

DISPOSAL OF ATOMIC POWER PLANT WASTES

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THE discussion will be in two parts, the first dealing with a description of the Yankee Atomic Power Reactor and the waste problems associated with such a reactor; the second with a brief summary of present practices and research developments in the disposal of radioactive waste materials.

YANKEE ATOMIC POWER REACTOR, ROWE, MASSACHUSETTS**

The plant site is located in the town of Rowe, Massachusetts on the east bank of the Deerfield River at a point approximately three-quarters of a mile south of the Vermont-Massachusetts border adjacent to the Sherman hydroelectric station of the New England Power Company. This location offers advantages to Yankee in cooling water supply, transmission economy, and a favorable land area. According to the 1950 census 28,892 people live within 10 miles of the plant and 104,293 (including North Adams, Massachusetts) live within 20 miles. The basic land use is agriculture and forest products with light industry in Adams and North Adams. No water supplies are listed for the 42 mile reach of the Deerfield River to its confluence with the Connecticut River. Meteorological data interpolated from nearby Weather Bureau stations generally indicate the significance of rugged topography in affecting seasonal turbulence and inversions in site-area valleys. Additional meteorological studies are underway at the site.

The plant will be similar to a standard steam-electric station except for the boiler equipment. In the Yankee plant the conventional boiler will be replaced by a pressurized water reactor contained

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**The information in this section has been taken from the following documents: (1) Anonymous, "Pressurized Water Reactor for New England—A New American Project," *Atoms*, Vol. 7, pp. 440-442 and 447, December, 1956; (2) A brochure prepared by the Yankee Atomic Electric company, describing the plant to be constructed at Rowe; (3) Statements contained in the 19th and 20th Semi-Annual Reports of the Atomic Energy Commission to Congress pertaining to the Yankee Reactor; and (4) News releases contained in the February, March, May, July and October, 1956, issues of *Nucleonics*.

in a large steel sphere. This is the general type of reactor being built at Shippingsport, Pennsylvania by Westinghouse for the Duquesne Power and Light Company, and similar to the unit powering the Nautilus.

The orthodox section of the plant will house a 134,000-kilowatt tandem compound turbine with its condenser and auxiliary equipment. Adjacent to the conventional buildings will be the 125-foot steel sphere known as the vapor container. Inside this steel shell is the actual reactor steam generator with its controls, auxiliaries, and heat exchangers. The heart of the reactor is the core which is cylindrical in shape, about 6 feet in diameter and approximately 8 feet tall. This will stand in a large water filled pressure vessel about 9 feet in diameter and some 30 feet high. Its steel walls will be over 8 inches thick and it will weigh about 175 tons.

Cooling water will enter the top of the vessel from the four coolant pumps, flow downward through the thermal shields, and make a single pass upward through the core heating channels. No penetrations of the reactor vessel are planned below the top of the core, thereby making loss of water covering in the core impossible, except for vessel rupture. Outside the pressure vessel wall is additional radiation shielding consisting of a water-filled thermal shield tank and approximately 8 feet of concrete wall. The reactor core consists of four similar quadrants, each containing 19 fuel assemblies for a total of 76. These assemblies will be made up of over 20,000 stainless steel tubes. Each vertical 8-foot long pencil-sized tube is filled with about 300 pencil eraser-sized pellets of slightly enriched uranium oxide. Some 24 control rods will be located to move up or down inside the pattern of tube assemblies to control the neutron reaction.

Water in the pressure vessel will be heated to 560°F. under 2,000 psi and will return from the heat exchanger at 510°F.; the secondary will make 600 psi steam. High purity for the light water coolant-moderator will be maintained in order to ensure the smallest possible release of activated particles to the waste disposal system. The all stainless steel construction will ensure that the corrosion rate is low, and a hydrogen gas corrosion inhibitor will be used to further reduce corrosion. Furthermore, the use of oxide fuel and the anticipation of a small amount of clad failures will result in only small releases of fission products from the uranium core material.

A purification system consisting of demineralizers will be de-

signed to control loop water purity and to remove activated particles. The demineralizers will be of the "regenerated in place" or "throw away" type based on plant economics and technology of handling waste disposal.

PROTECTIVE EQUIPMENT

Protective equipment and facilities will be provided at the plant to safeguard physical property and for personnel protection. Equipment connected with personnel protection against radioactivity includes the vapor container, radiation shielding, and radiation monitoring.

The design of the vapor container will be such that it will contain the pressure buildup resulting from a maximum credible accident to this particular pressurized water reactor plant. Considering the oxide fuel to be used and the stainless steel cladding, the maximum credible accident has been tentatively chosen as being the pressure buildup resulting from a rupture and the release of the entire volume of the primary loop at average operating temperature.

Radiation shielding during normal plant operation will include a thermal shield tank and the necessary concrete shield to reduce the dose levels at the surface of the vapor container to about 6 mr/hr at full power. The radiation level at continuously manned stations outside the container will be designed below 2 mr/hr at full power. Radiation monitoring will be provided throughout the plant, and in the area surrounding the plant which makes up the site. Plant monitoring will include airborne particle detectors, boiler leak detectors, area monitors, neutron detectors, and gamma detectors.

FUEL HANDLING AND SPENT FUEL STORAGE

Other equipment is that associated with the fuel handling and spent fuel storage. Suitable manipulators, shielding, and underwater storage facilities will be provided to remove and allow spent fuel to decay for the period required prior to shipment in air-cooled container casks. No decladding or reprocessing activities are planned at the facility.

WASTE DISPOSAL SYSTEM

It is reported that a waste disposal system for the temporary holdup and ultimate disposal of activated primary loop and fission product solubles, insolubles, and gases resulting from normal oper-

ation or operation with clad failure will be provided. The design will be such that no waste will be discharged from the plant above the permissible levels outlined in National Bureau of Standards Handbook 52. The waste disposal system will include cartridge demineralizers and gas absorbing filters and possible evaporators for the concentration of the waste, such that ultimate disposal in solid form can be accomplished if required. Large volume holdup of liquid waste will be provided by storage tanks. The possibility of plant operation with clad failures will depend on further research on the uranium oxide fuel materials and the adequacy of the waste disposal system.

WASTE DISPOSAL FACILITIES, SHIPPINGSPOINT, PENNSYLVANIA*

Although the Yankee reactor is larger than the Shippingspoint pressurized water reactor, having a power level of 500 megawatts heat energy of 134 megawatts electrical energy in contrast to 264 and 60 megawatts, respectively, the waste disposal facilities provided at Shippingspoint may be considered as a basis for discussing the waste problem at Yankee. Obviously, there will be differences in the quantities of radioactive wastes produced because of the difference in power levels, but the sources of wastes will be similar. Another difference will result from the types of fuel elements employed; zirconium-enriched uranium, Zircaloy-2 clad elements will be used at Shippingspoint whereas uranium oxide pellets clad in stainless steel will be used at Yankee.

The wastes produced at Shippingspoint have been categorized into eight types as follows:

Type I. Reactor plant systems spent resins. Spent resin from the mineralizers will be transferred by flushing, in the form of a slurry, directly to underground storage facilities.

Type II. Cold laundry and monitored drains.

Type III. Hot laundry and special monitored drains.

Type IV. Decontamination room waste.

Types II, III, and IV constitute low level wastes, which are processed by holding in service building hold tanks, monitoring and

*This information was given to the writer by Mr. E. D. Harward, Public Health Service, on assignment to the Shippingspoint Reactor Project, and by Mr. J. G. Terrill, Jr., Chief, Radiological Health Program, Public Health Service, Washington, D. C., and was taken from the paper, "A Sanitary Engineering Approach to Reactor Waste Disposal," by J. G. Terrill, Jr., and M. D. Hollis, which was presented at the Annual Meeting, American Society of Civil Engineers, October 17, 1956, Pittsburgh, Pennsylvania (5).

release of each into a pair of tanks in the low level waste storage area. These wastes will be evaporated, the evaporator bottoms being discharged to the spent resin storage tanks or packaged for disposal at sea. The vapor condensate will pass to the surge and decay tanks before release into the Ohio River following mixing with condenser stream flow.

Type V. Reactor plant effluent. A portion of the cooling water will be wasted and discharged to the surge and decay tanks for hold-up. The liquid fraction will be passed through ion exchange resins and will feed into the spray recycle tank or into the gas stripper. From the gas stripper the liquid will flow to test tanks where it will be monitored prior to mixing with condenser water and release to the Ohio River. The gases contained in the reactor plant effluent discharge to the vent gas surge drum and into the gas decay drums before release into the atmosphere following monitoring.

Type VI. Combustible waste. This consists of laboratory wipes, contaminated clothing, other contaminated combustible material. It will be burned in an incinerator with the gases passing through a wet gas scrubber before release through the stack, the ash being slurried for disposal in the spent resin storage tanks.

Type VII. Noncombustible wastes such as tools, metal turnings, etc., will be packaged, shielded, and shipped via railroad or truck to a point for disposal at sea.

Type VIII. Noncombustible items too heavy to ship will be disposed of by burial at the site.

In all probability many of the same techniques will be utilized in handling the various wastes produced during the operation of the Yankee atomic power plant.

WASTE DISPOSAL IN THE ATOMIC ENERGY PROGRAM

Waste materials from the atomic energy industry are gaseous, airborne, liquid and solid and occur in any phase of the industry from the mining of the uranium ore to the ultimate use of a specific radioisotope in industry, research, or medicine. These waste materials differ from those with which we have been concerned in the past in that they are radioactive. Unless properly controlled, they could be damaging to human and other tissues.

GASEOUS AND OTHER AIRBORNE WASTES

Gaseous or airborne particulate wastes vary greatly with their origins, and include tiny particles of radioactive material originating from failure of a fuel element in an air-cooled reactor, particulates and iodine from fuel processing plants, and particulates from plutonium fabrication facilities (6). Many of these problems have been solved through the development and use of special high-efficiency filters and iodine gas removal units.

Air used as a coolant for a reactor is prefiltered to remove particulates which would become radioactive when irradiated. High-efficiency filters of glass or kraft paper and asbestos are also used to remove radioactive particulates from gas that has passed through a reactor. Short-lived radioactive isotopes of gases, such as iodine, in the waste streams from chemical processing plants can be released to the atmosphere through dilution from a tall stack under favorable meteorological conditions.

Studies in micrometeorology have shown that a wide variety of conditions will affect the dispersal of stack discharges, and have indicated what hazards could arise should serious disruptions occur in normal operations involving radioactivity.

SOLID WASTES

Solid wastes are divided into two groups: combustible and non-combustible. The latter includes such materials as machine turnings, contaminated equipment, etc., whereas the former includes burnable contaminated trash. In general, the wastes are disposed of by incineration, in the case of combustible materials, and packaging for ocean disposal or direct burial into the ground.

Burial

Five national burial sites, located at the Hanford Works, Washington; National Reactor Testing Station, Idaho; Los Alamos Scientific Laboratory, New Mexico; Oak Ridge National Laboratory, Tennessee; and Savannah River Project, Georgia, are operated by the Atomic Energy Commission for the disposal of solid wastes (7). The physical factors affecting the desirability of any particular site are topography and geology, surface and ground water hydrology, meteorology, soil conditions, and transportation facilities. From results reported by Morgan (7) the costs of operating such disposal sites ranged

from \$1.52 to \$9.40 per cu yd of material. These costs did not include sample containers, processing and handling at source, etc., but only capital and operating costs at the site, and may be compared with specific costs at given sites. At Oak Ridge, the annual burial load amounts to about 5 acres per year at a cost of approximately \$2.00/cu yd (8); at the National Reactor Testing Station (9) approximately 3800 cu yds of solid wastes have been disposed of since 1952 from Rocky Flats, Colorado, at a reported cost exclusive of packaging and handling of \$21 to \$35/cu yd (10).

Incineration

Studies to evaluate the effect of incineration waste materials containing P^{32} , Sr^{89} , and I^{131} in an institutional incinerator are described by Geyer *et al.* (11). Their results indicate that of the P^{32} charged into the incinerator, about 12% was retained on the stack wall and 2% was recovered from the stack gas; of the Sr^{89} , 9% was retained on the wall and 1% recovered in the gas; while of the I^{131} , about 11% was deposited on the wall and 80% escaped in the gas. Ash activity was assumed to be the balance of the charge not otherwise accounted for.

The use of a special type of incinerator has been described by Silverman and Dickey (12) for reduction of combustibles contaminated with low level amounts of radioisotopes. Maximum daily load is set at 200 $\mu\mu\text{c}$ for all isotopes except I^{131} which is set at 500 μc . The stack gas effluents had activity levels of 7×10^{-11} $\mu\text{c}/\text{ml}$. Maintenance costs over a two-year period of operation, during which 3400 lbs of assorted combustibles containing 27,100 μc of activity were handled, amounted to less than \$1/month.

Rodgers and Hampson (13) report operating data on the incinerator designed and built at Argonne National Laboratory to handle 100 feet³ of combustibles daily. Overall decontamination factor (ratio of influent feed to effluent gas concentration) was 2.3×10^7 . Cost of incineration amounted to \$2.68/ft³ for 8-hour and \$1.60/ft³ for 24-hour operation schedules as compared with solid storage costs of \$9.00/ft³. At present the combustibles are being shipped to Oak Ridge for burial at a cost of \$1.50/ft³ thus removing the economic justification for incineration.

LIQUID WASTES

For convenience, although the breakdown is somewhat arbitrary, the problems of liquid waste management will be considered under three headings: Low, intermediate, and high level wastes. Low level wastes are those which, if decontaminated by a factor of 100 or 1000, would approach permissible limits (14) for human exposure. The range of their activity would be from 10^{-4} to 10^{-3} microcuries per milliliter. High level wastes are those requiring shielding to protect persons handling them from exposure to damaging radiation and may contain 100 or more curies per liter. They are generally associated with the chemical processing of the nuclear fuel for the recovery of the fissionable materials. Intermediate wastes also require shielding and must be handled with considerable care. They may be the high level wastes which have lost a considerable fraction of their activity as a result of decay, the residual wastes following recovery or separation of the strontium or cesium isotopes, the wastes resulting from pilot plant operation, or lower level wastes that may have been concentrated by evaporation.

Low Level Wastes

Low level wastes are those most frequently encountered at the present time at least insofar as release into the environment is concerned. Depending upon their source, they are handled or disposed of in various ways. They may be contained in the residues following the recovery of uranium ore in which case they will contain essentially the daughter products of the uranium itself. Generally, these waste materials are discharged into the stream either directly or from tailings ponds. Studies by the Public Health Service have shown that water, mud, and biological samples collected below uranium mills have activity levels higher than similar samples collected upstream (15). The public health significance of these higher levels has not been evaluated as yet.

Another source of low level wastes results from the use of natural waters as reactor coolants. In passing through the reactor, activity will be induced in the normal constituents of the water as a result of neutron bombardment, and the water may pick up corrosion products and other materials. Most of the activity is relatively short-lived and is associated with the lighter elements. The largest source of such wastes is the Atomic Energy Installation at Hanford

where pretreated Columbia River water is used to cool the reactors. Before return to the Columbia River, the water passes through specially designed retention tanks which permit maximum decay of the radioactivity.

Low level wastes are produced in any laboratory where radioactive materials are used. They are extremely variable in composition—both from the standpoint of their chemical as well as radioactive constituents—and are generally released into the environment following limited treatment. At Oak Ridge National Laboratory, for example, they pass through settling basins before release into the Tennessee Valley System through the Clinch River. In general, the activity level in the Clinch River below the plant discharge is at the 10^{-7} microcurie per milliliter level indicated for mixed fission products or activities of unknown composition. Plans are underway to construct a treatment plant for the removal of strontium and other activities present in the wastes during peak activity levels. The plant is flexible in design and will permit the use of either phosphate coagulation or lime-soda ash softening to precipitate the strontium before the wastes are released to the Clinch River.

Brookhaven National Laboratory concentrates its waste by evaporation, mixes it with concrete, places it in steel drums, and carries it out to sea for disposal.

In Great Britain, the low level wastes which arise from the Atomic Energy Research Establishment at Harwell are discharged into the Thames River following treatment consisting of chemical coagulation. Treatment must be such that the quantities of radioactive materials discharged not exceed 20 curies per month or 5 curies in any one day as determined by the formula:

$$\begin{aligned} & \text{Ra (curies)} \times 2500 + \text{Other Alpha (curies)} \times 420 + \\ & (\text{Ca}^{++} + \text{Sr}^{++}) \text{ (curies)} \times 50 + \text{Remaining Beta (curies)} \\ & \leq 20 \text{ (curies) per month.} \end{aligned}$$

This formula was arrived at by applying a factor of 1/100 to the maximum permissible concentration recommended by the International Commission on Radiological Protection (16).*

Burns (17) reports that a small plant has been in operation at

*The British selected a factor of 1/100 because of the large numbers of people located downstream on the Thames River below the Harwell Establishment. This factor may be compared with a value of 1/10 used in this country when recommending levels for the general population.

Harwell for some considerable time treating mixed fission product solution in water of a low solids content. By treating this waste with phosphate and iron salts followed by a sulfide treatment and passing the liquid through columns of Vermiculite, decontamination factors of the order of 1000 have been obtained. If necessary, the water could be treated further by an electric deionization process which is expected to increase the decontamination factor to 100,000. If this is achieved it should be possible to reuse the water for many purposes and thus effect an economy.

Other formulas were developed for use in connection with the discharge of radioactive materials from the operations at Windscale, near Sellafield, England. Here the limiting levels for discharge into the Irish Sea were determined by the accumulation of activity in fish, in an edible sea weed, and on the beach sands. The formulas are:

$$\frac{\text{Total Alpha (curies)}}{200} + \frac{\text{Ru (curies)}}{5000} +$$

$$\frac{\text{Total Beta (curies)}}{20000} \leq 1 \text{ (curie) per month}$$

$$\frac{\text{Sr (curies)}}{2500} + \frac{\text{Total Alpha (curies)}}{300} \leq 1 \text{ (curie) per month}$$

The most common method of disposal, particularly by users of radioactive isotopes following application in medicine, research, or industry, is to discharge these materials into the sewer. Although this practice does not appear to be objectionable at present levels of activity, it may become necessary for public health authorities to maintain vigilance over the quantities of radioactive materials discharged as the use of radioisotopes increases.

Considerable information has been published on the effectiveness of water treatment methods for the removal of low level radioactive contamination. It will not be reviewed here, but the principles involved may be applied successfully to the treatment of low level wastes. Methods that may be used include coagulation (18) (19), softening (18) (19), ion exchange (18) (19) (20), permselective membranes (21) (22), clays (19) (23), powdered metals (24), etc.

Intermediate Level Wastes

In the past, storage was used for the retention of all liquid radioactive wastes containing appreciable quantities of radioactive materials. More recently, consideration has been given to the release of radioactive materials into the ground and into the oceans. If this can be accomplished without hazard to the environment, the cost of waste treatment and disposal may be materially reduced.

Ground Disposal. This method has been practiced at the Hanford site for approximately 10 years. Under the rather unique geologic, hydrologic, and meteorologic conditions at this site approximately 10^9 liters of active waste containing several hundred thousand curies of fission products have been released into the ground for storage (25). At the Oak Ridge National Laboratory, four waste pits have been scooped out in the weathered Conasauga shale overburden and have received the quantities of waste reported in Table I (26). Experience indicates that the nitrate moves through the soil most rapidly followed by the anionic ruthenium present.

It is reported that about 130,000 gallons of waste are discharged daily into the soil at the Savannah Project (27).

In the use of the ground, as will be defined more specifically under high level wastes, one must always be certain that the amount of radioactive material transmitted to the ground water will be at a sufficiently low level that the maximum permissible concentration levels will not be exceeded in its use as a source of water supply. Furthermore, it must be remembered that once radioactive materials are introduced into the ground, control of them has been lost.

Ocean Disposal. Except for the reference made to the Windscale operations in England little if any radioactive materials are discharged directly into the oceans. Some materials are discharged but these are in packaged form. The radioactive materials, in the form of evaporator concentrates, precipitated and dewatered sludges, etc., are mixed with concrete directly or placed in previously prepared concrete shielded cylinders, and then shipped out to sea for disposal. It has been reported (28) that approximately 20 to 30 tons of waste are disposed of annually on the East Coast as compared to 4 to 5 tons on the West Coast. Disposal must take place beyond the 1000 fathom level (29) in this country, and the British disposals are carried out in water more than 2000 fathoms deep, several hundred miles from

TABLE I—WASTE PIT CHARACTERISTICS
(as of October 31, 1955)

Waste Pit Number:	1	2*	3
1. Design capacity—gal	180,000	1,000,000	1,000,000
2. Dimensions of pit— length, width, depth—ft	100x20x15	210x100x15	210x100x15
3. Period of use	8/1/51-10/5/51	6/20/52-10/31/55	1/24/55-10/31/55
4. Raw waste added—gal	123,000	1,331,760	1,434,600
5. Waste from Pit 3—gal		398,200 ^a	—398,200
6. Total volume waste—gal	123,000	1,729,960	1,036,400
7. Beta activity added—curies	389	15,975	18,911
8. Beta activity taken from Pit 3—curies		3,449 ^a	—3,449
9. Total beta activity—curies	389	19,424	15,462
10. Radioisotopes present Per cent of total			
Cs ¹³⁷ -Ba ¹³⁷	60	60*	77
Ru ¹⁰⁶ -Rh ¹⁰⁶	40	40*	12
Sr ⁹⁰ -Y ⁹⁰			11
11. pH of waste added	~12.5	~12.5	~10-11
12. Approximate construction cost—dollars/million gal	\$14,500	\$14,500	\$14,500
13. Approximate cost/gal waste capacity—cents	1.45	1.45	1.45
14. Approximate cost—gal waste discharge to pit—cents	2.12	< 0.84 ^b	< 1.40 ^b

*Decanted liquid from Pit No. 3 and added the material to Pit No. 2 after 1/24/55.

^aVolume of waste and activity decanted from Pit No. 3 and added to Pit No. 2. Activity estimated from activity added just before decanting.

^bBased on total gal waste volume in pit. Equal to line 13 values divided by line 6/1,000,000 values.

the British Coast, and in areas approved by the Minister of Agriculture, Fisheries and Food (30). The need was pointed out (28) for special reinforcing of the concrete in the containers to provide strength to meet the impact when the container strikes the hard ocean floor and the high hydrostatic pressures in 500 or 1000 fathoms of water.

In describing Brookhaven experience with ocean disposal, Ginell (31) reports that about 660 drums of waste totalling 600 curies were dumped at a cost of \$20 per drum exclusive of overhead. North American Aviation (32) ships its stored wastes to sea at a cost of approximately 71 cents per gallon based upon 100, 55-gallon drums per dumping 60 miles off the coast of Southern California at a depth greater than 800 fathoms.

High Level Wastes

These originate principally from the chemical processing of spent fuel for the recovery of the fissionable material. Associated with the irradiated fuel is the entire spectrum of fission products which occurs when the uranium atoms are split. The fission products must be separated from the fuel to permit further processing or reuse of the fuel in the reactor. Following removal from the reactor, the irradiated fuels are generally stored under water for periods of from 90 to 120 days to permit decay of the short-lived fission products. The problems of handling and disposing of the product are intensified when shortened storage periods are employed.

After decay, the fuel material is processed. The processing varies depending on whether the fuels came from a heterogeneous or a homogeneous fuel reactor and on the materials used in the construction of the fuel element. The separations processes that are being utilized or are under study include precipitation, ion exchange, fractional distillation, solvent extraction, high temperature processing (volatility), and pyroprocessing.

The characteristics of a typical reactor-fuel-processing waste as reported by Wolman and Gorman (28) are given in Table II. The wastes contain the fission products, some unrecovered fissionable material, and the various acids and salts required for dissolution of the material. These wastes, which are highly acid, may be stored in stainless steel tanks or they may be neutralized and stored in mild steel lined concrete tanks. Tank storage is the method presently em-

ployed at all atomic energy plants. Thus far we have had little over a decade of experience with such tanks without any serious results. However, the storage facilities have been located in relatively isolated or remote areas of the country. With the impetus being given to the use of nuclear energy for power development, it is reasonable to expect that nuclear power reactors will be located, and they are being located, close to large centers of population.

TABLE II—CHARACTERISTICS OF TYPICAL REACTOR FUEL PROCESSING WASTES
(High-Level Waste)

Gross Beta activity,	1.6×10^6 - 2.2×10^{10} cpm/ml
Alpha activity,	6.0×10^3 - 6.0×10^5 cpm/ml
Radioactivity, 1 to 4×10^2 curies per gallon (neutralized)	
Effective life, about 600 years	
Heat generation, 1 to 3 BTU/hr/gallon	
Power equivalent, 1 gm U^{235} -24,000 kwh (100% efficiency)	
Fission product wastes, 1 gm U^{235} forms 1 gm fission products	
Wastes from processing 0.5 to 5.0 gal waste solution/gm U^{235} consumed	

Waste chemistry:

Ions	Concentration in moles per liter
Al	0.5-2.5
NO_3	2.0-8.0
H	0.5-3.0
N_a	0.1-0.2
F	2.0-3.0
Zr	0.3-0.6
SO_4	0.3-0.6

Specific gravity, 1.1 to 1.4

In the reactor types under consideration at present, particularly those employing solid fuels, chemical processing of the fuel was to take place at one of the AEC sites having such facilities. However, on January 5, 1956, the AEC advanced a policy aimed at having commercial chemical plants ready to process spent fuel elements from the first privately owned power reactors (33). In addition, the Commission would supply the plants with an initial base load of spent fuel from one or more of perhaps 20 reactors. Since these facilities will probably be near proposed reactor stations or at least near transportation facilities, it is probable that they too will be near centers of population.

In the case of homogeneous reactor stations, the chemical processing plant, at least for the partial treatment of the fuel, will have to be located adjacent to the reactor. Under these conditions, the location of facilities for the storage of the large quantities of waste resulting from an expanding nuclear power economy presents a more difficult problem. Accordingly, considerable thought and energy is being given to the development of more satisfactory and permanent methods of disposal. Some of the proposed methods have been studied on a laboratory scale and are now being investigated in pilot plant facilities. Others are only in the thinking stage. The methods that have been demonstrated or suggested will be discussed in somewhat greater detail in the sections which follow.

Storage. Storage itself is not being abandoned even though the cost on a per gallon basis seems rather high. Prices quoted range from about 30 cents to \$2.00 per gallon of tank capacity. However, when calculated on a kilowatt hour basis the cost is not high. Because power reactor wastes will have higher activity levels, it is necessary to provide facilities for cooling the waste solution. In conventional terms, the energy release in separations plant waste is small, up to 1 watt per liter of solution, but its management is complicated by the large volumes involved and nonuniform distribution promoted by the tendency to form precipitates in wastes normally made alkaline to minimize corrosion (34).

Grandquist and Tomlinson (35) point out that with a special type of fuel recovery process and mechanical dejacketing of the fuel elements about 20,000 gallons of highly active waste would be produced by irradiation of natural or slightly enriched uranium to about 2500 megawatt-days per ton. The waste volume would be expected to self-boil perhaps for 50 years. At Hanford nonboiling wastes in 500,000 to 1,000,000 gallon tanks can be stored in reinforced concrete tanks with mild steel liners at a cost of 20 to 25 cents per gallon and self-concentrating wastes may be stored at a cost of 40 to 50 cents per gallon of tank space. Since this cost amounts to but 0.01 to 0.05 mils per kilowatt of electrical power produced, there is no pressing economic need to develop a better system of waste handling (35).

Separation and Storage. Glueckauf (36) suggests that the quantitative removal of Sr^{90} and Cs^{137} from the wastes and separate storage from the bulk of radioactivity will permit consideration of rela-

tively short-time storage (about 20 years) for the residual activity. By such separation and storage it may be possible to recover the more usable radioisotopes and to release the residual activity under suitable conditions into either the ground or the oceans. A similar scheme is under investigation at the Oak Ridge National Laboratory where the removal of specific radionuclides (strontium, cesium, zirconium, niobium, yttrium, ruthenium, and cerium) from simulated wastes by precipitation (37) and solvent extraction (38) has been evaluated. The separation of the waste into such components with separate storage of the more hazardous fraction has advantages in that less storage space will have to be provided, less elaborate storage may be provided for the bulk of the wastes, self-boiling of the wastes will not take place, and there will be no need to provide special tank cooling facilities.

Jonke (39) has described a process for converting aqueous nuclear wastes to solid form by injection into a fluidized bed of heated solids, where evaporation and calcination of the waste to oxides is effected. Another calcining operation has been described by Hatch, Regan, Manowitz, and Hittman (40) in which simulated high activity waste streams containing 30 wt % salt were converted to anhydrous free-flowing melts.

Specific studies for decontaminating aluminum waste solutions were described by Blanco, Higgins, and Kibbey (41). They utilized a scavenging precipitation and an Al-resin ion exchange technique. Gross beta-gamma decontamination factors of 10^3 (influent over effluent activity) were obtained with 97 to 99 per cent of the aluminum appearing in the eluate. The Cs and Sr decontamination factors were 10^3 to 10^4 , respectively. A further development was reported by Higgins and Wymer (42) who decontaminated an $\text{Al}(\text{NO}_3)_3$ -nitric acid radioactive waste. Ninety per cent of the niobium and 95 per cent of the ruthenium and zirconium were removed by a ferric hydroxide-manganese dioxide scavenging precipitation. The aluminum nitrate was converted to a dibasic aluminum nitrate by destructive distillation and dissolved by 12-hour digestion at 160°C . The strontium, cesium, and rare earths were removed by cation exchange in a continuous contactor. The decontaminated waste can be volume-reduced to about $6M$ aluminum before disposal and then stored cheaply. The fission products can be eluted from the column, concentrated to a

small volume of highly radioactive waste, and stored with all necessary precautions.

At Los Alamos (43), a radioactive waste containing Ba^{140} , La^{140} , Sr^{90} , and Y^{90} was passed through a cation exchange resin column. Storage in the column results in the decay of short-lived Ba and La isotopes. Within reasonable limits of raw waste quality, the ion exchanger reduces the Sr^{90} concentration to permissible levels for discharge. The resin column was regenerated on exhaustion and spent regenerant treated by chemical precipitation and vacuum filtration.

Another scheme, reported by Glueckauf and Healy (44), for the separation of cesium and strontium involves the following: The fission products are taken to dryness to remove all nitric acid and water; then the dry material is roasted for about an hour at $300^{\circ}C.$, whereby all the nitrates except those of alkali and alkaline earth metals are decomposed into oxides. The solid is then leached with warm water which dissolves the cesium and strontium nitrates, leaving behind all the water insoluble oxides. Usually about 95 per cent of the cesium and 85 per cent of the strontium can be extracted. The roasting temperature is critical to $\pm 10^{\circ}C.$, otherwise the leached material either contains too many impurities (at $<290^{\circ}C.$), or retains too much cesium and strontium with the oxides (at $>310^{\circ}C.$). The strontium and cesium recovered in this way may be further concentrated for separate storage and the residual fission products may be stored for a shorter time in separate facilities.

Fixation of Wastes and Firing. Ginell (45) and Hatch *et al.* (46) (47) (48) have conducted experiments dealing with the fixation of radioactive contaminants on montmorillonite clays. In their process, the fission products were passed through a column containing extruded clay (spaghetti) and the clay was fired at temperatures up to $1000^{\circ}C.$ Subsequent leaching showed that the amount of material leached was a function of the initial firing temperature. At temperatures approaching $1000^{\circ}C.$ the amount of leaching was small and stabilized quickly. More recent investigations by the Brookhaven group have extended this method of disposal to high level wastes. Because of the large amounts of nitric acid or aluminum nitrate in the waste solutions, and because these quickly saturated the montmorillonite clay, it was necessary to pretreat the wastes to remove these high concentrations of stable materials. This was done original-

ly through the use of permselective membranes. More recently, the pretreatment has been modified and now includes kiln-drying of the aluminum nitrate-fission products waste to form aluminum oxide. The aluminum oxide plus fission products is leached; the leachate containing a fraction of the fission products is then passed through montmorillonite clay; the clay columns are fired; and the activity fixed permanently.

The approach used by the Health Physics Division, Oak Ridge National Laboratory, was somewhat different in two respects. One, adsorption on the indigenous Conasauga shale was investigated; and two, fixation following mixing of the Conasauga shale and acid aluminum nitrate waste with specific quantities of limestone and sodium carbonate was evaluated. It was found that sintering at temperatures above approximately 500°C. could fix much of the activity. Subsequent leaching with water showed little release of activity with the exception of cesium. The next approach investigated the possibility of utilizing the heat of decay to fix the activity directly to the soil without the need of any external heat. The amounts of heat available have been reported by Perring (49) and could be sufficient, with enough concentrated activity, to permit self-fixation of the waste.

From studies reported by Johnson *et al.* (50) at ORNL it was concluded that liquid containing between 100 and 1000 curies per gallon may be disposed in insulated concrete lined earth pits and will self-heat to temperatures adequate to become fixed in clay-flux mixtures. The mixtures used in these experiments were developed by McVay, Hamner, and Haydon (51). They combined Conasauga shale, soda ash, and limestone with a simulated aluminum nitrate-nitric acid waste solution to form a ceramic mass at a temperature as low as 1050°F.

The Oak Ridge studies led to the development of the so-called pilot plant hot-pot experiment which employed clay and outside sources of heat. Fixation of fission products from acid aluminum nitrate waste was effected and studies are now being planned in which high level wastes will be employed directly to determine the self-heating and self-fixing characteristics of such mixtures.

Studies at Los Alamos (52) showed that alpha activity in filter cake (4000 c/m/g) and raffinate wastes (126,000 c/m/ml) were fixed for safe disposal when fired with clay. Beta-gamma wastes at the 400 millicurie level have not been satisfactorily fixed by this method. Ad-

ditional studies are underway to determine the influence of such factors as clay mixtures, controlled firing, fluxes and glazes, and leaching solutions. Patrick (53) has proposed a method of synthesizing aluminum silicates which eventually metamorphose to feldspatic structures with alkali or alkaline earth oxides. He is applying this method to the removal of strontium and cesium from high level wastes.

Mawson (54) refers to pilot plant studies made at Chalk River, Canada in which a 2.5*N* nitric acid waste of high activity is mixed with nepheline syenite, and fused at a temperature slightly below 1000°C. forming an opal glass. Tests have shown that a very small amount of activity is leached initially, but after a short time very little activity comes off.

Amphlett (55) (56) has investigated this problem in England and reports (56):

"The use of natural silicate materials or soils as bases for formation of unleachable products, by mixing with the waste and firing to high temperatures, offers promise. Higher loadings can be achieved than by means of ion exchange, and inactive ions and acid do not appear to affect the adsorption of activity. This method may enable absorption of bulk wastes in one operation, and because of the shorter-lived nuclides are included there is a possibility of self-fixation on a reasonable scale. No preparation of the material is required, and the process may be applicable to slurry and solid wastes as well as to solutions. If possible, the process should be aimed at melting the mixture to form a glass, in order to achieve the maximum density and hence the maximum concentration factor. This requirement is of course subject to leaching properties, loss of activity by volatilization during firing, and the possibility of lowering the melting point by the use of suitable additives. It is clear that much further work is required.

Ground Disposal. Before burial of radioactive materials the geology and hydrology of the particular disposal site must be well understood. The hydrological and geological factors affecting ground disposal at Hanford, Brookhaven, Oak Ridge, Idaho Falls and Savannah were discussed by Brown *et al.* (57) de Laguna (58), and Theis (59). Theis (60) also described the general types of formations useful for ground disposal. These include 1) sandy, relatively permeable formations; 2) permeable cavernous formation; 3) jointed or otherwise

fractured rocks; 4) relatively impermeable shales; or 5) deposits of salines—rock salt or gypsum. Thurston (61) points out that many geological principles and methods used in the petroleum industry are applicable to radioactive waste disposal. The preparation of artificial cavities in salt and shale for the storage of hydrocarbons may be adaptable to the shallow underground disposal of a comparable volume of radioactive wastes. Furthermore, the natural porosity of many sedimentary rocks may be suitable as reservoirs for the safe disposal of wastes at great depths.

The factors which must be evaluated in considering the feasibility of disposal of high level radioactive wastes into the ground include: 1) the chemical and radiochemical content of the waste; 2) the effectiveness of retention of the radioisotopes in the available soil column above the ground water table; 3) the degree of permanence of such retention, as influenced by subsequent diffusion, leaching by natural forces, and additional disposal; 4) the natural rate and direction of movement of the ground water from the disposal site to public waterways, and possible changes in the characteristics from the over-all liquid disposal practices; 5) feasibility of control of access to ground water in the affected region; 6) additional retention, if any, on sands and gravels in the expected ground water travel pattern; 7) dilution of the ground water upon entering public waters; 8) maximum permissible concentrations in those public waters of the radioelements concerned; 9) the temperature and pressure effects resulting from the heat of decay; and 10) the effect of waste discharges on present or potential mineral wealth of the region. High level wastes are not discharged into the ground in any location at the present time.

The reactions of radioisotope solutions with Hanford soils were described by McHenry, Rhodes and Rowe (62) with respect to the effects of concentration, pH, total ion concentration, and the type of soil. A significant increase in Sr adsorption in the presence of phosphate ion is shown to counteract the adverse effect of high salt concentrations. Experimental evidence and theoretical considerations illustrate the very low diffusion rate of strontium. Preliminary investigations were carried out by Kaufman *et al.* (63) on the feasibility of high level radioactive waste disposal by injection into isolated geological formations. Density effects and exchange reactions between the simulated Sr wastes and columns of clay or oil sand

were studied. They observed that exchange reactions may retard the advance of radiostrontium concentration fronts to as much as 1/40 that of the liquid front.

Ocean Disposal. As reported earlier, the only direct discharge of radioactive waste materials into the ocean is at Sellafield, England, where the wastes from the Windscale plant are discharged into the Irish Sea. In other instances, packaged materials are dumped in specified disposal areas, but in all cases these are of low- or intermediate-level wastes. There has been no discharge of high-level radioactive materials. Some of the problems facing the oceanographer in determining the feasibility of such a disposal scheme have been reported by the British in describing their studies prior to the Irish Sea discharges (64) (65) (66). It is obvious that mixing, either desired or undesired, will play a major role in defining suitable disposal areas, if such exist.

With respect to ocean disposal of radioactive materials, the National Academy of Sciences (67) states:

"Sea disposal of radioactive waste materials, if carried out in a limited, experimental, controlled fashion, can provide some of the information required to evaluate the possibilities of, and limitations on, this method of disposal. Very careful regulation and evaluation of such operations will, however, be required. We, therefore, recommend that a national agency, with adequate authority, financial support, and technical staff, regulate and maintain records of such disposal, and that continuing scientific and engineering studies be made of the resulting effects in the sea."

Cost. Zentlin (68) has estimated the allowable cost of waste disposal to be \$4 per gallon in the case where the reactor burnup is 5000 megawatt-days (heat) per ton, the processing volume 1200 gallons per ton, and the allocated cost for waste disposal 2 per cent (or 0.16 mills/kwhr_e).

Non-Radioactive Wastes

Not all wastes produced by the nuclear energy industry are radioactive. Where acid dissolution of partially spent fuel materials is practiced, large quantities of nitrate wastes will result. Christenson *et al.* (69) have developed an activated sludge process without diffused air which has a maximum rate of nitrate reduction in the order of 70 ppm per hour. Their feed, based upon a 6-liter feed vol-

ume, is 600 ppm methanol, 250 ppm nitrate nitrogen and 10 ppm PO_4 . This mixture is reduced in the 8-hour contact time to no nitrogen and about 40 ppm oxygen consumed.

Liquid wastes from the production of hafnium-free zirconium contain 1.8 pounds of ammonia per pound of zirconium produced along with trace amounts of cyanides, thiocyanates, sulfates and chlorides. McDermott (70) indicates that the process wastes, most objectionable from the pollution standpoint, amount to 10 gallons of concentrated ammoniacal waste per pound of zirconium produced. The waste is held in basins before trucking to a large river for disposal by dilution.

In the uranium and thorium refining and reduction operations large quantities of fluoride containing wastes are produced by dumping in a pit at a cost of \$0.54 per cubic foot (71). Liquid wastes are released in such a manner that fluoride content of the stream does not exceed 1.2 ppm (background fluoride level of stream is 0.4 ppm).

SUMMARY

The Yankee Atomic Power Reactor installation, planned for Rowe, Massachusetts, has been described. Since little information has been made available relative to provision for waste disposal facilities, the disposal facilities associated with the power reactor installation at Shippingsport, Pennsylvania, a similar type reactor, have been listed and the types of wastes produced noted.

The techniques in use or under study for the handling, storage, and/or disposal of solid, gaseous or airborne, and liquid wastes have been discussed. Burial, incineration, or disposal at sea have been suggested for handling solid wastes; collection, concentration, and dilution for gaseous or airborne wastes; and storage, separation, fixation on clays and other materials, ground disposal, and ocean disposal for liquid wastes. Examples are also given of non-radioactive wastes that pose potential pollution problems.

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