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Radiochemical Processing--Off-Site Transportation and Ultimate Storage Problems

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ABSTRACT

Safe and economic methods of handling radioactive materials "off-site" are required for the successful operation of nuclear chemical plants. These occasions arise in the shipment of spent fuel, radiactive, isotopes, and liquid wastes. An unsolved problem exists in the development of techniques and sites for the final disposal of waste products.

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RADIOCHEMICAL PROCESSING—OFF-SITE TRANSPORTATION AND ULTIMATE STORAGE PROBLEMS

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INTRODUCTION

The operation of nuclear chemical plants imposes a number of requirements for off-site handling of radioactive materials. Paramount among these is the shipment to the plant of spent fuel from the reactors. In addition to these shipments which are required for routine operation, shipments of fission products and other especially prepared isotopes for commercial use is required. Finally, a potential requirement for off-site handling in the future is the shipment to permanent disposal sites of high-activity wastes which are currently stored on an interim basis at the processing plant. While no compromise can be made with safety, the most economical means of achieving safe shipment and disposal must be found if reactor-produced power is to be competitive with power from other sources.

This paper is divided into two parts. The first part reviews the techniques and problems related to shipment of radioactive materials. The research and development related to ultimate disposal methods for radioactive wastes are reviewed in the second part.

Part 1. TRANSPORTATION OF RADIOACTIVE MATERIALS

Transportation of radioactive materials by rail, motor truck, air, or water is controlled by the Code of Federal Regulations (CFR), the ICC Regulations (Table 1), and, in some cases, state and local regulations. The regulations cover allowable quantities to be shipped, safety precautions, fabrication of carriers, necessary packaging, labeling, and numerous other details and specifications that must be met, and should be studied prior to design, fabrication, or shipment of carriers. Radioactive materials now being shipped include low-activity material, e.g., unirradiated fuel elements, radioactive isotopes, and ores (covered by Title 49 CFR); and high-activity materials, e.g., spent fuel elements (covered by Title 10 CFR) and some radioactive wastes (covered by Title 49 CFR and ICC Regulation Tariff 10).

Radioactive Isotopes

In the shipment of radioactive isotopes the activity levels and energy release are generally so low that radiation shielding and heat dissipation do not constitute difficult problems. In many cases the container packing is sufficient shielding; in the most severe cases a few inches of lead may be required.

Table 1. Some of the Regulations on Shipment of Radioactive Materials

Federal Regulations*	Equivalent ICC Regulations
Title 10 Part 72 CFR (proposed) (spent fuel)	Tariff No. 6 (water shipments) H. A. Campbell, agent 30 Vessey St., New York, N. Y.
Title 49 Parts 71-78 CFR (dangerous articles)	Tariff No. 8 (truck shipments) F. G. Freund, agent 1424 16th St., N. W., Washington 6, D. C.
Title 14 Part 49 CFR (by aircraft)	Tariff No. 9 (rail shipment) H. A. Campbell, agent
Title 49 Part 146 CFR (over water)	Tariff No. 10 (radioactive materials) H. A. Campbell, agent
U. S. Postal Guide (through mails)	
Civil Air Regulations (explosives and other dangerous articles)	
Handbook of Federal Regulations (radioactive materials.	

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*These documents are on sale by the Superindent of Documents, Government Printing Office, Washington, D. C.

Isotope shipments from ORNL are routine, as indicated by the following summary of shipments made during the ten-month period, July 1959 through April 1960.

Major Categories	No. of Shipments	Curies
Co-60	47	124,000
Cs-137	194	78,700
H-3	117	14,600
I-131	2,153	540
P-32	1,776	130
C-14	244	14
Other	377	13,700

Spent Fuel Shipments

Spent fuel is shipped from the reactor site to the processing site for recovery of fissionable and fertile material. The shipping containers must be designed with provision for adequate heat removal, shielding, and safeguards against accidental formation of a critical array. In addition, the ease of decontaminating the surfaces of the cask must be taken into consideration when specifying materials and details of construction.

The relative sizes, weights, and geometries of some first generation reactor fuels that will have to be shipped from reactor sites to processing plants are shown in Fig. 1. The ORNL Graphite Reactor slug and the MTR assembly are typical fuels routinely shipped in the past; so it is apparent that a significant extension in handling and shipping techniques will be required for power reactor fuels. In addition, these fuels will have been subjected to higher nuclear burnup than fuels from current research reactors and will consequently generate proportionately more fission product decay heat.

Heat Removal: Specifications in Title 10 CFR pertaining to heat removal stipulate that the hottest spot in the fuel element or built-in poison must be at least 180°F below its respective melting point, assuming a dry, air-filled cask, with natural convection cooling. In cases where forced convection cooling is used, the primary gas or liquid coolants must not be circulated outside the carrier in order to safeguard against exposure in the event of contamination with fission products, and any liquid coolants must be maintained at least 20°F below their boiling points. The system should operate at approximately atmospheric pressure with the temperature of the external accessible surface of the carrier not exceeding 180°F.

In conforming with these regulations, practice has been to restrict the amount of heat evolved in a carrier to that quantity which can be dissipated by natural convection and radiation , from the carrier surface, and this can be done through decay cooling prior to shipment and/or by restricting the number of fuel elements per carrier. Forced convection cooling within the carrier has not been economic because of the increased complexity and additional weight of shielding required. Safety could also be compromised due to the difficulty in guaranteeing that the system will not develop a leak or that there will be no power failure.

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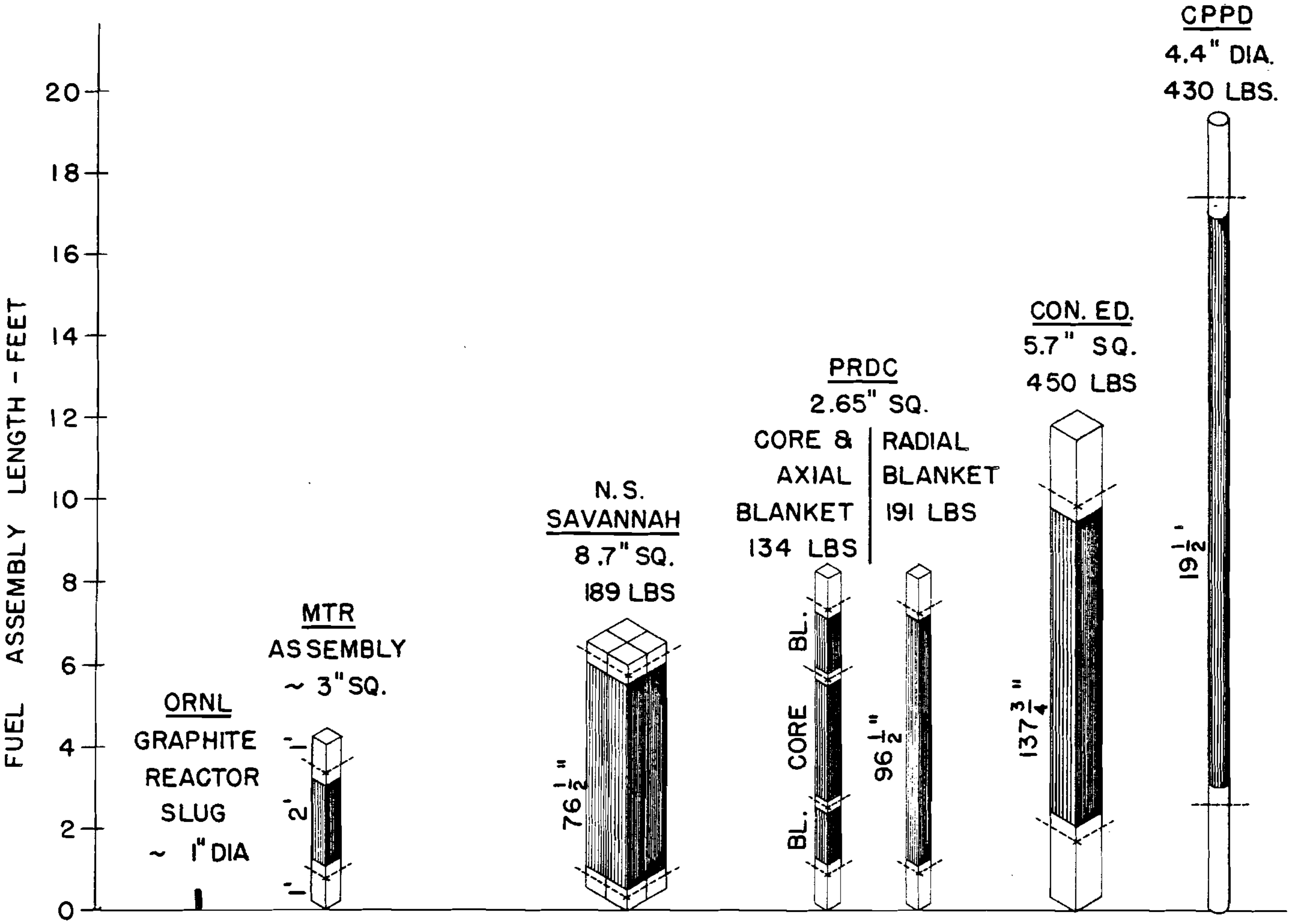

Fig. 1. Relative sizes and weights of power reactor fuel assemblies. Dotted lines indicate portions that could be removed prior to shipment, and dark portions indicate fuel-bearing sections. (1)

Calculations have been made to determine the heat that could be dissipated by radiation and natural convection from the surface of a 54-in.-o.d. carrier containing Consolidated Edison elements, irradiated to 23,000 Mwd/tonne burnup (Fig. 2). A carrier of this size could provide space for 12 elements; but the results show that if the cask surface temperature is to be kept at 180°F, no more than three elements, 12 months' decayed, can be carried. In this example it is apparent that the cost of providing better heat transfer must be balanced against the higher fuel inventory charges associated with longer decay times and the higher costs required to ship fewer elements per carrier. It seems likely that there will be sufficient economic incentive to justify the development of carriers equipped for mechanical removal of heat.

Shielding and Structural Integrity: For safe shipment of radioactive materials, carriers must be designed for ample biological shielding and with sufficient structural strength to prevent rupture in the event of any credible accident. Title 10, CFR specifies that the external radiation levels must not exceed 200 mr/hr at any accessible surface of the cask or more than 10 mr/hr at a distance of 1 meter from the cask except when shipped by rail, in which case the latter limit is extended to 10 mr/hr at 3 meters. It is further specified that the structural integrity of the carrier be such that it will remain intact after striking solid concrete while traveling 44 ft/sec (a 30-ft drop), and that the fuel elements and poisons will not rearrange themselves in a more nuclearly reactive configuration.

Because of its reasonable cost, high density, and easy fabrication, lead has been used almost exclusively in this country as a carrier shielding material. However, its low melting point and poor structural properties leave much to be desired. Other possible shielding materials include cast iron, steel, and depleted uranium. For a comparable sized cavity and equal shield effectiveness, a cast iron or steel carrier may weigh as much as 15% more than a lead-filled carrier and occupy 70% greater space. In many applications this would not be too great a penalty to pay for the greater structural strength and better thermal properties possessed by iron and steel provided fabrication costs could be reduced to a level competitive with those of lead.

In addition to possessing reasonably good structural and thermal properties, depleted uranium as a shielding material offers the advantage of requiring the minimum mass of shielding for cavities of any given size and geometry. The

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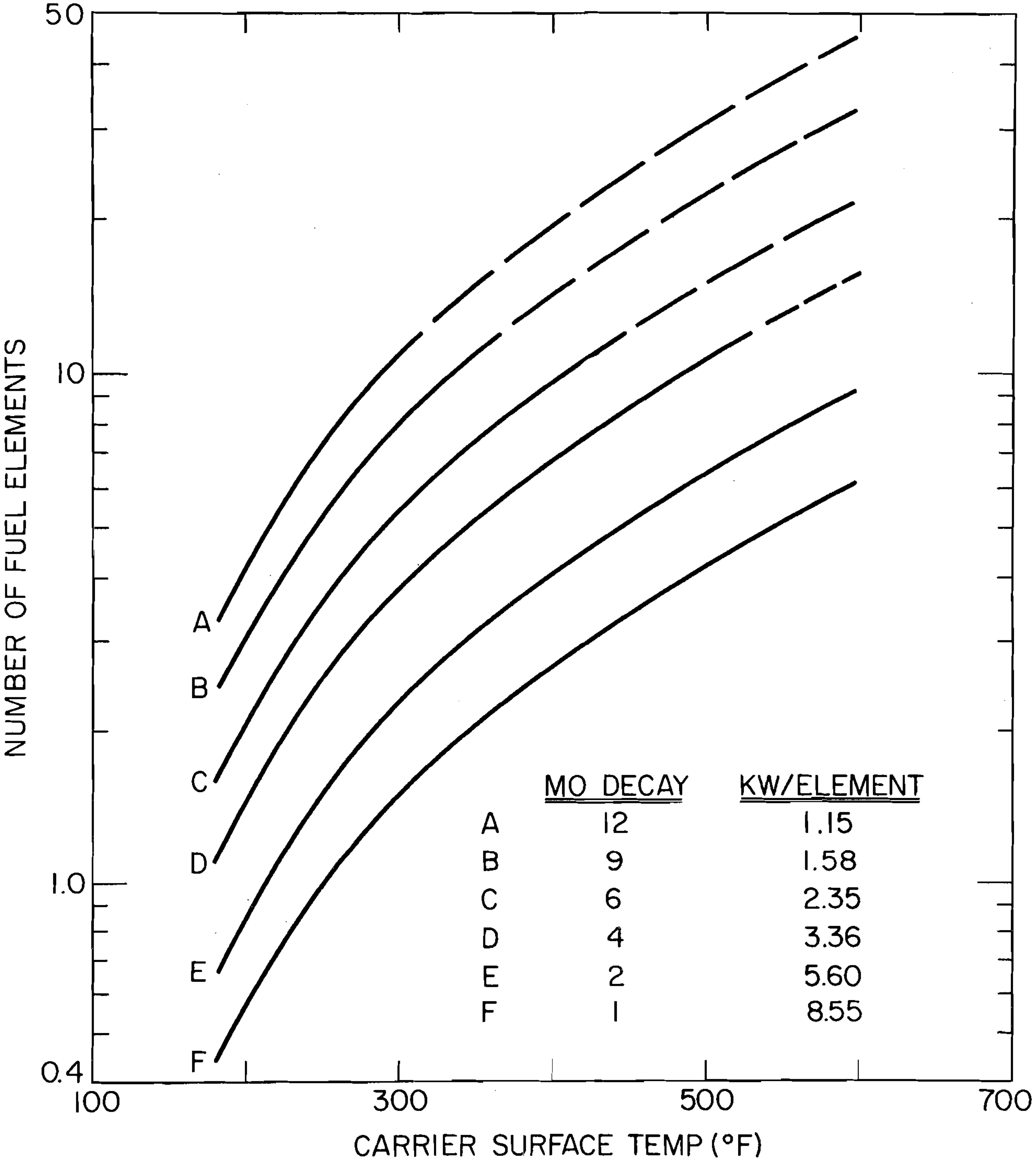

Fig. 2. Carrier surface temperature as a function of number and time of decay of average Consolidated Edison fuel elements. Basis: Fuel Irradiated to 23,000 Mwd/tonne and shipped in 54-in.-o.d. carrier with ambient temperature at 100°F.

saving in weight through using uranium rather than lead can range as high as 60% for shielding small cavities several cubic centimeters in volume but drops to less than 10% savings in weight for cavities over 100 ft^2. The major drawback in the use of uranium is in fabrication problems and raw material costs.

Criticality: In shipping the maximum number of fuel assemblies per cask consistent with shielding and heat removal, provision must frequently be made to avoid conditions that could lead to a criticality incident. Criticality can be prevented by limiting the mass (or concentration) of material, controlling the geometry, or using nuclear poisons. Title 10 CFR, Part 72, stipulates that shipment of up to 75% of a critical mass may be shipped in a carrier without special provisions for criticality, but any shipment exceeding this amount must be made in carriers with the fuel spaced in geometrically safe configurations. Normal structural materials must be ignored as nuclear poisons in these considerations, but specific poisons such as boron or cadmium may be built into the carriers as lattice or spacers, or taken into account when present in the fuel elements themselves.

The minimum single critical batches, completely reflected, of various materials are (2)

U^{235}	0.7 kg
U^{233}	0.5 kg
Pu^{239}	0.5 kg
20% enriched U^{235}	0.96 kg of U^{235}
5% enriched U^{235}	1.6 kg of U^{235}

Since most individual fuel elements in the power reactor program contain more than a critical mass, shipments must normally be made with criticality control.

Shipment of Liquids

Because of the ease with which they can be dispersed, radioactive solutions are inherently more hazardous to ship than are comparable amounts of radioactivity in solid form. Proportionately greater care must be taken to limit temperature and pressure rises within the cask and to prevent leakage in the event of accident. Specifications pertaining to liquid shipments are detailed only regarding the handling of relatively small quantities of solution typical of the isotope sales program. Shipments of larger quantities of solutions have been relatively rare and have been considered individually as special cases. The largest operation to date consists of approximately 30 shipments of 3- to 5-year-old waste by rail freight from the Idaho Chemical Processing Plant to Oak Ridge National Laboratory for fission product recovery. Shipments were made in 500-gal cylindrical tanks shielded with 3.5 in. of lead (3). The waste averaged about 10 beta curies per gallon in activity and did not require special provisions for cooling while in transit or for criticality control.

Economics and Safety Considerations

As is the case for other parts of the nuclear fuel cycle, it has not been possible to assign a definite limit to shipping costs allowable in a competitive nuclear power economy; however, in an 8- to 10-mill economy, Guthrie (4) has suggested that these charges should probably not exceed 0.2 mill/kwh$_e$. In the case of a reactor operating at 25% thermal efficiency, a fuel burnup of 8,000 megawatt days/ton, and an 80% load factor, this would be equivalent to $10 per kilogram of low-enriched fuel. The total shipping costs are primarily a function of the capital costs of the casks, the transportation charges, and the contingency or liability costs. The capital costs may vary from $0.25 to more than $1.00 per pound for current carriers and are interrelated to a considerable degree with liability costs since additional safety or integrity can seldom be built into a cask without increasing its initial cost. On the other hand, it appears that additional safety features can be adopted without greatly affecting the over-all economics when it is realized that, in the case of a carrier in use 50% of the time, the transportation costs alone over five years will exceed the initial capital investment by a factor of 4 or 5.

Transportation costs by different modes of conveyance as a function of distance from Oak Ridge, Tennessee, are presented in Fig. 3. Rail express is several times more expensive than either motor or rail freight; however, decontaminated, empty carriers can be returned via rail express at about 65% of first class rates. Barges have not been used for transportation of radioactive materials pending an analysis of the hazards involved, but they appear to be especially attractive for very heavy shipments. As a general rule, anything weighing over 25 lb is usually shipped by rail or motor freight as opposed to the more expensive rail express. Motor freight can handle 15 to 20 tons routinely and above 20 tons using special equipment. For casks with a gross weight in excess of 40 tons, rail shipment or possibly barges are the only practicable methods.

Very little experience is available on which to base liability costs. Knapp (5) has attempted to estimate the order of magnitude of the costs that

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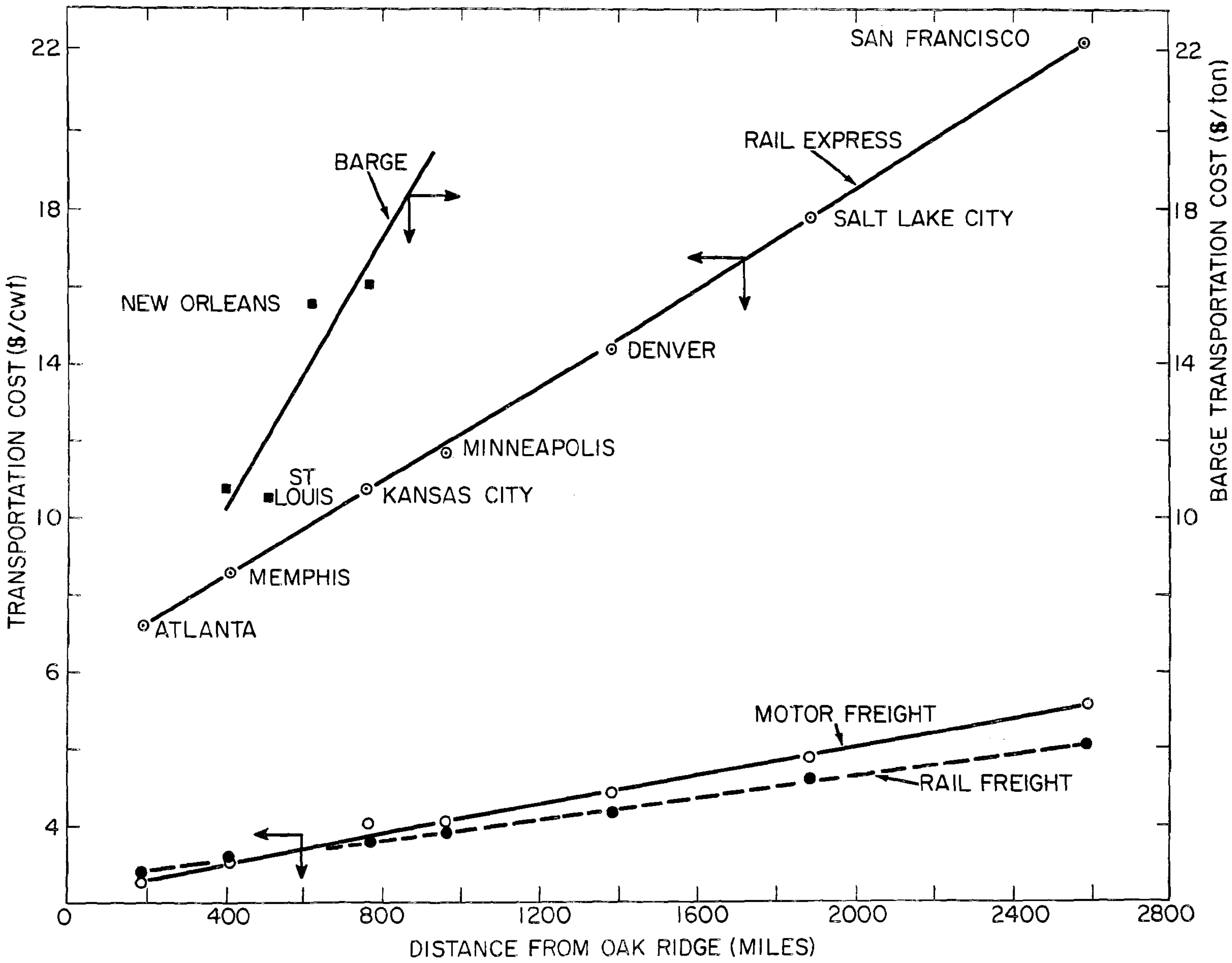

Fig. 3. Transportation costs as a function of distance for various modes of conveyance.

should be placed on 1,000-mile shipments to cover the liability incurred from a serious contamination accident. By gathering information on the frequency and severity of accidents, it was determined that one might expect a serious accident every 100,000,000 vehicle miles for ordinary methods of transport. A contingency cost for a $5,000,000 accident was computed to range between 1 and 4% of the basic transportation cost, which suggests that insurance premiums for such shipments should not be prohibitively expensive.

Part II: ULTIMATE DISPOSAL PROBLEMS

Although great quantities of radioactive waste are produced as a result of nuclear chemical plant operations, it has been routine practice to store or dispose of them on-site without the necessity of off-site handling or shipment. Very dilute gaseous and liquid wastes are released to the environment through stacks or surface waterways, seeps, and lagoons. Solid or packaged wastes, usually containing minor quantities of activity, are buried in carefully chosen areas above the ground water table. The high-activity liquid wastes containing concentrations of radioactive materials greatly in excess of the levels considered safe for release are stored in tanks buried near the surface of the ground.

While it is likely that the growth of a nuclear power economy and an expanding population over the remainder of this century will result in requirements for additional cleanup and decontamination of low-level wastes prior to their release, the permanent disposal of high-level wastes represents a major unsolved problem. Tank storage of these solutions is considered to be no more than a temporary expedient, for the presence of biologically hazardous isotopes of long radioactive half-life imposes the requirement that the wastes be isolated from mankind and his immediate environment for many centuries. At the present time about 67,000,000 gallons of high-level waste is in storage at various AEC sites. In addition to continued waste production at Government installations, it is estimated that the wastes generated by an expanding nuclear power industry will amount to 36,000,000 gallons in 1980 and 300,000,000 gallons by the year 2000, assuming current aqueous chemical reprocessing methods are used (6).

The composition of these wastes depends on the processing technique used and the fuel being processed. Table 2 summarizes some significant characteristics of wastes from recovery of various types of fuels by solvent extraction. The volume of waste produced per kilogram of uranium recovered is greater for

Table 2. Characteristics of Anticipated High-Activity Wastes

Fuel Type	Vol., gal/kg U	Composition	Fission Product Level,[a] per gal		
			120 days decay	1 yr decay	10 yr decay
Natural or slightly enriched U or UO_2	0.06	6 M HNO_3	60,000 curies (220 watts)	21,000 curies (68 watts)	1,850 curies (4 watts)
Highly enriched U-Al alloy	125	1.3 M HNO_3 1.6 M $Al(NO_3)_3$	1,390 curies (4.8 watts)	400 curies (1.4 watts)	30 curies (0.07 watts)
Highly enriched U-Zr alloy	450	1.1 M HNO_3 0.8 $(NH_4)_2ZrF_6$ 0.5 M NH_4F 1.1 M $Al(NO_3)_3$	390 curies (1.3 watts)	110 curies (0.4 watt)	9 curies (0.02 watt)
Highly enriched U-stainless steel cermet	65	50 g/liter SS 0.69 M $Fe(NO_3)_3$ 0.16 M $Cr(NO_3)_3$ 0.08 M $Ni(NO_3)_2$ 0.12 M $Al(NO_3)_3$ 3 M HNO_3	1,260 curies (9.2 watts)	140 curies (2.6 watts)	52 curies (0.13 watt)

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[a] Fission product levels based on 10,000 Mwd/ton burnup of slightly enriched uranium and 30% burnup of slightly enriched uranium of an average thermal neutron flux of 3 x 10^{13} n/cm^2/sec.

enriched uranium alloy fuels than for unalloyed U or UO_2 fuels because the nonradioactive alloying material remains with the waste and limits the degree of concentration obtainable. The wastes containing dissolved stainless steel, aluminum, and zirconium with fluoride and nitrate ions will be less amenable to tank storage than the nitric acid wastes because of their corrosivity and, in some cases, chemical instability. Neutralization results in the formation of voluminous precipitates which complicate the problem of removing radioactive decay heat. Although temporary storage in tanks may be used to allow for decay of short and intermediate-level activities, final repositories are required which afford greater security and permanence than do the present tanks, and possibilities for establishing sites for this purpose, both at sea and on land, have been considered. In all cases a large amount of work yet remains to be done before technical feasibility based on both safety and economics is established.

Sea Disposal

Because of the generally inadequate knowledge of the effects of radioactivity on marine resources, the oceans have been used to only a very limited extent for disposal. In this country since 1946, low-activity solid wastes arising mainly from research and industrial uses of radioisotopes, and totaling about 22,000 curies at the time of burial, have been packaged in steel and concrete containers and dumped in 1,000 or more fathoms of water at sites off both the Atlantic and Pacific coasts (7). In addition, about 1,000 curies per day of neutron-activation products reaches the estuary of the Columbia River as a result of AEC reactor operations upstream at Hanford, Washington. The British have carried the practice a step further by pumping low-activity liquid wastes, containing an average of 3,000 curies of activity per month, from chemical processing plants into the Irish Sea (8).

All these operations are carefully supervised and monitored, and studies made of them to date indicate that larger amounts could be disposed of under less exacting restrictions without resulting in harmful effects. However, there remain sharp differences of opinion both in the United States and abroad concerning the over-all safety of ocean disposal based on uncertainties in mixing, dilution, and the degree of assimilation of radioisotopes by marine organisms. Any extension of the present conservative practices seems destined to come about only as adequate programs of research and education establish the necessary degree of confidence that both human health and the marine environment are being adequately safeguarded.

Based on the controversy pertaining to low-activity wastes, it is, at best, premature to decide whether any high-activity wastes should be dumped at sea. It has been suggested that disposal of these wastes might be possible in certain very deep waters of the ocean, which are believed to remain isolated from the surface layers for periods of hundreds of years (9). Even after more is learned pertaining to the residence time, currents, and mixing processes in the deep sea, the engineering problems of waste shipment to these areas and their subsequent monitoring appear so complex as to render disposal of high-activity wastes in the ocean infeasible for many years.

Land Disposal

The possibilities for ultimate disposal on land that are currently receiving emphasis are: conversion of liquid wastes to solids and storage of the solids in selected or prepared sites; and direct discharge of liquid wastes into salt cavities or deep, permeable formations. The preparation of solids is the more conservative of these two approaches and is generally expected to be more expensive than direct disposal procedures, but at the same time it offers two distinct advantages which put it in a favored position as a long-term solution to the problem. First, immobilization of the radioisotopes in solid form would permit their shipment to ultimate disposal sites which may be remotely situated from desirable chemical plant locations. It is likely that the hazards of routinely shipping high-activity liquid wastes would be intolerable; consequently, direct disposal of them would be possible only if the chemical plant should be located at the disposal site. Secondly, the solidified wastes afford easier surveillance and the possibility for recovery if failure of the disposal site should occur.

Converstion to Solids: Processes for converting high-activity wastes to solids all have as their objective the production of thermally stable, chemically inert solids of high bulk density and thermal conductivity. The degree to which these properties are achieved will influence the manner and cost of their storage. At Brookhaven National Laboratory (10) and Chalk River (11) emphasis is placed on fixation of the radioisotopes in solids of very low solubility. In the first case, the waste solution is passed over extruded clay which sorbs the radioisotopes, and these sorbed activities are subsequently "fixed" by firing at about 900°C. In the Canadian process, waste is mixed with nepheline-syenite and heated to 1,350°C to form a glass. Other processes are designed to reduce the bulk wastes to dryness and calcine the residual solids in the absence of ingredients added specifically to achieve fixation. For this purpose a fluidized bed calciner is being studied at Idaho Chemical Processing Plant (12), a pot calciner at Oak Ridge National Laboratory (13), a radiant heat spray calciner at Hanford Atomic Products Operation (14), and a rotary ball kiln at Brookhaven National Laboratory (15). To compensate for the higher leachability and in some cases lower structural stability of these solids, greater reliance would necessarily be placed on the long-term integrity of their primary containers.

To date, none of these processes has been demonstrated on a pilot plant scale with high-activity wastes, although the fluidized bed calciner is currently undergoing such tests in Idaho. It seems likely that several of the processes will find eventual application, depending on their economics, the types and volumes of wastes to be processed, and the type of permanent storage to be used.

Possibilities for permanent storage of the solidified wastes have been seen to exist in specially constructed concrete vaults and in certain natural geologic formations such as limestone and salt. Because of a lack of information concerning the nature of the solids to be stored and many of the geological and environmental factors involved, it has been possible to define only in a general sense the criteria for a suitable ultimate disposal site. Besides the high degree of permanence and isolation that is required, the site should be accessible to commercial shipping and transportation facilities; it must be accessible to men and the remote equipment necessary for handling or arranging the wastes in place; it must be possible to provide for the natural dissipation of the heat of radioactive decay; and, finally, it should provide a dry environment where long-term corrosion of the waste containers would be held to a minimum. As is noted below, salt deposits meet these and other considerations to a unique degree and are therefore considered a prime possibility for disposal of solid wastes. On the other hand, mined repositories in shale or limestone formation might be developed to serve the needs of specific sites where salt is unavailable.

Direct Disposal: Under the auspices of the National Academy of Sciences-National Research Council, a committee of geologists and geophysicists was established to consider the possibilities for disposal of high-activity wastes in the United States. This committee concluded that disposal in salt was the most promising method for the near future and that storage in deep porous beds and sedimentary rocks by means of deep wells was a good possibility for the more distant future (16).

Large deposits of salt exist in many well-defined and accessible locations in the United States, and commercial mining operations create annually spaces that are greatly in excess of the expected volumes of high-activity waste production at the end of this century. In addition to offering an isolated and relatively uniform chemical and mineralogical environment, salt is plastic under load, and deposits are impervious to water. At the same time, the compressive strength is great enough to allow cavities to be mined which are structurally safe and accessible to personnel and equipment. Considerable work has been done at the University of Texas (17) and Oak Ridge National Laboratory (18) on such problems as the chemical interactions between salt and typical waste solutions, dissipation of radioactive decay heat, and structural properties of salt. These studies have led to the initiation of a series of field experiments now being conducted in portions of a Carey Salt Company mine near Hutchinson, Kansas. The experiments, which are being conducted with non-radioactive synthetic waste solutions, are designed to investigate the structural integrity of salt cavities; the migration of radioisotopes from cavities; the migration of the cavity itself due to condensation of water and dissolution of salt; temperature gradients around the cavities; and production of gases by chemical reactions between the salt and wastes. It is expected that the field experiments will establish the conditions under which liquid wastes could be safely stored in salt, following which an economic analysis should enable its practicability to be assessed. Because of the problems associated with decay heat removal and off-gas production during storage, it seems likely that salt can best be applied to disposal of high-activity solid and low-activity liquid wastes.

The feasibility of disposal into subterranean porous formations through deep wells is suggested by the technology already established by the petroleum industry for disposal of brines and for the secondary recovery of oil. A subcommittee appointed by the American Petroleum Institute has reviewed the problems pertaining to this method of disposal and confirmed its possible application (19). The problems are rather more complex than those related to disposal in salt and can be expected to take longer to resolve. Relatively little is known geologically concerning the very deep porous formations such as might be suitable. They will have to be located and defined carefully. A very thorough chemical and mineralogical knowledge of the disposal reservoir will be required before chemical compatibility between the waste and residual liquids and solids can be assured. A careful analysis of the thermal problems incurred by radioactive decay heat must be made. Adsorption of radioisotopes on the solids concentrated over a relatively small area could cause "hot spots" and result in undesirable effects. Other problems may arise from corrosion of the well casing by the wastes and the possibility of plugging the well with suspended solids. Laboratory investigations have been initiated on the interactions of waste solutions with typical clay minerals (20), preliminary computations of the magnitude of the heat problem have been made (21), and surveys of existing information on the geology of potential disposal sites are under way. If this work continues to support the feasibility of deep-well disposal, it is expected that exploration and field testing would consitute the next phase of development. The promise of considerable pre-injection treatment that may be required and the irreversible nature of this disposal method render its eventual use questionable.

Economic Considerations

The fraction of nuclear power costs allowable to waste disposal has not been established, and it is difficult to form a realistic estimate of the eventual costs of ultimate disposal methods now under consideration because of the preliminary status of their development. However, the cost of storing wastes in tanks of present design on a "perpetual care" basis has been estimated to lie between 0.1 and 0.15 mill/kwh of nuclear electricity produced (22). This corresponds to only 1 to 2% of the cost of power in an 8 to 10 mill/kwh economy and cannot be considered unreasonable. Estimates indicate costs for ultimate disposal by methods currently under study should fall in the same range, and if later developments bear out these expectations, waste disposal should not constitute a barrier to the development of economically competitive nuclear power.

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