Then the mixture must be separated, purified, and the neptunium refabricated into target elements for the next cycle of irradiation.

The future price or value of Cm^{242} has been placed at $300/g. This is far below present projections (Table III), and has been set at this level because of the anticipated low cost of the target, Am^{241}, plus the fact that it may be used to produce Pu^{238} and thus would have to be valued at appreciably less than Pu^{238}. Although curium should also be recovered from the ion exchange system like americium, the smaller quantities involved and the probable requirement for special procedures to cope with the intense and damaging alpha activity justifies a substantially higher production cost for the Cm^{242}. The credit for Cm^{242} in fuel should also be based on its use as a source of Pu^{238}. A cost figure of $150/g was thus concluded as reasonable for Cm^{242}. Fairly pure Cm^{242} for heat source uses would have to be specially produced via Am^{241} irradiation.

The price for Cm^{244} is that from Reference (3). The production cost was concluded to be between that of americium and Cm^{242} and was thus set at $100/g. As for Am^{243} the credit is potentially so large as to encourage plutonium fueling, optimization for neutron capture instead of fission and the offering of premium prices for by-product, high exposure plutonium with values based on Pu^{240} and Pu^{242} content in addition to the fuel values of Pu^{239} and Pu^{241}.

THE PLUTONIUM ISOTOPES

Essentially all considerations of plutonium value have been concerned with its use as a nuclear fuel. Its possible value as the only source material for manufacture of valuable higher isotopes has been specifically avoided, subordinated, and/or discounted.[11] In view of the efforts to produce significant quantities of Cm^{244} over the next few years and beyond, it seems unreasonable to continue to avoid consideration of credits to the value of plutonium for such production. This production will be realized with hardly more than an unintentional investment in neutrons resulting from the normal utilization of plutonium as a fuel in thermal reactors. Through processing by conventional means, very significant quantities of by-product curium (and other valuable higher isotopes) will then be recovered at costs potentially far below those now being experienced in the special production programs.

The conventional studies, which do not take into account the value of the higher isotopes, result in conservatively low values for plutonium. A similar but somewhat less significant situation exists for recycled U^{235} fuels, if a value is placed on U^{236} in view of its role as the precursor of Np^{237} and eventually extremely costly (valuable) Pu^{238}.

In the process to produce plutonium via the irradiation of U^{238} all of the five long-lived plutonium isotopes are produced. In irradiations of short duration, formation of Pu^{239} is dominant. However, in power reactor operation which has as an objective the conversion of as much low cost U^{238} into plutonium as possible to gain heat from the fission of such plutonium, a substantial part of the total plutonium exists as isotopes above Pu^{239}, only one of which is significantly fissionable in the thermal spectrum (Pu^{241}). Heretofore, by-product plutonium has been valued only as fissionable material relative to the equivalent value of U^{235}. This figure has been set at \$10/g of Pu^{239} plus Pu^{241} (Table IV). No credit has been acknowledged for any of the other plutonium isotopes. Cm^{244} can be obtained only by irradiation through the whole chain of plutonium isotopes, and has great value. Therefore, the precursors of Cm^{244}, all of the plutonium isotopes, certainly have some value. In this study, the value of the neutrons absorbed which are useful for the production of higher isotopes has been accepted as a reasonable basis for establishing the minimum value of these plutonium isotopes. The production cost of plutonium is merely the share of the fuel processing cost (Assumption 9, p. 4) born by each gram of plutonium in a ton of 25,000 MWd/t fuel. This figure amounts to very close to \$2/g.

The consideration of plutonium value for higher isotope formation based on the incremental value of invested neutrons is complicated by the fact that in the formation of the successive plutonium isotopes two of them have substantial fission cross sections, thus they are less valuable as higher isotope precursors. In the following discussion this factor is taken into account when arriving at plutonium isotope value.

Plutonium-238

Pu^{238}, although it exists in fuel, has no value because it cannot be separated. It can, however, be recovered from the curium fraction via

the ageing process required to obtain to obtain reasonably pure Cm^{244}. Cm^{242} credit is therefore shown only as credit for the Pu^{238} provided via this route. The processing cost for Pu^{238}, is then the value of the Cm^{242} plus the cost of separating the Pu^{238}. The quantity of Pu^{238} available via this route is also minor. Processing of fuel with a minimum ageing period, and thus recovering curium before so much of it has decayed to Pu^{238} which would otherwise be "lost" in the usual mixture of plutonium isotopes, would be a means of possibly doubling the amount of Pu^{238} that could be eventually recovered in fairly pure form. Such Pu^{238} would always be contaminated with appreciable Pu^{240} derived from the decay of Cm^{244}.

Plutonium-239

Pu^{239} at the modest value as a fissionable material ($10/g) is still a most valuable material in unprocessed reactor fuel. Although it is intended for use as a power producer, additional credit may also be taken for it as a precursor of the higher isotopes. Since 26.7% of the Pu^{239} does not fission on capturing a neutron, 26.7% of the value of the neutrons that went into the initial formation of Pu^{239} may be taken as credit for higher isotope formation. However, at the Pu^{241} position another large fraction of neutron captures result in fission instead of a new isotope. Therefore, the value of Pu^{239} for higher isotope production is further reduced by a factor of 28.5%. Thus, neutron credit must take these value deficiencies into account. With this situation emphasized, the credit value of Pu^{239} for higher isotope production and for fission is estimated as follows:

$$\text{Credit} = \frac{\$ \text{ value of 1 g-atom of neutrons} \times 0.267 \times 0.285}{239} + \$10^{*}$$

$$= \frac{\$2500 \times 0.267 \times 0.285}{239} + 10 = \$10.80/\text{g (call it } \$11/\text{g)}.$$

The 26.7% figure is the ratio of absorption cross-section to total cross-section for Pu^{239}. The 28.5% figure is the ratio of absorption cross-section to total cross-section for Pu^{241}. These ratios are not fixed but can be varied and thus may be optimized for maximum higher isotope production.

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*Accepted value as fissionable material.

Plutonium-240

Pu^{240} is nonfissionable in the thermal neutron spectrum and therefore to date has not had a value set on it as have both Pu^{239} and Pu^{241}. It may be regarded, however, as valuable as a fertile material required for the formation of fissionable Pu^{241}. If value of fertile material were proportional to the cross-section, Pu^{240} would be worth about one hundred times that of U^{238}.* With depleted U^{238} valued at a high figure of \$4.50/lb in the form of the oxide (=\$0.01/g), Pu^{240} may be worth \$1.00/g. This is, however, regarded as sufficiently significant in this study to justify inclusion as a modest credit.

As in the case of Pu^{239}, the neutron credit in Pu^{240} is depreciated by the fact that the successor, Pu^{241}, is fissionable. Only 28.5% of the neutron absorption result in formation of the next higher isotope. Therefore the neutrons absorbed to form Pu^{240} from U^{238} are only 28.5% effective in attaining higher isotopes. The credit for neutron investment in Pu^{240} should thus be:

$$\text{Credit} = \frac{\$2500 \times 2 \times 0.285}{240} + \$1 = \$6.94 \text{ (call it } \$7/g).$$

Although the neutron credit is much greater than for Pu^{239}, the total value is less because of the high fission value credit to Pu^{239}.

Plutonium-241

Pu^{241} is the other readily and thermally fissionable isotope of plutonium formed by neutron capture in Pu^{240}. It is also valued at \$10/g in mixture with the other plutonium isotopes for use as a thermal reactor fuel. Its cross section for capture and fission in the thermal range is about like Pu^{239}, 28.5% of the absorptions result in capture to form Pu^{242}; the balance results in fission. Thus, for higher isotope production only 28.5% of the neutrons absorbed to form it can be credited for higher isotope manufacture. The credit for neutron investment in Pu^{241} and for fission value should thus be:

$$\text{Credit} = \frac{\$2500 \times 3 \times 0.285}{241} + \$10 = \$18.86/g \text{ (call it } \$19/g).$$

---

*This is also an oversimplification since the cross sections are highly variable and dependent on specific reactor conditions.

Pu^{241} has a half-life of 13 yr. Its decay is predominantly via beta emission to produce Am^{241}. Plutonium which originated from fuels of prolonged exposure will be rich in Pu^{241}. On storage this Pu^{241} converts to Am^{241} which can be recovered in fairly simple solvent extraction or ion exchange processes. The pure Am^{241} thus recovered can be used as a heat source or can be irradiated to produce Cm^{242}, an isotope of extremely high specific power (120 W/g). As discussed earlier, Cm^{242} on decay converts to Pu^{238}: another isotope of considerable interest as a heat or power source.

Plutonium-242

Pu^{242} formed by neutron capture in Pu^{241} is essentially nonfissionable in the thermal neutron spectrum, has a very long half-life (379,000 yr) and only a modest absorption cross section (23 barns). In uranium fuels of prolonged exposure or preferably in highly burned plutonium fuels the Pu^{242} content can reach very high concentrations–in some situations can approach well over 95%. Such compositions would be ideal as target material for higher isotope manufacture, Am^{243}, Cm^{244} and beyond. Since it is non-fissionable its only apparent use is for such production, and should also receive full credit for neutron investment all the way from U^{238}. Its credit on this basis is then:

$$\text{Credit} = \frac{\$2500 \times 4}{242} = \$41.33/\text{g (call it \$41/g)} \quad .$$

Other Plutonium Isotopes

The "Chart of the Nuclides" shows ten other plutonium isotopes. With the exception of Pu^{236} and Pu^{244}, they are generally all of short half-lives–minutes, hours, and days–and are thus essentially without interest as practical recovery possibilities. Pu^{236}, a member of a decay chain dominated by U^{232} and Th^{228}, would be of considerable interest as a heat source if it could be produced cheaply. A practical production process is not apparent. Pu^{244} has by far the longest half-life of any plutonium isotope (76,000,000 yr). Unfortunately, between it and Pu^{242} is Pu^{243}, an isotope with a half-life of only 5 hr. However, intense irradiation of highly concentrated Pu^{242} in a very high flux may be a means of producing Pu^{244} in practical quantities and in high concentrations.

If quantities of Pu^{244} of fairly high isotopic purity could thus be obtained, the possibility then exists for the production of appreciable amounts of fairly pure Cm^{245}. This curium isotope may be of interest as a fissionable material with a critical mass much smaller than that for Pu^{239}. It may thus be particularly advantageous for use in very small compact power reactors. The route to this product via Pu^{244} is the only apparent one, other than via isotopic separation of mixed curium isotopes.

Table V summarizes plutonium isotope values as determined above.

TABLE V

SUMMARY OF VALUE OF PLUTONIUM ISOTOPES

	Use	Estimated Value,[a] $/g
Pu^{238}	Heat source	500
Pu^{239}	Nuclear fuel; higher isotopes	11.00
Pu^{240}	Fertile material; higher isotopes	7.00
Pu^{241}	Nuclear fuel; higher isotopes (and Am^{241})	19.00
Pu^{242}	Higher isotopes	41.00

(a) Based on credit for neutrons which are effective in producing higher isotopes. This comment does not apply to the estimated value of Pu^{238}.

The calculations shown for the estimation of these values for plutonium isotopes assume "Nuclide Chart" values for cross sections, etc. It should be realized that in a given reactor situation, wide variations in cross section can be obtained. With significant values being assigned to plutonium isotopes, conditions may be optimized for maximum production of certain isotopes. The credit values thus obtained could differ greatly from those shown above.

SUMMARY OF DATA ON ESTIMATED VALUES

The economic data developed in the preceding discussions are summarized in Table VI. In discussion of fission product values questions

TABLE VI

ECONOMIC DATA FOR ISOTOPES AND ELEMENTS

Isotope or Element	Future Prices, $/g	Estimated Production Cost, $/g	Net Value in Fuel, $/g	Approximate g/ton 25,000 MWd/t	Estimated Value, $/ton Spent Fuel	
Kr^{85}	300	200	100	17	1 700	Fission Products $7 660
Sr^{90}	20	18	2	411	822	
Tc^{99}	10	9	1	628	628	
Rhodium	6	4	2	337	674	
Ruthenium	2	1.50	0.5	1 707	854	
Palladium	1.25	1	0.25	976	244	
Xenon	2	1.70	0.30	3 907	1 196	
Cs^{137}	10	9	1	950	950	
Ce^{144}	25	22.50	2.50	241	260(a)	
Pm^{147}	30	25	5	133	335(b)	
U^{236}	---	0	11	3.277	36 070	The Higher Isotopes $104 110
U^{238}	6.75/lb	6.75/lb	0	8.9×10^5	0	
Np^{237}	100	80	20	316	6 320	
Am^{241}	100	50	50	56	2 800	
Am^{243}	500	50	450	23	3 450(c)	
Cm^{242}	300	150	150	87	39 120	
Cm^{244}	1,000	100	900	22	19 800	
Pu^{238}	500	350	150	--	3 450	Plutonium Isotopes $99 185
Pu^{239}	11	2	9	5 453	49 100	
Pu^{240}	7	2	5	1 517	7 585	
Pu^{241}	19	2	17	1 412	24 000	
Pu^{242}	41	2	39	386	15 050	
			Value: per short ton		$210 955	
			per pound		105.50	
			per kilogram		232.50	
			per kWH_e		1.055 mills(d)	

(a) At 1 yr after discharge.
(b) At 1 yr half-life.
(c) As Pu^{238}, value included with plutonium isotopes.
(d) At 33 1/3% efficiency thermal to electrical

arise relating to the comparative prices of some of the common but costly commercial elements. Table VII lists current prices of many such materials.

TABLE VII

CURRENT PRICES[a] OF SOME COSTLY COMMERCIAL ELEMENTS[b]

Element	Cost	Element	Cost
Beryllium	$54-66/lb	Palladium	$32-34/troy oz
Boron	$25/lb	Platinum	$98/troy oz
Cesium	$100-150/lb	Rhenium	$600/lb
Gallium	$1.35/g	Rhodium	$182-185/troy oz
Germanium	$0.28/g	Rubidium	$100/lb
Gold	$35/troy oz[c]	Ruthenium	$55-60/troy oz
Hafnium	$75/lb	Scandium	$3/g (minimum)
Indium	$2.40/troy oz	Selenium	$4.50/lb
Iridium	$90-95/troy oz	Silver	$1.29/troy oz
Lithium	$9-11/lb	Tantalum	$30-49/lb
Niobium	$36/lb	Tellurium	$6/lb
Osmium	$150-160/lb	Thallium	$7.50/lb
		Zirconium	$4.25-5.00/lb

(a) From Handbook of Chemistry and Physics, 45th Ed., 1964-1965, and Engineering and Mining Journal, February 1965.

(b) This list excludes the rare gases, rare earths, and the radioactive elements.

(c) A troy oz is equivalent to 31.10 g.

PLUTONIUM RECYCLE AND HIGHER ISOTOPE FORMATION

Although consistent data on the effects of plutonium recycle on the formation of all isotopes of interest are not yet at hand, information on production of the higher isotopes is reported in Reference (6). In view of the large number of possible fueling combinations and reactor conditions that can be conceived, only three representative situations are summarized in

Table VIII. The data in the table emphasize the results obtained at widely differing plutonium-isotopic compositions. It is certain that in view of the yield curves shown in Figure 2, major changes in concentrations of the fission products with masses in the region of the low mass peak would be realized. The estimated production effects under conditions of plutonium recycle in natural uranium for these fission products would be:

Kr^{85}	-- reduced by about one-half	(to 8.4 g/ton)[a]
Sr^{90}	-- reduced by about one-half	(to 239 g/ton)[a]
Tc^{99}	-- essentially unchanged	(to 630 g/ton)[a]
Rhodium	-- increased 50%	(to 478 g/ton)[a]
Ruthenium	-- increased 15%	(to 2029 g/ton)[a]
Palladium	-- almost doubled	(to 1660 g/ton)[a]

TABLE VIII

PLUTONIUM RECYCLE AND HIGHER ISOTOPE PRODUCTION
(g/ton at 25,000 MWd/t)

Isotope	Low Enrichment Uranium (as in Table VI)	Plutonium[a] in Natural Uranium	Plutonium[b] in Natural Uranium
U^{236}	3277	403	326
Np^{237}	316	46	33
Am^{241}	56	175	793
Am^{243}	87	392	1300
Cm^{242}	23	89	273
Cm^{244}	22	126	351
Pu^{238}	18	175	647
Pu^{239}	5453	6581	6620
Pu^{240}	1517	4820	16462
Pu^{241}	1412	3282	8813
Pu^{242}	386	1569	8676
Total Plutonium	8786	16427	41218

(a) Plutonium composition in feed = 76%, Pu^{239}; 18% Pu^{240}; 5% Pu^{241}; 1% Pu^{242} [Reference (6), Case 3002]

(b) Plutonium composition in feed = 22%, Pu^{239}; 46% Pu^{240}; 22%, Pu^{241}; 10%, Pu^{242} [Reference (6), Case 5000]

In situations where the plutonium fuel is high in Pu^{241} or in reactor conditions which favor the formation and fission of Pu^{241}, the effects as indicated above would be further emphasized by the expected corresponding shift in the low mass peak.

PRODUCTION OF OTHER ISOTOPES

Although this study has been concerned with an estimation of the value of fission products and isotopes produced in the course of normal power reactor operation, the facilities required for the processing and recovery of these by-products would very likely be directly applicable to the processing of essentially any other isotope which may be produced in reactors. An isotope plant may have a great deal of flexibility in processing and materials handling capability. Its output can thus encompass a very broad array of products. Once a plant is set up to handle such active alpha emitters as Cm^{242}, such energetic gamma emitters as Ce^{144} and Ru^{106} (Rh^{106}), and such combinations of alpha and neutron emitters as Cm^{244}, modest changes or rearrangements in chemical processing facilities, such as tanks and columns with associated conventional control instrumentation, is all that is needed to assure broadest processing capabilities. This situation encourages consideration of a greater range of products which may be accepted from the reactor operator. The following list presents examples of several of the other isotopes that may be considered for recovery, purification, or packaging in a versatile fuel processing-isotope recovery plant combination.

Isotope	Possible Use
Co^{60}	Heat and gamma source
Tm^{170}	Heat source
Tm^{171}	Heat source
Tl^{204}	Heat source
Po^{210}	Heat and alpha source
Th^{228}	Heat and alpha source
Pa^{231}	Intermediate isotope, "new" element
U^{232}	Heat and alpha source
Am^{242} (152 yr)	Heat source; fissile material
Pu^{244}	Intermediate isotope
Cm^{245}	Fissile material

CONCLUSIONS

A review of the results as shown in Table VI leads to conclusions relating to the value of fission products, the value of the neutrons, the importance of credit for effective neutrons, and the importance of credit for the higher isotopes:

In general, fission products do not appear to have great value to the reactor operator (not much basis for significant credit). This is due to the relatively low inherent value per unit coupled with an indicated high production (processing) cost. This conclusion should not be taken to mean that fission products do not have value; it merely means that their recovery does not promise much as a basis for credit to the reactor operator or owner of the spent fuel. The fuel processor may find, however, attractive possibilities for profit among these. The reality of such profit potential could influence the bid value of fuels for reprocessing with benefit to the reactor operator. However, the pressure for increased value is expected to be small.

The values of Pu^{239} and Pu^{241} as fissionable materials with what appears to be realistic future selling prices and processing costs are significant and amount to about \$73,000 or nearly 75% of the total estimated plutonium value.

The value of the neutrons at \$2500/g which was used in arriving at the credit for reactor operator investment in a number of the higher isotopes as well as the plutonium isotopes results in high additional values. This justifies a more critical evaluation of the value of the neutron to support or modify these credits. Similarly, the extremely high credits assumed for Am^{243} and Cm^{244} should receive attention from the standpoint of more accurately assessing their worth. It is unlikely that in view of the rather low processing costs projected for these elements that such high values could be sustained.

In view of the beneficial potentialities of an integrated fuel processing and isotope recovery facility in producing most economically a number of unique products, it appears that a fairly detailed engineering study of the nature and economics of such an arrangement and the process technology required justifies consideration.

Particularly, it would be of interest to establish the technology for routine recovery of the rare gases and noble metals, and to determine the feasibility of routine americium and curium recovery and separation in the chromatographic ion exchange facilities of the isotopes plant. It should be noted that the recovery of americium in such facilities has just recently been fairly certainly established by work at Battelle-Northwest.[12]

The assumptions on depleted uranium value and processing cost are over-simplified for convenience in this study. It was assumed that the fuel processing cost would be shared equally by uranium and plutonium with the value of uranium set such that it would just cover the cost of processing. This value for depleted uranium is probably too high even for the future when it will be intensively used as a fertile material. However, whatever lower value is used with a corresponding increase in processing cost shared by plutonium would not alter the results of this study to the extent that the general conclusions would be changed.

Although there are many points of this study that may be argued, it is felt that changes would be compensating or minor. For example, the fission product values are so conservatively low compared with the total by-product values that they could even be neglected without changing the picture insofar as credit to the reactor operator or cost of power are concerned. However, if, say, Cm^{244} and Am^{243} were found to have no future value, the impact could be great since of the total credit shown about one-third is attributed to the net value of these two isotopes. It is, however, felt to be unreasonable to consider these as having little or no value in view of the extreme efforts being made today to produce these isotopes at correspondingly very high costs per gram: far greater than are shown in this study.

If it is accepted that neutrons have value that can be credited through sale of those products obtained by neutron capture, the incremental neutron value as used here is certainly a conservative figure. Furthermore, the net yield of the neutrons as used in this estimate is probably unrealistically high (giving low costs). Doubling the value of the neutrons (which still may be a conservative, low, basis) would add about $60,000 of total credits to the figure as listed in Table VI. Such a change would compensate the

unrealistic assumption that perhaps Cm^{244} and Am^{243} would have lesser value. This whole concept of substantial value or credit for by-products as they exist in the unprocessed fuel is therefore based largely on the assumed value of higher isotopes plus the value of the neutrons used in the formation of the precursors for these higher isotopes.

It is re-emphasized that this study attempts to assess conditions as they may exist in a free and competitive reactor fuel processing economy-with reactor operators negotiating for credit for the elements and isotopes as they exist in the unrefined and unprocessed state in the spent fuel elements. The fuel processors would be striving for improved markets and overall process economy for these products and negotiating with reactor operators or fuel owners for most favorable fuel prices and processing charges. It is, of course, unreasonable to assume that the present bases would actually, totally, and precisely exist for this complex assembly of possible products. However, it is felt very strongly that the results as disclosed here form a practical basis for consideration of potential markets, prices, and credits for many of these potential by-products of an extensive nuclear power economy.

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75	CLEARINGHOUSE FOR FEDERAL SCIENTIFIC AND TECHNICAL INFORMATION

*New listing or change in old listing.