

CURRENT STATUS OF THE DEPARTMENT OF ENERGY'S PROGRAM
FOR RECOVERY AND UTILIZATION OF NUCLEAR BYPRODUCTS

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ABSTRACT

Nuclear waste from Defense Nuclear Production contains many useful, valuable, and strategically important materials. These materials have a wide range of known and potential applications in medicine, public health, energy, food technology, industrial technology, and national security. The separation and removal of several of these materials from the nuclear waste stream has the potential to facilitate waste management and yield potential economic, safety, and environmental advantages in the management and permanent disposal of the residual nuclear wastes that have no known redemptive value.

The current three primary program elements, byproduct recovery, byproduct application, and the impact assessment of these prior activities on nuclear waste management, will be implemented only when it can be clearly determined to be safe, environmentally acceptable, and cost effective. Goals have been set for an integrated research, development, demonstration, and technology transfer program for byproducts applications from defense and other nuclear activities. It is intended that the research, development, demonstration, and technology transfer will be a cooperative, resource sharing activity among DOE Defense programs and known and potential users in the Federal, State, and private sectors, as well as foreign states and international agencies.

Emphasis is currently being placed on applications with the two highest activity byproducts, strontium-90 and cesium-137. One of the strontium-90 initiatives is the design and construction of a relatively high power (500 watts) radioisotope thermoelectric generator. Cesium-137 applications for both sterilization of sewage sludge and low-dose disinfestation of food are being cooperatively developed and demonstrated with other Federal agencies and industrial groups.

In addition, tritium radioluminescent airfield lights are being developed jointly with the Department of Defense and the States of Alaska and Florida. These lights have already been demonstrated in airfield lighting applications during highly successful operations.

Areas that are addressed include: status of current byproduct applications, current supply of byproduct materials to meet potential needs resulting from commercialization, and possible ways of increasing the future supply of byproduct materials from defense and civilian nuclear fuel cycles.

INTRODUCTION

Because of the potential value of byproduct recovery and utilization as well as a desire to convert a perceived liability into a national resource, the Department of Energy has established a policy to encourage widespread use of byproducts in meeting unique national needs and in solving societal problems. In concurrence with this policy, goals have been set for an integrated research, development, and demonstration program for use of byproducts from the defense fuel cycle and other nuclear activities.¹ The effort, under the direction of DOE's Office of Defense Waste and Byproducts Management, includes byproduct recovery, development of useful applications, and an evaluation of the impact of byproduct recovery and use on nuclear waste management and disposal. All program elements will be implemented when it can be clearly shown that they are safe, environmentally acceptable, and cost effective.

Thus, the recovery of byproducts from nuclear waste and their beneficial use depend on one or more of the following guidelines:

- The byproduct must have unique properties that match unique requirements of a specific application.
- The byproducts must be present in sufficient quantities in defense fuels to justify recovery and beneficial use.
- Recovered byproducts must be marketable at an economic advantage acceptable to the user and the producer.
- High value may be assigned to a byproduct or recycled material because the material meets a critical military or national strategic need.

- Removal of selected byproducts may reduce the cost for managing the residual waste.

Nearly 500 isotopes are produced in nuclear reactors by fission and irradiation processes. Several of these can be considered recoverable at the present time using the above guidelines, and others may be recoverable as future conditions warrant. Candidates for recovery and utilization at this time include the following grouped by potential application:

Process irradiation
cesium-137

Radioisotopes for heat and power
strontium-90 cesium-137
krypton-85 promethium-147
plutonium-238 (from neptunium-237)
curium-244

Special value and unique isotopes or elements
americium-241 technetium-99
ruthenium rhodium
palladium krypton-85
xenon

Byproducts useful as targets for production of other elements
neptunium-237 americium-241
curium-244

Byproducts useful for luminescence applications
tritium krypton-85
promethium-147

Emphasis is currently placed on application for the two highest activity byproducts strontium-90 and cesium-137, although other byproducts may have strategic value or other known applications in medicine, public health, industrial technology, or national security. Also, a major effort has been expended to develop the use of tritium for radioluminescent lighting.

This paper is an overview of the DOE sponsored studies of the use of beneficial byproducts. More detailed papers on various aspects of the program will be presented later in this session.

FOOD IRRADIATION ACTIVITIES

The DOE Byproducts Utilization Program supports current initiatives in food irradiation.² The goal of these initiatives is to develop and remove the impediments to commercial beneficial uses of cesium-137, which decays with emission of gamma radiation. Cobalt-60, an activation product from irradiation of cobalt-59 also producing gamma radiation, is currently the major isotopic source for irradiation treatment but it is currently in short supply.

The focus of food irradiation activities in the Byproducts Program is on low-dose radiation treatment of foods at levels below 100 krad. These doses are appropriate for insect disinfestation of fresh produce and for destruction of parasites in meat. This focus is somewhat different than the historical AEC low-dose food irradiation activities that stressed the potential extension of the shelf life of fresh foods through radiation treatment, but is consistent with the research and development approach of the Atomic Energy Commission (AEC) during the "Atoms for Peace" program of the 1960's. The following research, full-scale facility validation and technical support activities are supported by DOE to foster the use of cesium-137 in irradiation of food.

Research

Cooperative research in food irradiation is being carried out with the National Laboratories, U.S. Department of Agriculture (USDA), and several universities. The objectives of these research efforts are to define the parameters critical to the irradiation treatment of agricultural commodities. First, the dose must be established that effectively inactivates specific pests so as to bring commodities in compliance with Federal quarantine regulations. Secondly, the effective dose must not adversely affect those commodity properties such as color, odor, taste, etc.

Citrus

The DOE is cooperating with the USDA/Agricultural Research Service in Florida to determine the viability of irradiation treatment of grapefruit as an alternative to ethylene dibromide (EDB) fumigation quarantine treatment. Several small-scale irradiation treatments of Florida grapefruit have been completed at the Sandia National Laboratories' cesium irradiation facilities. These small-scale treatments have led to larger scale studies conducted by USDA in various commercial cobalt-60 irradiation facilities and have established a basis for the implementation of this treatment by industry faced with an Environmental Protection Agency ban on the use of EDB as a citrus fumigant after September 1, 1984.

Dried Fruit and Tree Nuts

Cooperative efforts are under way with the USDA/Agricultural Research Service, USDA/Economic Research Service, Oregon State University, the California Almond Board, and the California Prune and Walnut Board to assess the feasibility of treating almonds, raisins, and walnuts with radiation as a partial replacement for fumigation with methyl bromide. These commodities are produced in tremendous quantities (about 26,000 tons/day) in California during harvest and must be fumigated several times to destroy feeding insects. Final fumigations applied after the commodities are packaged can leave residues of the chemical greater than those acceptable to foreign importers. A final treatment with radiation could potentially accomplish the same disinfestation result with a concurrent reduction in residual chemical fumigant levels. Insect susceptibility to irradiation, product responses, taste panel tests, and economic feasibility studies are being conducted by Oregon State University to assess this potential application of food irradiation technology.

Fresh Pork

Efforts with the USDA Animal Parasitology Institute, the National Pork Producers Council (NPPC), and Iowa State University have led to the initiation of a design for a demonstration facility to irradiate fresh pork to inactivate trichina and other parasites which pose a potential health threat to man. Much of the data gained in this activity supported by the DOE will be used in an NPPC petition to the FDA to include irradiation of fresh pork in their final regulations. An assessment of consumer attitudes regarding food irradiation in general and pork irradiation in particular has been conducted under the direction of NPPC and has indicated important issues (e.g., wholesomeness, effectiveness, etc.) that must be addressed if irradiation treatment of foods is to be accepted by consumers.

Shellfish

The DOE is supporting the University of Lowell, Massachusetts, in efforts to assess the potential for

irradiation treatment of shellfish (clams, oysters) harvested from potentially polluted beds along the eastern coast of the United States to safely clean them prior to their introduction to the marketplace.

Clams from lightly polluted beds are cleansed (depurated) in clean seawater exposed to ultraviolet light where pathogens residing in the tissues as a result of pollution are expelled into the clean seawater and destroyed. This technique is not applicable to clams harvested from moderately or highly contaminated beds. Initial research has shown that shellfish can withstand substantial doses of gamma radiation during depuration.

Irradiation of Northwest Agricultural Products

Pacific Northwest Laboratory (PNL) is currently conducting a DOE-sponsored experimental program in conjunction with the U.S. Department of Agriculture (USDA) laboratories in Washington State. The goal of the program is to evaluate the potential benefits of irradiation to the agricultural industry in the Northwest. The near-term objective is to evaluate the need for a demonstration irradiator to irradiate a variety of products in sufficient quantities to establish the market potential and to stimulate public acceptance. Initial efforts have concentrated on irradiating apples and cherries to disinfect them of codling moths and cherry fruit flies. Irradiation appears to be the treatment of choice to meet the quarantine requirements for export to the Far East.

Transportable Cesium Irradiator (TPCI)

To conduct meaningful research on irradiation treatment of fresh commodities, the effect of complicating variables (such as time since harvest, temperature of storage and shipment, distance and time of shipment, etc.) must be minimized. For research results to be valid, irradiation of the particular pests of concern must be conducted at the appropriate stage in the life cycle of the pests. These factors have dictated the need for a small-scale flexible research facility that can be transported to sites where infested commodities are harvested or collected for determination of the applicability of irradiation. The transportable irradiator will be a flatbed-truck-mounted cesium irradiation unit and is being fabricated by the Foster Wheeler Corporation and will contain approximately 250,000 curies of cesium-137 in four WESF capsules. Operation should commence in 1985 under a license by DOE or the Nuclear Regulatory Commission as appropriate. TPCI will be capable of irradiating unit-size cartons of commodities as part of research initiatives on various products. Use is projected at multiple locations throughout the country, including USDA and DOE laboratories as well as by commercial firms interested in gaining limited basic research data on the feasibility of irradiation treatment of their specific products.

Demonstration Irradiators

To accomplish the technology transfer goals of the DOE program, several full-scale facilities are being designed and fabricated.

These facilities will serve as a validation of cesium irradiation technology and will address technical and institutional issues such as licensing, economics, operational reliability, etc., which will influence the potential for commercial implementation of the technology. Two full-scale food irradiation facilities are planned: the cesium agricultural commodities irradiator (CACI) and the fresh pork irradiator.

Cesium Agricultural Commodities Irradiator (CACI)

The cesium agricultural commodities irradiator is currently being designed by Rockwell International to contain 3 million curies of cesium-137 to evaluate the benefits of irradiation on citrus fruits and various other agricultural commodities. This facility is designed to determine irradiation conditions and will provide sufficient quantities of irradiated commodities for assessments of economics, storage, and marketability. It will also serve as a tool to evaluate feasibility of radiation treatment of commodities from the Caribbean Basin area, which must currently be fumigated with EDB in order to meet United States quarantine restrictions.

The commodities irradiator project will also involve the Agency for International Development (AID) as a cooperating agency. The facility will be used by USDA and AID to train students, faculty, and operators from developing countries of the Caribbean Basin and will be utilized by visiting scientists to conduct research on commodities of regional interest. The economics of irradiating pork appears favorable with the costs by large processors estimated at less than 1 cent per pound. Construction of CACI is planned for the Oakland, California, area.

Fresh Pork Irradiator

This facility is being designed to validate for the pork packing industry the concept of using irradiation treatment to produce trichina-safe fresh pork. The facility will use 2 to 3 million curies of cesium-137 in Waste Encapsulation and Storage Facility (WESF) capsules and a conveying mechanism for whole or split hog carcasses, which simulates the systems used in packing plants.

It is DOE's intent to site this facility at a university to serve as host for a broad-ranging irradiation research institute, using the irradiator as a foundation. The facility will be used to extensively evaluate the options for irradiation of fresh pork and will conduct assessments of irradiation effectiveness; potential changes, if any, caused in the meat by irradiation involving taste, color, odor, and other properties; the economics of the process; and the need for specialized temperatures or atmospheric environments.

DOE is competitively soliciting a site for this irradiation institute; selection is expected in FY 1985 and design and fabrication activities will start shortly after site selection.

Technical Support Activities

In addition to food irradiation research and facility validation initiatives being supported by DOE, several efforts supporting the technology in a broader fashion are being pursued.

Beneficial Uses Shipping System (BUSS)

The DOE is fabricating a large radioactive shipping container able to carry 1 million curies of cesium-137 in 16 capsules at a time. This cask will be compatible with the DOE Waste Encapsulation and Storage Facility (WESF) at Richland and the demonstration irradiators. The BUSS will enable more economic transport of large quantities of isotopes and will be licensed by the DOE and the Nuclear Regulatory Commission (NRC) for potential use in commercial irradiation facilities. The thick-walled BUSS body has been fabricated, and analyses are nearing completion so that licensing activities can commence.

WESF Capsule Evaluation

The WESF capsules available for use as radiation sources have been extensively tested by DOE to ensure their integrity under a variety of operating and potential accident conditions. These tests have been documented by DOE in various reports and have been recently summarized in a Pacific Northwest Laboratory report,³ which has served to expedite our negotiations with the NRC concerning the acceptability of using these sources in DOE, as well as commercial irradiation facilities. DOE is currently interacting with the NRC to determine specific acceptable irradiator designs and conditions for safe use of the cesium-137 capsules as radiation sources. Additional tests of the capsules are being conducted to justify use of the sources in irradiators under varying operating conditions.

Under the planned terms of DOE's lease of cesium capsules to industry, DOE will accept responsibility for return of the sources for ultimate disposal once their radioactivity levels have decreased below those justifying continued use. Because DOE has an ongoing interest in the performance of these sources over time, capsules from both DOE facilities and commercial facilities may be periodically removed for examination and evaluation of their performance. These evaluations will contribute to the data base regarding capsule performance under diverse conditions and will assist in guiding future DOE byproduct encapsulation activities.

Agency for International Development (AID) Cooperation

The DOE has recognized the international interest in food irradiation and its potentially dramatic applicability to many developing countries of the world. In those countries where refrigeration and transportation systems may be inadequate, radiation may play a significant role in increasing food supplies through the reduction of postharvest losses. DOE is cooperating extensively with AID to assist in evaluating the potential for irradiation in the Caribbean Basin, Central and South America, and Southeast Asia.

Cesium-137 Supply

Cesium-137 is one of the major fission products generated in nuclear reactors and, thus, is a byproduct from both power and defense reactor fuels. To aid waste management, this isotope has been recovered and encapsulated from the defense reactor waste at Hanford since 1967; 1575 capsules, containing about 86 million curies of cesium, have been produced. Material from a few of the capsules has been used for various purposes and radioactive decay has occurred, leaving a current inventory of about 77 million curies.

At present, DOE has more requests for cesium-137 sources than are available. An additional 40 million curies will be separated from Hanford defense waste through 1991. Although current plans stipulate disposal of this material, it could be made available if there is demand for it. The stored defense nuclear waste at Savannah River contains about 100 million curies with another 100 million to be generated through the year 2000. However, facilities for purification and encapsulation are not currently available at Savannah River.

A study nearing completion by DOE is evaluating the desirability and cost of recovering cesium-137 from Savannah River and from future waste to be generated at Hanford. This study examined the feasibility and projected costs of separation and encapsulation at Savannah River and also that of recovery at Savannah River and shipping to Hanford as a dried solid sorbed

on an ion-exchange resin. Both approaches appear to be feasible provided the regulations permit shipment of the material and the demand justifies the recovery and encapsulation costs. Projected costs for future cesium capsules at Hanford are of the order of \$0.80 to \$1.00/Ci. Present generation cesium from Savannah River is projected to cost about \$1.00/Ci if shipped to Hanford for encapsulation. Similar costs were projected if the encapsulation were performed in modified existing facilities at Savannah River but were more than doubled if new facilities were constructed. Stored waste at Savannah River could also be recovered in the Defense Waste Processing Facility (DWPF) and is projected to cost about \$1.50 and \$1.25/Ci if encapsulated at Savannah River and Hanford, respectively.

The largest source of byproduct cesium is in the spent fuel from commercial power plants where an estimated 500 million curies are contained. This amount will increase to about 11 billion Ci by the year 2020. Recovery of the cesium-137 from spent fuel can be considered only if fuel reprocessing were undertaken or if spent fuel disposal requires removal of the most volatile radioactive species before storage. Presently available cesium would supply only eight average-sized (10 MCi of cesium-137) irradiators. Thus, at best, the present cesium could only aid in meeting the immediate demand; and additional cobalt-60, cesium-137, and radiation generating machines would be necessary for future requirements.

STRONTIUM-90 REMOTE POWER SOURCES

Fission product strontium-90, along with its daughter product yttrium-90, is primarily a beta emitting isotope which generates 149 watts/Ci of thermal energy. This energy has been used for nearly 25 years for small remote power generators. Most have been used for defense and space purposes and some sold to foreign countries.

Strontium-90 fueled generators using thermoelectric conversion have been manufactured with an output of up to 100 watts of power and licensed by NRC. DOE now has under development a 500 watt radioisotope thermoelectric generator (RTG). This demonstration unit will be discussed in a later paper, and therefore, is not included here.

Strontium-90 has also been investigated as an energy source for Rankin, Brayton, and Stirling cycle engines. New developments in long-life high performance Stirling engines indicate that advantage can be taken of their much higher energy conversion efficiency (25 to 35 percent compared to the 5 to 8 percent for RTG's) in many applications. Work at PNL⁴ has evaluated the potential for high efficiency radioisotope power sources. They are currently designing and constructing a strontium-90 fueled heat source to power a 1 kW Stirling engine to evaluate and demonstrate the potential for remote power generation. This design incorporates a lead gamma shield and strontium-90 in WESF strontium capsules into a unit that would weigh less than 3000 kg, and use about one third the strontium-90 required for a 500 We RTG. Thus, the existing limited supply of encapsulated strontium-90 would be more effectively used.

Strontium-90 Supply

Currently there are 80 kW of usable strontium-90 encapsulated in WESF capsules stored at Hanford.¹ Successful performance of isotope power generation would increase the demand for strontium-90 and stimulate the desirability of continued recovery of strontium at Hanford and other defense production facilities and even from power reactor fuels. About 850 kW of

strontium-90 are contained or will be produced by the year 2000 in defense reactors. Spent commercial fuel will contain about 5000 kW by the year 2000.

TRITIUM POWERED RADIOLUMINESCENT (RL) LIGHTS

Tritium powered radioluminescent lights have been under development by the DOE for remote, austere, and tactical airfield lighting applications where electrical utility or portable power is unavailable or difficult to obtain. Because the technology has promise for use in both military operations and civilian applications, user organizations such as DOD, the Department of Transportation and Public Facilities of the State of Alaska, the State of Florida, and others have work co-sponsored with DOE to meet their specific objectives. In order to plan and coordinate the work, a technical working group (TWG) has been formed composed of individuals from various DOE, DOD, and civilian sponsors, and the DOE laboratories at Oak Ridge (ORNL) and Richland (PNL), participating in the work. This group is chaired by one of the authors (WCR).

Self-powered radioluminescent lights have been manufactured and demonstrated for runway, taxiway, landing pad, threshold, visual approach slope indicator, landing and marking purposes for both fixed-wing and rotary-wing aircraft during tests conducted in Alaska, North Carolina, Washington, and other locations throughout the past three years.⁵

The lights can be designed to be transportable, rugged, and easily installed. They can be used to locate drop zones, to light remote or temporary runways to rapidly repair tactical runways, and were recently used in highly successful military tactical operations during the Grenada invasion.

Their benefit in Alaska was shown New Years Eve (1984) during the Alaskan testing when a resident of Central, Alaska was badly burned in a house fire and had to be evacuated in a severe snowstorm. The fact that the Central, Alaska airstrip was lighted with a reliable lighting system--radioluminescent lights--is credited with saving his life.

RL lights appear to be especially suited for use in rural Alaska where there are over 700 military and civilian airfields. Most of these are for fixed-wing, land-based aircraft but some are used for rotary-wing aircraft and seaplanes. Only 10 percent of these runways have lighting. The Arctic and sub-Arctic environments of Alaska place special demands on runway lighting systems. Adverse weather conditions of precipitation, wind, and extremely low temperatures occur year round and cause conventional lighting equipment breakdowns and frequent maintenance. During the winter, there are few hours of daylight in which to make repairs; the cold weather hampers these efforts.⁶

Further testing and demonstration for Air Force use is planned for Germany during April and May of this year. An airfield lighting system has been shipped to Germany for the Salty Demo 85 exercises where it will be evaluated as a part of the rapid runway repair systems, which will keep our military air ports operating after a first strike. In addition, further evaluations are planned in Alaska during the 1985-1986 winter in cooperation with the FAA to obtain appropriate approvals for use of the lights for air taxi operations in Alaska.

SUMMARY

Significant progress has been made in the development of useful applications of byproducts for beneficial use. The current demand for cesium-137 exceeds the availability and the demand for radiation sources is expected to increase markedly with the onset of irradiation of food.

Tritium powered lights now have demonstrated utility in defense applications and a permanent runway is expected to be installed on a rural airfield in Alaska this summer. We expect this technology to be transferred to commercial suppliers in the next few years.

The limited supply of strontium-90 is the major deterrent to expanded use in the many applications for remote power generators.

The removal of the most highly radioactive and heat generating isotopes may prove to be a major benefit to the handling and ultimate disposal of nuclear waste.

Clearly there is a need for continued and expanding work on both recovery and uses of byproducts from nuclear reactors. We at DOE hope to continue to play a major role in this effort.

REFERENCES

1. U.S. DEPARTMENT OF ENERGY. "Department of Energy Plan for Recovery and Utilization of Nuclear By-products from Defense Wastes--Executive Summary." DOE/DP-0013, Vol. 1. (1983).
2. "The Status of the Technical Infrastructure to Support Domestic Food Irradiation." Hearing before the Subcommittee on Energy Research and Production of the Committee on Science and Technology House of Representatives Ninety-Eighth Congress, Second Session. No. 122. (1984).
3. TINGEY, G. L., E. J. WHEELWRIGHT, and J. M. LYTLE. "A Review of Safety Issues that Pertain to the Use of WESF Cesium Chloride Capsules in an Irradiator." PNL-5170. Pacific Northwest Laboratory, Richland, Washington. (1984).
4. MC ELROY, J. L. and J. A. POWELL. "Nuclear Waste Management Semiannual Progress Report April 1983 Through September 1983." PNL-4250-4. Pacific Northwest Laboratory, Richland, Washington. (1984).
5. HAFF, K. W., J. A. TOMPKINS, L. J. HULT, and C. L. BUPP. "Evaluation of Arctic Test of Improved Tritium Radioluminescent Lighting." ESL-TR-84-19. Engineering & Services Laboratory, Air Force Engineering & Services Center, Tyndall Air Force Base, Florida. (1984).
6. JENSEN, G. A. and L. E. LEONARD. "Radioluminescent Lighting for Alaskan Runway Lighting and Marking." PNL-5328. Pacific Northwest Laboratory, Richland, Washington. (1985).