THE POTENTIAL FOR LARGE SCALE USES FOR FISSION PRODUCT XENON

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ABSTRACT

Of all fission products in spent, low enrichment, uranium, power reactor fuels xenon is produced in the highest yield - nearly one cubic meter, STP, per metric ton. In aged fuels which may be considered for processing in the U.S. radioactive xenon isotopes approach the lowest limits of detection. The separation from accompanying radioactive ^{85}Kr is the essential problem; however, this is state of the art technology which has been demonstrated on the pilot scale to yield xenon with pico-curie levels of ^{85}Kr contamination. If needed for special applications, such levels could be further reduced. Environmental considerations require the isolation of essentially all fission product krypton during fuel processing. Economic restraints assure that the bulk of this krypton will need to be separated from the much more voluminous xenon fraction of the total amount of fission gas. Xenon may thus be discarded or made available for uses at probably very low cost.

In contrast with many other fission products which have unique radioactive characteristics which make them useful as sources of heat, gamma and x-rays and luminescence as well as for medicinal diagnostics and therapeutics fission product xenon differs from naturally occurring xenon only in its isotopic composition which gives it a slightly higher atomic weight, because of the much higher concentrations of the ^{134}Xe and ^{136}Xe isotopes. Therefore, fission product xenon can most likely find uses in applications which already exist but which can not be exploited most beneficially because of the high cost and scarcity of natural xenon. Unique uses would probably include applications in improved incandescent light illumination in place of krypton and in human anesthesia.

BACKGROUND

Since the earlier review of prospects for use of fission product xenon[1] sufficient changes or advances have come about to encourage this reassessment or update. The changes include revised estimates of fission product xenon availability, of isotopic composition, experience with long term retention in stored, spent fuel, actual production of fission product xenon at ORNL from material recovered from the processing of fuel at the Idaho Chemical Processing Plant, successful testing by Airco of an industrial process for removing ^{85}Kr from xenon down to the pico curie level, expansion of potential uses particularly in the medical field and demonstration of the use of the ORNL xenon by the deposition of heavy metals by the sputtering technique at PNL. A related factor is the continued and expansion of use of natural krypton in improved incandescent lamps.

Xenon Resources and Properties

Xenon is one of the six so-called "inert" gases (which include He, Ne, Ar, Kr, and Rn) that occur in the atmosphere. Only radon is naturally radioactive, but it has such a short half-life (3.8 days for its longest lived isotope) that it can be neglected as a constituent recoverable from the atmosphere. Of the others, only helium occurs in practical concentration in nonatmospheric sources. By far the most important source of helium is certain fuel-type natural gases, from which it is routinely extracted commercially in large quantities. Fastovskii, Rovinskii, and Petrovskii[2] tabulated the abundance of these rare gases in air (Table I). Other properties of these gases are given in Table II.

The isotopic composition of atmospheric (natural) and fission product xenon is as shown in Table III. The isotopic composition of fission product xenon is quite different. This difference in isotopic composition accordingly gives differences in average atomic weight and gas density. Calculated values are shown on Table III. The atomic weight of fission product xenon can thus be expected to be about 134.2 (versus 131.3 for natural xenon). Likewise the density should be about 6.027 (versus 5.897 as shown in Table II).

The Fission Resource

Although on an atomic percent basis xenon is among those elements which are produced in highest yield in the fission of uranium and plutonium, its true position is apparent only when the overall complex of processes and relationships in the productive application of fission is realized. Precursor decay, neutron absorption cross sections, atomic weight and fission yield all go together to place xenon on a weight basis in position of being the most prevalent element of the some forty that are produced in the fission process. In addition, in view of the very short half-lives of accompanying xenon isotopes, it is also the only fission product element which from normal sources is of considerable value and which can be claimed to be essentially free of radioactivity by the time it normally be processed. In other words, although other essentially stable elements are also produced in high yield in the fission process, they are of relatively low price (value) such as zirconium, molybdenum, neodymium, lanthanum and praseodymium and can be more economically recovered from conventional natural mineral resources.

TABLE I. Minor Constituents of Dry Air*.

Gas	Amount in Air Vol%	 ppm	 Wt%	Relative Abundance (Xe = 1) By Volume	 By Weight
CO$_2$	0.033	330	0.050	3,840	1,282
Helium	5.239×10^{-4}	5.239	0.724×10^{-4}	61.00	1.86
Neon	1.818×10^{-3}	18.18	1.267×10^{-3}	211.5	32.5
Argon	0.934	9340	1.288	108,700	33,000
Krypton	1.14×10^{-4}	1.14	3.29×10^{-4}	13.25	8.44
Xenon	0.86×10^{-5}	0.086	0.39×10^{-4}	1.0	1.0

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* Primarily from Reference (2).

TABLE II. Some Properties of the Inert Gases*.

Gas	Atomic Weight		Density (STP), g/liter		Boiling Point of Liquid, °K	Heat of Vaporization at b.p. cal/g mole 1 atm	Heat Capacity (1 atm, 25°C), cal/g mole (°C)	Thermal Conductivity (1 atm, 0°C), cal/(cm)(°C)(sec)
Neon	20.183		0.900		27.07	424	4.969	110.1×10^{-6}
Argon	39.948		1.784		87.27	1555	4.969	40.5×10^{-6}
Krypton	83.80		3.749		119.80	2154	5.005	20.7×10^{-6}
Xenon	131.30	134.2**	5.897	6.027**	165.05	3020	5.022	12.1×10^{-6}

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* Reference (3)

** Estimated values for fission product xenon.

TABLE III. Isotopic Composition of Xenon.

Isotope	Natural,* at. %	Fission product,** at. %
124	0.10	--
126	0.09	--
128	1.91	0.13
129	26.4	
130	4.1	
131	21.2	10.34
132	26.9	16.61
134	10.4	29.21
136	8.9	43.70
Average Atomic Weight	131.3	134.2

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* Reference (4)

** ORNL data. These values will vary slightly depending on the element being fissioned (^{235}U, ^{239}Pu or ^{233}U) the exposure, etc. They will also vary somewhat depending on the amount of contamination introduced from atmospheric xenon.

A metric ton of typical, low enrichment, uranium, power reactor fuel exposed to 33000 MWD/T is estimated to contain 5470g (907.6 liters, STP) of xenon[5]. By the year 2000 about 65000 tons of such fuel will have been produced[6] representing a xenon resource amounting to about 355.5 metric tons. A representative fuel processing plant of 2100 tons per year capacity[7] would be releasing about 11 metric tons of xenon per year (but would not at this capacity be able to keep up with the rate that additional fuel would be accumulating in the year 2000!). Thus, because of the magnitude of its potential availability and for economic (and other) reasons, xenon is one of the few fission products which can be considered as favorably competitive with "natural" materials. The future fission resource of xenon is impressively large. Furthermore, there has been little or no evidence that consideration has been given as to how fission product xenon of large-scale availability and potentially low cost could be usefully employed.

Although fission product xenon at the time of processing will be essentially without activity from radioactive xenon isotopes, it will be associated with radioactive krypton (^{85}Kr) which has a rather long half-life (10.7 years). However, there is such a wide difference in the boiling temperature between xenon (165°K) and krypton (120°K) that separation via cryogenic distillation is feasible as an effective separation and purification method.

The single factor which will have the greatest positive impact on the future realization of large-scale, low-cost availability of fission product xenon is concerned with the air pollution problem of the release of fission product krypton and the accompanying ^{85}Kr to the atmosphere (about 10^7 Ci per year from a typical processing plant). Elimination of this environmental pollutant is already mandated. This will undoubtedly involve the separation and prolonged storage of the krypton gas after separation from the much greater volume of the accompanying fission gas xenon. The intentional discard of the purified xenon resulting from such a process seems incomprehensible. Instead, the likely modest cost associated with final cryogenic purification and packaging would appear to be justified in view of the potential value of this by-product. The price of such material should therefore be relatively low, probably about $1 per liter, STP. The report of Exxon[7] indicates a 1978 cost of about 42 cents per liter. This is far below, probably by a factor of ten or more, existing prices for natural xenon.

At this point comparisons between the fission and atmospheric resources of xenon seem appropriate. With low-enrichment fuels exposed to about 33000 MWD/T processing will release about 5.5 Kg (900 liters) of xenon and 0.371 Kg (100 liters) of krypton per ton of fuel[5]. A representative plant should process about 10 tons of fuel per day (or 2100 tons per year)[7]. Essentially no detectable xenon radioactivity will exist in fuel one year after reactor discharge. The radioactivity of processed xenon will depend therefore on the effectiveness of the process used to remove krypton and its associated radioactive ^{85}Kr. Concentrations of ^{85}Kr of less than 8% in the total fission product krypton can be expected (about 11500 curies per ton of fuel). For the aged U.S. fuel at hand in the future when a processing plant may be expected to be in operation the ^{85}Kr content could be 2% or less[5]. In any case depending on the dissolution process used and the effectiviness of minimizing extraneous air inleakage, the vented fission gas could be an extremely rich composition of xenon and krypton compared with the conventional liquid air process. With regard to the effectiveness of separating krypton from xenon, ORNL about twelve years ago processed several hundred liters of ICCP xenon. The purity attained was 99.93% with an average ^{85}Kr activity of 0.165 micro-curie per liter. At about the same time Airco took a simulated mixture of about this activity (by spiking their commercial xenon with ^{85}Kr) and using special but state of the art cryogenic techniques brought the activity down to the pico curie level - about one millionth of the initial microcurie level. We believe this adequately demonstrates the capability of achieving such extreme purity and also believe that for most common uses such refinement will not be needed. The Exxon study[7] indicated that the bulk xenon from the cryogenic krypton separation process involving 150 day old fuel would have an ^{85}Kr content of about 3.6 microcuries per liter. It is likely that such material could be employed directly for many uses without further purification.

The data on the concentration of xenon in air indicates the rarity of this gas as obtainable from the atmosphere. Obviously, recovery of even reasonable amounts requires the processing of enormous tonnages of air and likely at high cost. The very large scale use of oxygen in the steel industry has resulted in the building of large liquid air plants. Although these plants generally do not yield separated xenon and krypton, the opportunity exists to provide facilities by which the xenon and krypton could be recovered in fairly high yields (85%) from intermediate concentrates that could be separated. Table IV shows the impressive difference between xenon recoveries possible in a large oxygen plant and a representative fuel processing plant.

Using these data it can be shown that in processing a cubic mile of air the oxygen plant would have to operate 3.23 years and in this time would recover 303003 liters of xenon. In contrast the nuclear fuel processing plant could recover the same amount of xenon in about a month (35 days).

Prices

The prices of the rare gases krypton and xenon as commercially obtained from liquid air processes are understood to be about $0.65 per liter (STP) for krypton and about $4.00 per liter for xenon, each for orders of one thousand liters or more. The Exxon study[7] indicated that the selling price for krypton (including all the ^{85}Kr) would be about $31.00 per liter but only about $0.42 per liter for xenon. The reason for this great difference relates to the fact that krypton removal during fuel processing is required to isolate the radioactive ^{85}Kr and that essentially all the costs are thus borne by the need for the krypton processing. Xenon costs therefore relate to hardly more than the load-out costs. It appears then that fission product xenon could be available at a price near, if not less than, the current price of krypton obtainable from atmospheric resources. Furthermore, the quantities of fission product xenon that could be made available would be equivalent to that for krypton from atmospheric resources or at least for that amount of krypton that the market can consume at the present price. It is our understanding that world-wide the availability of xenon is in the range of a few hundred thousand liters annually and for krypton it is in the range of a few million liters.

Uses

The uses for xenon today, though extensive, are generally very specialized and require rather small quantities. In addition, these uses are not sensitive to price. Typical applications include:

thyratron tubes, half-wave rectifier tubes, stroboscopic lamps, bactericidal lamps, point light-source lamps, research lasers, gaseous and liquid scintilation counters and lamps for small and large scale intense illumination (photography). There are also a number of experimental uses of which some are highly price sensitive and also which could consume large amounts. These are particularly in the illumination, metallurgical and health science fields including general lighting, human anesthesia and computed tomography (CT or Cat Scan). In illumination with lower costs and equivalent availability xenon could compete with and in some situations readily displace krypton. Krypton has for many years been used in Europe as well as in the U.S. as a filler gas for certain tungsten filament lights. In this application it partially displaces lower-cost argon and is today's principal use for krypton. However, it should be realized that krypton because of its scarcity and cost could replace only a small fraction of the argon already used in this way. The higher atomic mass (about double) and lower thermal conductivity (about one half) of krypton yields some major advantages over argon. Filament deterioration is significantly inhibited by the higher atomic mass of krypton. Its lower thermal conductivity can also result in smaller sized lamps. The direct benefits of these advantages are that the filament can be operated at higher temperature to yield more light per watt of energy expended without concurrent loss of lamp life or longer lamp life can be assured at equivalent filament temperatures. In addition the smaller size advantage allows a reduction in the requirement for energy intensive raw materials such as glass. Xenon has a substantially higher atomic mass (one and one half times) than krypton; furthermore, its thermal conductivity is also substantially less (about sixty percent) of that for krypton. There features coupled with the possible equivalence in price suggests very strongly that xenon could provide even further improvements in similar illumination applications and with attendant economic justification. The significantly lower projected price for fission product xenon and the attendant major increase in availability strongly supports the possibilities for beneficial applications in the medical field principally for use as a very safe general anesthetic as well as in "Cat Scan" (CT or Computed Tomography) applications where xenon has been found to significantly enhance imaging. Even with today's high cost of xenon Pullicino et al[8] state that this use and in the range of $80 to $100 "per scan adds little to the total cost of patient management". The experimental study of the use of xenon in human anesthesia is well documented. Cullen and Gross[9] in 1951 are credited with the first use of xenon as an anesthetic for conventional surgical procedures on man. Their gas mixture comprised 18 to 22% oxygen, the balance being high-purity xenon. This anesthetic gas was essentially a synthetic air in which all the nitrogen had been replaced by xenon. In a review on inert-gas anesthesia, Featherstone and Meuhlbaecher[10] reported the following observations on the use of xenon as an anesthesic for man: In one case of light anesthesia, there was no evidence of reaction to pain and full recovery was noted in 5 minutes. In another case a satisfactory degree of anesthesia was achieved within a few moments. Excitement during administration was absent, and recovery occurred in 2 or 3 minutes after the administration of xenon was stopped. Man was the only species that could be anesthetized with the xenon gas mixture at atmospheric pressure. Other animals (monkeys and dogs) can be anesthetized with xenon mixtures but only at elevated pressure (2 to 3 atm). Xenon-induced anesthesia is characterized by the unusual brevity of the induction and emergence periods. Equivalent mixtures using krypton were ineffective when administered to human subjects.

For such uses xenon is somewhat limited by the depth to which desired anesthesia can be obtained. Thus for some extensive major surgical procedures conventional anesthetics such as "Halothane" and cyclopropane would continue to be preferred, however for less critical procedures the use of xenon may be preferred because of its overall safety advantages- absence of toxicity and lack of flammability. If in such applications the gas could be used in a conventional recycle type system, we understand that only about eight liters of gas would be required for a single operation. If this were the general practice, a single, representative nuclear fuel processing plant could supply sufficient xenon for over 230,000 operations per year. The entire stockpile of fuel accumulated by the year 2000 would similarly supply sufficient xenon for over 7,000,000 surgical operations.

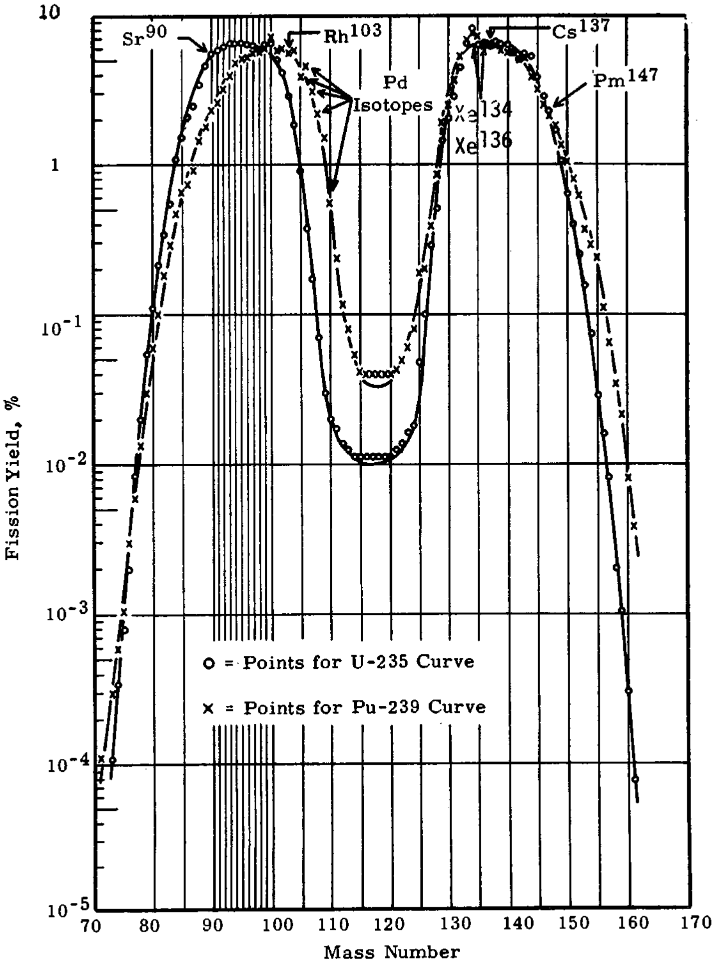
In the metallurgical field fission product xenon has been experimentally tested at PNL for heavy metal deposition by the sputtering technique. This test was successfully demonstrated in producing a massive deposit of copper. This technique with xenon is expected to be even more suitable for use with the heavier metals. The use of sputtering is being developed as a means of metal fabrication and alloy formation to produce unusual shapes and alloy compositions which can not be readily produced by other means. Low cost xenon could be a significant factor in the commercial development of such materials.

Conclusions

The applications as discussed above suggest that most of the existing commercial uses are insensitive to price. Furthermore, these uses are not restricted by the present rarity or limited availability of xenon. It is concluded therefore that future exploitation of fission product xenon will be in new applications which are sensitive to price and where large scale-availability and supply are important. The future applications which can then be considered when commercial, nuclear fuel processing is realized include applications in the health sciences field, metallurgy (sputtering) and special incandescent illumination devices. In addition uses less demanding of large scale availability but still encouraged by availability and price will include expanded use in the field of high intensity illumination, gaseous isotope separation and other heavy ion concepts as well as radiation detection and analysis applications.

TABLE IV. Comparison of Daily Xenon and Krypton Production between Oxygen and Fuel-Processing Plants.

Plant	Krypton, liters(STP)	Xenon, liters(STP)	Krypton in mixed gas, vol.%	Xenon in mixed gas, vol.%
Oxygen (10^3 short tons/day, 85% recovery for Xe)	3407	257	93	7
Fuel processing (10 metric tons/day, 95% recovery for Xe)	936	8620	9.8	90.2



Yields from Thermal Fission of U^{235} and Pu^{239}
From: Katcoff, Nucleonics 18 No. 11, 201 (1960)

REFERENCES

1. C. A. Rohrmann, Fission-Product Xenon and Krypton-An Opportunity for Large-Scale Utilization, Isotopes and Radiation Technology. Vol. 8, No. 3, Spring 1971.

2. V. G. Fastovskii, A. E. Rovinski, and Yu. V. Petrovskii, Inert Gases, USAEC Report AEC-tr-6725, translation from the Russian by J. Schmorak, Jerusalem, Israel Program for Scientific Translations, 1967.

3. AIRCO Rare and Specialty Gases, catalog of Air Reduction Co., Inc., 150 E. 42nd St., New York, N.Y. 10017, 1968, p. 18.

4. General Electric Co., Chart of the Nuclides, Twelfth Edition, 1977.

5. W. J. Zielenback, BMI-X-686, Annual Report, Task 83 on Studies of Beneficial Uses for Krypton-85 in Energy Applications, September 30, 1977.

6. DOE/RL-82-1, Spent Fuel Storage Requirements, Table B-1, page 40, June 1982.

7. XN-FR-ER-2 Rev. 1 Exxon Nuclear Company, Inc., Final Report for Study of the Separation and Recovery of Select Radioisotopes from Commercial Nuclear Fuel Wastes, January, 1978.

8. P. Pullicino, G. H. In Boulag and B. E. Kendell, Xenon Enhancement for Computed Tomography of the Spinal Cord, Neuroradiology by Springer-Verlag 1979.

9. S. C. Cullen and E. G. Gross, The Anesthetic Properties of Xenon in Animals and Human Beings, with Additional Observations on Krypton, Science, 113:580-582 (May 18, 1951).

10. R. M. Featherstone and C. A. Muehlbaecher, The Current Role of Inert Gases in the Search for Anesthesia Mechanisms, Pharmacol. Rev., 15:97-121 (1963).

11. S. Katcoff, "Fission-Product Yields from Neutron Induced Fission, Nucleonics 18(11):201 (1960).