

AN OVERVIEW OF BYPRODUCT UTILIZATION

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ABSTRACT

Useful and strategic byproducts exist in the high-level nuclear waste of the defense and civilian fuel cycles. These byproducts can be used for energy sources, process photon sources, light-producing electron sources, strategic metals, and other useful applications. The partitioning, extraction, and use of byproducts has the potential for significantly contributing to the closure of the civilian fuel cycle, for facilitating the management and disposal of residual high-level waste materials from both fuel cycles, and for using the peaceful atom to solve a number of major societal problems both in the U.S. and abroad.

BACKGROUND AND HISTORICAL OVERVIEW

The concept of byproduct utilization is not a new one; it evolved from the Atoms-for-Peace initiatives of the late 1950's,^{1, 2} work on isotope production and development in the 1960's,³ and a new look at the concept in the mid-1970's which was presented at the Second Annual Waste Management Symposium here in Tucson⁴ and in hearings in the U.S. Senate.⁵ In the 1980's, the U.S. House of Representatives also held hearings on the subject.⁶

In the 1980's a number of synergistically related events occurred which tended to support the program as a new Federal initiative. In summary, these events included the following:

- Federal commitments and resolves to close the nuclear fuel and waste cycles;
- Increasing recognition that the separation and use of byproducts can facilitate the management and disposal of high-level wastes;
- New requirements for secure, rugged, and independent energy and light sources in the defense and civilian sectors;
- Changes in regulatory philosophy both here and abroad which are beginning to allow radiation processes to compete more effectively with conventional processes for treating various commodities;
- The advent of effective advanced isotope separation processes to extract and refine byproducts; and
- An emerging awareness that U.S. sources of strategic materials are insecure.

At the same time significant progress has been made in byproduct applications engineering. Experimental work at the Sandia Laboratories irradiator fueled with cesium, a defense fuel cycle byproduct from Hanford, Washington, has demonstrated the efficacy of byproduct irradiation in sanitary

engineering, agricultural and food processes to eliminate pathogens and certain chemical carcinogens. The use of radioluminescent lighting with isotopes has recently been demonstrated in a number of field experiments in the Continental U.S., Hawaii, and Alaska.

The extraction of byproducts and recycle of metals in reprocessing defense materials is currently being evaluated for application at both the Hanford and Savannah River Plants.

The above topics will be briefly summarized below. Subsequent papers will address these subjects in some depth.

FUEL CYCLE CLOSURE

Fuel cycle closure (reprocessing) is inherent in the defense fuel cycle. In addition, cesium and strontium byproducts have been separated at Hanford and have facilitated waste management there. Cesium, noble metals, and process mercury for recycle may also be partitioned from the defense wastes at the Savannah River Plant. The extraction of additional byproducts is being considered at both locations. The defense fuel cycle has progressed to the point of reprocessing, and work is either underway or planned for the immobilization of waste residues for geologic disposal.

A comparable degree of closure has not been effected in the civilian fuel cycle. The commercial fuel and most of the waste now reside in the form of about 9000 tons of spent fuel elements stored at 80 or so nuclear power plants and in two inactive reprocessing plants around the country. Although reprocessing of this fuel is encouraged by Federal policy, the private sector has shown no interest in making the heavy investments that a commercial reprocessing industry would require. The only remaining alternative would be to merge the defense and civilian fuel cycles, for example, at Federal facilities and sites. This alternative has not been seriously considered or systematically evaluated, although existing and projected defense facilities and sites can be modified to satisfy both cycles. In such a merger, the partitioning of byproducts can enhance the capabilities and economics of the combined fuel cycle. The economic benefits accruing from the adaptation of proven processes, facilities, and

personnel; proven waste management, interim storage, and waste immobilization techniques; and use of common waste disposal facilities and sites may be in the tens of billions of dollars; and both fuel and waste cycles could be closed.

The joint closure of the U.S. defense and civilian fuel cycles warrants an in-depth evaluation based solely on technical and economic factors. Then the advantages and disadvantages of maintaining two separate cycles could readily be compared with those of a merged cycle. It is quite possible that this is the only viable alternative to a "throw-away" fuel cycle, where the valuable constituents in spent fuel are abandoned.

By comparison a recent Soviet paper indicates that the USSR program "was planned from the very beginning with the closed fuel cycle" with the principal goal being to recover uranium and plutonium.⁷ This paper is also explicit that the Soviets are providing for "subsequent valuable element extraction"; strontium, cesium, technetium, palladium, and certain rare earths are specifically mentioned.⁷ There is no evident demarcation between the defense and civilian fuel cycles in the Soviet Union.

WASTE MANAGEMENT FACILITATION

There has always been a realization that partitioning, fractionation, or separation of certain radioisotopes or suites of radioisotopes could facilitate waste management and at the same time produce useful byproducts.⁸ More recently Cohen⁹ has independently evaluated the potential hazards from geologic disposal of high-level radioactive wastes and cited the dominant troublesome radionuclides. The coincidence of the isotopes having potential uses and those dominating the hazards of buried waste is striking. The convergence of these independently derived conclusions points in the direction of waste management facilitation via byproduct segregation.

Papers have been presented by Platt, et al.,¹⁰ and Northrup, et al.,¹¹ "on the desirability of removing heat and penetrating gamma radiation from high-level waste. The removal of strontium and cesium can reduce the thermal burden by 98% and the gamma flux by 97% in high-level nuclear waste 30 years after reactor discharge." This can facilitate the handling, interim storage, transportation, and ultimate disposal of the residual high-level waste. The residual waste, after strontium and cesium removal, is more stable and its long-term behavior in a geologic repository is more predictable, enhancing its environmental and safety attributes appreciably.

The initial extraction of other useful byproducts such as americium, neptunium, plutonium, uranium, and platinum family metals will reduce the waste to still safer radiation levels for disposal and will reduce the costs to manage waste.

Cohen's¹² analysis of the problematical constituents of high-level waste in geologic disposal shows that strontium and cesium dominate up to 200 years. The advantages of removing the heat and radiation existing in these materials and their uses are discussed herein and in subsequent papers of this symposium. Americium dominates from 1500 to 10,000 years; Americium-241 targets are transmuted by

neutron capture to make medical grade Plutonium-238 which has application in heart pacers and is one of the primary candidates to power the artificial heart pump. Neptunium-237 dominates the hazard from the geologic repository from 50,000 to 20,000,000 years.¹² Neptunium-237 targets are transmuted by neutron capture to make Plutonium-238 fuel for reliable, long-lived nuclear generators. Such generators have been used on 12 space missions to power lunar experiments, Martian landers and interplanetary spacecraft to explore Jupiter, Saturn, Neptune, Uranus, and Pluto--an example of both beneficial use and safe ultimate disposal of more than a million curies of trans-uranium material in space.

The isotopes utilized in terrestrial applications must be considered for recycle and disposal after use. Recycle will effectively diminish the heat and radiation of fission products during utilization, followed by disposal tailored to the isotope. Trans-uranium isotopes having long half lives can undergo recycle and reuse after reconcentration by chemical or advanced isotopic separation. This mode of disposal for used isotopes is similar to that considered for partitioned high-level wastes. Specific isotopes have more quantified and predictable long-term properties than the potpourri of isotopes in high-level wastes.

POWER AND LIGHT SOURCES

In his testimony to the House Armed Services Committee, the Under Secretary of Defense for Research and Engineering cites as "our most important hardware initiative" the modernization of U.S. military command, control, communications, and intelligence (C³I) capabilities. He cites the need for "flexible, reliable, and enduring C³I systems" and states that of the 20 most important basic military technology areas the Soviets are superior in two--power sources and conventional warheads.¹³

Isotopic power sources have a number of demonstrated and potential applications in low powered C³I systems. Isotopic power sources have performed for more than 10 million mission hours without failure in hostile space, marine, and terrestrial environments. They are inherently rugged, reliable, independent, and enduring and do not require fuel and maintenance over their life cycle. They utilize Plutonium-238 or Strontium-90 fueled heat sources and solid state thermal to electrical converters. It is not unusual for an isotopic power system to supply useful electrical power for 15 years without refueling or maintenance, totally unattended.

Aside from critical military applications, civilian applications exist for isotopic power systems for ground-based control, communication, meteorological and navigational systems for civil aviation and maritime activities.

Radioluminescent lights using beta emitting isotopes, such as Krypton-85 or tritium interacting with a phosphor have been developed for civilian and military applications. They do not present a source of ignition around combustible and explosive materials and have superior safety properties compared to electric lights. Since radioluminescent lights do not require an electrical power source, have a high operational reliability, and are rugged enough to be air-dropped, they have a variety of applications. These include delineation of runways, air drop zones,

taxiways, VTOL landing zones, and other avionic uses to be discussed at length in a subsequent paper.

RADIATION PROCESSING

Gamma radiation can be used instead of fumigation, thermal or chemical treatment to extend the shelf life and eliminate parasites, viruses, bacteria, molds, and insects in various commodities without degrading the commodity or imparting harmful chemicals to it. For example, ethylene dibromide, a common fumigant for pest control of agricultural products, has resulted in significant worker health effects, according to the Occupational Safety and Health Administration, and is a known carcinogen when ingested with the fumigated product. Gamma radiation is expected to displace chemical methods and today is routinely used on most medical products in the U.S.

In 1980 the U.S. Food and Drug Administration Irradiated Food Committee recommended that food irradiated at doses of 100,000 rads or less is wholesome without toxicological testing and that foods comprising no more than 0.01 percent of the daily diet and irradiated at 5,000,000 rad or less are considered safe for human consumption without toxicological testing.¹⁴ At about the same time, the IAEA, United Nations, and World Health Organization concluded that foods irradiated to 1,000,000 rads are unconditionally accepted with no requirement for wholesomeness testing. These events pave the way for the radiation processing of food and agricultural products in the U.S. Food irradiation plants are already operating routinely in Europe and Asia using cobalt.

In the U.S., the cesium irradiation program is centered around a 7250 kilogram product throughput per day pilot plant at Sandia National Laboratories.¹⁵ Tests on pathogen removal from sewage sludge have led to the authorization of a full scale plant for the City of Albuquerque. The irradiator has also been successfully used at low doses to disinfest grapefruit and mangos of the larvae of the fruit fly, as a safe substitute for fumigation with carcinogenic chemicals. More recent successful experiments on pork demonstrate that trichinae can be eliminated with low doses. The potential exists for eliminating trichinosis with radiation. Low dose applications look particularly promising for early commercialization for agricultural commodities.

THE ADVENT OF ISOTOPE SEPARATION

The advent of photochemical processes for separating elements and isotopes has added a new dimension to the concept of extracting and purifying byproducts from high-level nuclear wastes. Other authors will describe the laser isotope separation and plasma separation processes here today.

Laser and plasma separation could replace wet chemistry in some processes and also could serve to remove a radioactive isotope from stable isotopes of the same element to facilitate its use. For example, they could be used to separate krypton and xenon noble gases from one another and to separate Krypton-85 from stable krypton isotopes. Xenon could be used for conventional lighting, lasers, and anesthetics; stable krypton could be used for conventional lighting, and inert gas applications; and Krypton-85 could be used for electronic lighting and

non-destructive testing. These processes may be helpful for processing radioactive material from effluent gas streams.

Advanced isotope separation offers the first opportunity to separate significant quantities of platinum family metals from their radioactive isotopes. For example, the removal of Rhodium-102, Palladium-107, and Ruthenium-106 would leave behind stable platinum family metals for the National Strategic Stockpile. Chemical processes to extract platinum family metals from defense wastes are currently being explored.

The separation of other isotopes with advanced processes is also being explored. Among the candidates are isotopes with uses in nuclear medicine. Today there are 80,000,000 to 100,000,000 nuclear medical procedures conducted annually in the U.S. resulting in the saving of more than 10,000 lives each year. Nuclear medicine is the most successful result of President Eisenhower's Atoms-for-Peace initiative of the 1950's.

STRATEGIC MATERIALS

The Soviet Union and South Africa have historically produced and marketed about 90 percent of the platinum group metals; the U.S. has produced only a negligible amount of these materials. World yearly production is about 6 million troy ounces. The U.S. imports 94% of its annual needs and obtains the remainder from recycle. These materials are critical to both military and commercial technologies. The U.S. and the rest of the free world are vulnerable to supply interdictions and the cost of their importation contributes to balance-of-payments problems.

An example of this situation is reported in two recent Wall Street Journal articles.^{16, 17} The first article indicates that palladium prices increased dramatically from June to December, 1982. Palladium and platinum prices could rise further because "consumer inventories are very low." The U.S. strategic stockpile is far below its goals: 1.25 million ounces of palladium versus a 3 million ounce goal and 453,000 ounces of platinum versus a 1.3 million ounce goal. The second article¹⁷ states that the Soviets are cutting their supplies of palladium to the West by 25 percent. While the Soviets are the world's largest palladium producer, South Africa has reduced its annual output by one third this year. The price of palladium increased 50% from early October to mid-December of 1982.

While platinum does not exist in appreciable quantities in spent fuel, other fission product metals in spent fuel such as palladium, ruthenium, rhodium, and technetium can substitute for platinum in catalytic processes. For example, technetium¹⁸ and ruthenium¹⁹ can be used as a substitute for platinum in refining hydrocarbon fuels. The combined quantity of platinum family metals and technetium in U.S. nuclear wastes by the year 2000 is estimated to be 50 to 100 million ounces, including 1 to 2 million ounces of palladium.

Once these byproducts are recovered, they can be either cleaned up with isotopic separation, stored in a stockpile while their activity decays to usable levels, or used as is in closed cycle catalytic systems.

SUMMARY

The extraction and use of byproducts from the defense and commercial fuel cycles is not an end in itself but a means to an end. When integrated into the nuclear fuel and waste cycles, a byproduct option can serve to enhance the closure of both cycles. Otherwise, unique, valuable, and strategic national resources can be lost forever in a "throw-away" cycle or irretrievably sealed in a geologic repository. The byproduct concept is beginning to be demonstrated in the defense fuel cycle and can help to close the commercial fuel cycle. The large byproduct inventories in commercial spent fuel should be seriously considered when setting national policy and evaluating the economics of reprocessing.

REFERENCES

1. Hittman, F., "The Feasibility of Producing Power from Radioisotopes," American Nuclear Society, June, 1957.
2. Rupp, A. F., "Large Scale Production of Radioisotopes," Proc. Int. Conf. Peaceful Uses of Atomic Energy, 14, 68, 1956.
3. Rohrmann, C. A., "Values in Spent Fuel from Power Reactors," Isotopes and Radiation Technology, Vol. 6, No. 1, 1968.
4. Dix, G. P., "The Beneficial Uses of Nuclear Waste Products," Second Annual Waste Management Symposium, Tucson, AZ, 1975.
5. Dix, G. P., "The Beneficial Uses of Nuclear Waste - 1978," U.S. Senate Nuclear Waste Disposal Hearings, Part 1, Ser. No. 95-136, pp 251-271.
6. Committee on Armed Services, House of Representatives, "Beneficial Uses of Defense Nuclear Byproducts," HASC No. 97-3, March, 1981.
7. Dubrovsky, V. M., et al., "The USSR Experience in Nuclear Power Plant Spent Fuel Handling Including Storage and Transportation," IAEA-CN-42/88, Vienna, September 13-17, 1982.
8. Rupp, A. F., "A Radioisotope-Oriented View of Nuclear Waste Management," ORNL 4776, May, 1972.
9. Cohen, Bernard L., "Effects of ICRP Publication 30 and The 1980 BEIR Report on Hazard Assessments of High-Level Waste," Health Physics, Vol. 42 - No. 2, February, 1982.
10. Platt, A. M., Eshbach, E. A., "Rethinking the Management of High-Level Waste: The Need for Fractionation," PNL, April, 1981.
11. Northrup, C. J., Jardine, L. J., Steindler, M. J., "An alternative Strategy for Commercial High-Level Radioactive Waste Management," IAEA SM-261/34, June, 1982.
12. IBID 9.
13. De Lauer, Richard D., "Hearings on Military Posture - Research and Development - Title II," HASC No. 97-33, March, 1982.
14. FDA Bureau of Foods, "Recommendations for Evaluating the Safety of Irradiated Foods," July, 1980.
15. Sivinski, J., Ahlstrom, S., McMullen, W. H., and Yeager, J., "The DOE/EPA Sludge Irradiation Program," International Association of Water Pollution Research Meeting, March, 1982.
16. Behrmann, Neil and MacKay-Smith, "Russian Bear Learns New Tricks as Soviets Create a Bull Market for Their Palladium," The Wall Street Journal, p. 20, November 26, 1982.
17. Staff Reporter, "Soviets Seen Cutting Palladium Supplies by 25% to the West," The Wall Street Journal, p. 32, December 20, 1982.
18. Blackham, et al., "Technetium Catalyst for Hydrocarbon Reforming" Pat. 3,868,333, February 25, 1975.
19. Chemical and Engineering News, "Ruthenium-Catalyzed Hydrogenation Promising," p 44ff, September 8, 1980.