TREATMENT AND DISPOSAL OF WASTES IN NUCLEAR CHEMICAL TECHNOLOGY

Bernard Manowitz

Brookhaven National Laboratory, Upton, L. I., New York

I. Introduction

A. MAGNITUDE OF THE PROBLEM

Man has always been exposed to radiation from cosmic rays and other natural sources. Present developments in nuclear energy are such, that with the passage of time, the radiation background will be raised significantly by radiation sources of man's own making, if these are not controlled. From what is already known about the biological effects of radiation, an intensification of the radiation background is likely to lead to somatic and genetic effects in man. The former will occur in the population exposed, while the latter will accumulate and affect future generations. Although the National (N1) and International (I1) Committees on radiation protection have agreed upon maximum permissible concentrations of radioisotopes in air and water for continuous exposure, it is important to note their conclusion: "While the values proposed for maximum permissible exposures are such as to involve a risk that is small compared to the other hazards of life, nevertheless, in view of the unsatisfactory nature of much of the evidence on which our judgments are based, coupled with the knowledge that certain radiation effects are irreversible and cumulative, *it is strongly recommended that every effort be made to reduce exposures to all types of ionizing radiations to the lowest possible level.*"

Thus, an important task, responsibility, and challenge to the nuclear energy industry and its progress is a constant improvement in methods and facilities for control of radioactive waste. Simultaneous with an improved waste technology, cost reductions must be effected if the nuclear industry is to be competitive with other sources of energy.

A number of attempts have been made to assess the magnitude of the disposal problem the world may face during the next half century. Estimates have been made by Hatch (H1), Glueckauf (G2), Rodger (R4), and others. These estimates all tend to agree that the maximum plausible use of nuclear energy will result in the burnup of approximately 1000 tons of fissionable material (and the production of 1000 tons of fission products) per year. This is equivalent to approximately 2×10^6 megawatts, installed reactor capacity. Assuming this production rate, Rodger has calculated the equilibrium quantity of eight significant isotopes given in Table I. Also given are the maximum permissible concentrations of these isotopes in water and in air, and from these are calculated the volumes of water and air required to dilute the isotope in question to tolerance. Note that strontium-90 is the controlling isotope and that the dispersal volume of water needed, 2.6×10^7 cubic miles, is about 5% of the entire world ocean volume.

It should be noted that radioactive contaminants are not to be compared with their stable chemical counterparts. For example, because of the unchangeable character of the nuclear disintegration phenomenon, the natural purification processes which restore chemical and biological balances in the environments of living things, would have no effect on the radio-toxic properties of the radioactive elements. The time required for accumulations of strontium-90 to decay to background levels, would be measured in centuries. It is clear that an amount of radio-toxic materials such as is indicated in Table I, should never be introduced into our world, except under the most carefully planned and controlled conditions.

TABLE I

50-YEAR ACCUMULATION OF LONG-LIVED ISOTOPES AND REQUIRED DISPERSAL VOLUMES

Basis: 2.2×10^6 MW Installed Reactor Capacity—3 Tons of Fission Products per Day

Isotope	Accumulated Quantity in 50 years, curies	Maximum Permissible[c] Concentrations (μ curies/ml.)		Volume Required to Dilute to Tolerance (cubic miles)	
		Water	Air	Water	Air
Zr^{95}	1.3×10^{11}	4×10^{-3}	4×10^{-7}	7.8×10^{3}	7.8×10^{7}
Ce^{144}	1.1×10^{11}	4×10^{-2}	7×10^{-9}	6.6×10^{3}	3.8×10^{9}
Ru^{106}	1.0×10^{11}	0.1	3×10^{-8}	2.4×10^{2}	8×10^{8}
Pm^{147}	5.1×10^{10}	1	2×10^{-7}	12	6×10^{7}
Sr^{90}	8.6×10^{10}	8×10^{-7}	2×10^{-10}	2.6×10^{7}	1×10^{11}
Cs^{137}	8.1×10^{10}	1.5×10^{-3}	2×10^{-7}	1.3×10^{4}	9.7×10^{7}
$Tc^{99a} \perp$	2.0×10^{7}	3×10^{-2}	3×10^{-6}	0.2	1.6×10^{3}
$Pu^{239b} \perp$	2.8×10^{6}	1.5×10^{-6}	2×10^{-12}	4.5×10^{2}	3.4×10^{8}

[a] Decay neglected.

[b] Based on a loss of 0.1 % in processing.

[c] From National Bureau of Standards Handbook **52** (1953).

The problem of what to do with these large quantities of waste has received serious attention. Glueckauf (G2) has suggested separating out strontium-90 and cesium-137, storing them separately, and storing the remainder of the waste in tanks for 13 years, after which time these could be discarded into the sea. He calculates that with strontium and cesium removed, the other activities after 13 years would be negligible compared with the potassium-40 activity, which is naturally present in the sea. Rodger (R4) points out that the separation of strontium and cesium must be truly quantitative. Leaving as little as 0.001% of the strontium-90, will cause the storage requirement to be raised from 13 years to a century.

The general conclusion is that wastes should be reduced to solids and confined. If the most optimistic assumption is made, namely, that the inert salts can be held to a weight equal to the fission products themselves, at the 1000 ton/year rate, six tons per day of concentrate would be formed. This is not an impractical amount to consider storing permanently.

Of course, permanent storage implies facing a set of opposing forces tending toward dispersal. Corrosion and other forms of chemical and physical disintegration of containers would be unknown factors, involving the consideration of very long periods of time. All sorts of possible disturbances of the balance of natural forces would have to be taken into account, as well as the faculty of man himself to depart from principle in order to follow short-term objectives.

B. SOURCES OF WASTE

As our nuclear technology develops, the major sources of radioactive wastes will be reactor sites and fuel recovery sites.

Most of the present nuclear reactors have been burning solid fuel elements of either normal or enriched uranium. Thus far, it has been necessary to reprocess fuel in order to recover valuable fissionable or fissile material. It is possible that fuel elements will be developed for future reactors which can be burned to the point where it is not economically justifiable to recover fissionable materials. Obviously, this depends upon the value of these materials. Such a procedure would provide an optimum solution to the major part of the waste disposal problem. The fission products would still be locked in the fuel element, simple disposal techniques could be employed, and in fact, spent fuel elements would probably have secondary uses as radiation sources.

A second and more likely possibility is that fuel from future reactors will have to be reprocessed, resulting in the dilution and dispersal of the fission products into many media.

A third possibility is that fluid fuel reactors, such as the ORNL aqueous reactor, or the BNL liquid metal reactor, will supply some fraction of our future nuclear economy. These reactors are usually designed as completely self-contained systems including continuous reprocessing of fuel.

Various sources of waste in a complete reactor cycle are shown in Fig. 1.

The processes associated with fluid fuel reactors are too varied to be considered in detail here. The reader is referred to references (F1) and (D3) for further information.

There are three sources of contamination connected with the operation of solid fuel reactors, excluding uncontrolled nuclear reactions. The first source is an imperfection or failure of the jacket which protects metal fuel elements. If the coolant contains oxygen or an oxidizing agent, uranium will readily oxidize and particles of uranium containing fission products, will be carried out by the coolant.

The second source of contamination in normal reactor operation is induced activity by neutron irradiation of the coolant. The amount of activity formed in the coolant may be determined for each element. It depends upon the average neutron flux, the absorption cross-section for the element, the number of atoms of the element irradiated, the concentration of the element in the coolant, and the half-life of the isotope formed. In air coolants, A^{41} is the principal activity formed. Particulate matter may also contribute to air pollution. If water is the coolant, the dissolved impurities can be a source of appreciable radioactivity.

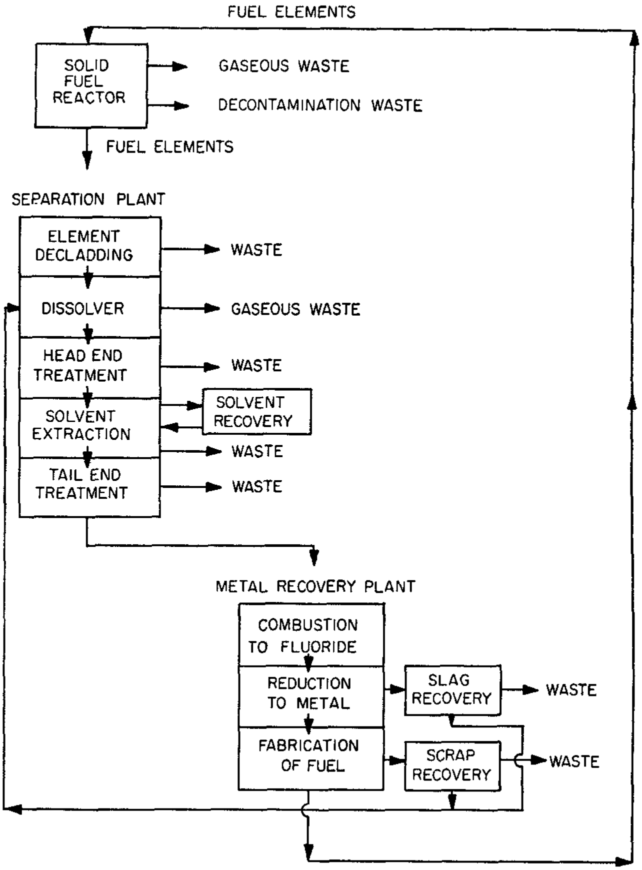

FIG. 1.

Leaks and auxiliary equipment failure, can be a third source of release of radioactivity.

The major source of radioactive waste is likely to be the separation plants designed to treat solid fuel elements and to separate and purify uranium and plutonium from the fission products. Detailed descriptions of such plants are given in reference (R1).

The first thing that will happen to the fuel sent to such a plant will be the removal of its cladding. This may be done mechanically or chemically, resulting in either a solid or a liquid low-level waste. Next the fuel itself must be put into solution. This may involve the use of some hazardous chemicals, depending upon the nature of the fuel. As long as the fuel is composed of uranium metal, it can be dissolved in nitric acid. However, enriched fuels in alloy form require solvents which will dissolve the fuels. For instance, hydrofluoric acid may be required for zirconium-uranium alloys. Typical separation processes employ solvent extraction as the primary means for separating uranium and plutonium from each other and from the fission products, accompanied by head-end treatments (usually precipitation) and tail-end treatments (usually adorption or ion exchange), to improve the purity of the product. These processes result in both high-level and low-level liquid waste streams, and both must be properly disposed of.

For a typical plant large enough to process fuel from five to ten reactors, the liquid waste generation picture is likely to be as follows:

The high-level waste stream may consist of from 10^5–10^6 gallons per year of an aqueous solution of various inert salts, ranging from 0 to 20% of total solids contaminated with 10 to 100 parts per million of fission products. The activity level of such a waste would be in the hundreds of curies per gallon range, so that even small samples of such waste would be dangerously radioactive.

The low-level waste stream may consist of 10^6–10^7 gallons per year of an aqueous solution of various inert salts ranging from 0 to 1% total solids contaminated with 0.01 to 0.1 parts per million of fission products. The activity level of such waste would be in the millicurie per gallon range.

Gaseous waste will be generated during the fuel-dissolving stage. Exit gases from the dissolver will contain contamination in the form of undesirable vapors (iodine), noncondensable gases (Xe, Kr), and particulate matter on which radioactivity has been adsorbed. Cell ventilation gases may also contain gaseous and particulate contamination.

There will be an inevitable accumulation of solid wastes at such a plant. In general, these may be categorized into contaminated equipment, contaminated burnable trash, and contaminated nonburnable trash.

II. Waste Disposal as a Consideration in Site Selection

A. REGULATIONS AND PRACTICES IN DISCHARGE OF RADIOACTIVITY TO THE ENVIRONMENT

Relatively few states have as yet adopted regulations governing the disposal of radioactive wastes. A considerable amount of work on determining the biological hazards of various radioisotopes, has been done by Morgan, Stone, and others, leading to the concept of the mean permissible concentration of radioisotopes in air and in water. Specifications proposed depend upon:

(1) The chemical and radiochemical nature of the fission products: for example, Ac^{227}, At^{211}, Po^{210}, Am^{241}, Pb^{210} and Sr^{90}—Y^{90} are generally among the more hazardous isotopes, the first four being among the 12 most hazardous in every case.

(2) The site in which they are deposited; for inhalation, the lung or gastrointestinal tract is usually the critical body organ.

(3) The length of time during which they are retained by the body, this factor being termed the biological half-life. (The longest biological half-lives are associated with Sr^{90}—Y^{90}, Sm^{151}, Pu^{239}, Ra^{226}, and Th^{232}.)

(4) The method by which they are adsorbed, i.e., whether as dissolved ions, particulates, or gases.

General tolerance levels for unknown mixtures of fission products are roughly as follows (as taken from "Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Concentrations in Air and Water" [N1]):

Kind of Uptake	Type	Maximum Weekly Dose
1. Total body irradiation	$\beta\gamma$	0.3 *r*/week
2. Ingestion in water, food, etc.	$\beta\gamma$	3×10^{-7} μ curie/cc., water, based on Sr^{90}
3. Inhalation of particulates	$\beta\gamma$	2×10^{-10} μ curie/cc., air, based on Sr^{90}
4. Inhalation of fission gases	$\beta\gamma$	3×10^{-9} μ curie/cc., air, based on I^{131}

It has been common practice to set the mean permissible concentrations for environmental exposure at $\frac{1}{10}$th of the above occupational exposure values. It is likely that these values will be incorporated into state and local regulations restricting the *concentration* of radio-nuclides that may be discharged to the local environment. It is probable, however, that some legislation will be developed that will also regulate the total amount of radioisotopes per year that may be discharged from a site. Agreements have been made between the AEC (Atomic Energy

Commission) and Brookhaven National Laboratory, for instance, that in addition to the above concentration requirements, no more than 1.5 curies per year of mixed fission products will be discharged to the local environment. Using this level as a guide, it will probably be permissible to discharge no more than one or two curies per year to the environment in populated areas. In relatively unpopulated areas, these requirements may be relaxed to perhaps 100–1000 curies per year. In cases where specific nuclides of short half-life are released, as for example, A^{41} from air-cooled reactors, the amounts permissible for release even in populated areas, are likely to be greatly in excess of 1000 curies per year.

B. Geology and Hydrology of Site

In any consideration of waste disposal, it is of prime importance to first obtain as good a picture as possible of the geology and hydrology of the site. Some information should be obtained on shallow and deep geology and on surface and ground-water hydrology. The presence of faults and the history of seismic disturbance should be determined. The chemical and physical nature of the soils should be ascertained.

It is important that these geologic data be determined both from the point of view of evaluating potential hazards, and from the point of view of evaluating the maximum amount of activity that may be safely discharged to the local environment.

C. Meteorological Problems

Weather factors are of importance in site selection, site construction and site operation. Preliminary advice on the climatological picture will often point out potential atmospheric pollution hazards and help in making a decision on selecting one particular site out of several.

The conventional use of meteorology will be most important during the construction period at a site. On some projects it may be necessary to furnish climatological data for prospective bidders, seasonal work scheduling, and on information not commonly available, such as freeze-thaw cycle frequency, and annual wet-bulb temperature frequencies and ranges.

After an atomic energy site begins operations involving the use of radioactive materials, meteorological services may be required under the following conditions: (1) if there is a continuous or intermittent source of radioactive effluent present in the area, or if there is a source potentially present to the extent that an atomospheric monitoring program is required; (2) if there is a daily or occasional requirement for weather forecasts, which may be required for the control of stack effluent; (3)

if there is a need for meteorological observations for documentation and historical purposes.

These records may be used in conjunction with testing and experimentation, or in the event of legal action arising from alleged radiation exposure.

The need for meteorological control should be carefully evaluated before being resorted to. "Meteorological control" implies stopping or slowing down plant operations for the duration of meteorological conditions, which are estimated to produce above-tolerance, ground radioactivity.

Two atomic energy sites where experience has been obtained in the routine forecasting for the control of radioactive effluents, are Brookhaven (B2) and Hanford (C2). The scheme for estimating probable ground concentrations at Brookhaven was based on O. G. Sutton's diffusion theories, on empirical relations, and on certain micrometeorological variables at the 355-foot level. A series of templates were prepared for a wide range of weather conditions, which gave the mean value of the dose rate averaged for 1 hour over an area 1 kilometer radius by 10 degrees, extending from the stack base outward for a distance of about 15 kilometers.

Since the radiation tolerance for both on and off-site locations was on a 7-day basis, the stack output for the previous 6 days was converted by means of the appropriate templates into radiation-dosage values. A decision could then be made as to whether continuation of reactor operations would result in dosages higher than the specified maximum.

As the experience at Brookhaven grew, it became evident that the curtailment of reactor operations due to meteorological conditions was sufficiently infrequent to warrant discontinuation of the regular forecast service. Accordingly, routine 24-hour forecasts of radiation dosages were discontinued on November 30, 1952. Since then, the 6-day dosages have been computed from the past meteorological and stack output records, but a forecast is prepared only when there is a chance of exceeding the permissible radiation limits.

At Hanford, a somewhat different forecasting procedure has been used. Teletype reports, records of instruments from a 410-foot tower, and pilot balloon observations four times daily, have been used to arrive at a "least dilution" factor (C2) which is likely to occur during a particular plant operation.

In planning for any new atomic energy area, the decision on the scope of the required meteorological program is fundamental to economic operations and safety. This is especially true where meteorological control of the operation is being considered. In most instances the use of reliable air-cleaning devices, sufficiently high stacks, or isolated sites will be more economical and much more satisfactory than operating a program only during favorable meteorological conditions.

III. Waste Treatment and Disposal Practices

A. LIQUID WASTES

1. *Collection and Pretreatment*

Liquid wastes at an atomic energy plant can, in general, be categorized as follows:

a. Uncontaminated Waste. These are usually cooling waters or process waters that are normally clean, but that may potentially be contaminated. Careful consideration should be given to finding ways in which to discharge washes known to be uncontaminated, into drains other than the special waste system to prevent unnecessary overloads on sampling and holdup equipment.

b. Low-Level Waste (μc./gal. range). At some reactor sites and at research laboratories, considerable amounts of liquid waste in the μc./gal. to mc./gal. range will be generated. These wastes are normally neutralized, sampled, and held up prior to disposal in site storage tanks, or in some cases, are finally disposed of by incorporation into concrete and burial at sea. Neutralizers for these "warm" wastes can be of several types—chemical, injection, or leaching tanks. Holdup tanks should have provisions for sampling, radioactive level recording, as well as convenient access for possible decontamination. Frequently a small diversion tank is placed just before the holdup tank, specially instrumented so that a potential slug of highly active waste will be diverted before being added to a large volume "warm" holdup tank. These tanks should be located so that no pumping has to be done until after the wastes reach them and have been neutralized. At least two holdup tanks should be present, so that the contents of one tank may be neutralized and disposed of, while the other is filling.

c. Low-Level Waste (mc./gal. level). These wastes result from second-cycle processes and cell washings at separations plants. Frequently, a portion of these are highly acid wastes which first go through an acid recovery process. At plants processing natural uranium, these wastes are neutralized and concentrated before storage. Neutralization and concentration are remotely controlled operations. Stainless steel equipment is used. Transfer of liquids from neutralization to concentration to storage is by jet. At plants processing enriched uranium, low-level wastes are concentrated but are stored in the acid state.

d. High-Level Waste (multicurie level). These wastes result from head-end treatments, in which a large fraction of the activity in the feed to an extraction process, is removed by a scavenge cake, and which result from first-cycle raffinates. Again, in plants processing natural uranium these wastes are neutralized and concentrated before storage, whereas at plants processing enriched uranium they are concentrated but stored as acids.

2. *Storage of Liquid Wastes*

a. Storage Tanks—High-Level Waste. Neutralized high-level wastes can be stored in reinforced concrete tanks lined with steel. Acid wastes are stored in thin-walled, stainless-steel tanks inserted in a secondary concrete tank. For volumes to be handled at separation plants, the tanks are very large ranging in capacity from 300,000 gallons (acid waste) to over 1,000,000 gallons (neutralized waste). For the purpose of shielding, the tanks are usually placed underground. An annulus between the stainless-steel and concrete tanks, provides a means for prompt detection of leaks. The concrete and steel construction for these facilities, requires the highest quality of materials and workmanship.

Means should be provided for temperature, pressure, liquid-level, and radiation measurement, as well as sampling. In some cases, provisions for cooling, liquid transfer, and agitation, are necessary.

At production plants, the delivery of wastes to individual tanks is made through a diversion box. The diversion box is the junction point for a manifold pipe system leading to the various tanks. Routing of wastes to specific tanks, is accomplished by means of jumpers which are placed in the diversion box and attached and detached remotely.

All long runs of pipe which carry high-level waste, are placed in concrete troughs with suitable provisions for expansion and movement of pipes caused by temperature changes. The troughs drain to the diversion box, which in turn may drain into a catch tank from which contaminated leakage can be readily delivered to the storage tanks.

The major problems to be overcome in storing high-level wastes are the following:

(1) *Cooling.* High-level waste may generate heat of up to 10 B.t.u./hr./gal., due to absorption of nuclear radiation. Since tanks are in the million-gallon capacity range, millions of B.t.u./hr. must be removed. In addition, acid wastes must frequently be held at low temperatures in order to minimize corrosion.

(2) *Corrosion.* As indicated earlier, some wastes must be stored in the acid condition, primarily because they would corrode mild steel, even if neutralized. Intense radiation fields produce free radicals in aqueous systems, which can affect corrosion rates. Fortunately, most wastes are oxi-

dizing systems in which peroxide radicals are formed, producing a corrosion-inhibiting oxide layer on metallic surfaces. Reducing systems, however, such as wastes with high chloride content, would tend to induce accelerated corrosion in radiation fields. Where large temperature differences may produce thermal stresses, stress corrosion must be considered.

Cooling coils can be provided in waste tanks for heat removal. Potential coil failure with cooling water contamination, may be regarded as the disadvantage of this method.

Waste can be permitted to boil within the tanks and the evolved vapors condensed and disposed of externally, or returned as liquid to the tank. Unstable conditions can result from this practice, however, for some fission products will concentrate in sludges at the bottom of the tank, and heat generated here can build up as superheat. Eventually the unstable system is disturbed, initiating rapid boiling, and steam is suddenly released at a rate in the order of ten to twenty times the normal rate. Temperatures as high as 176°C. have been observed at tank bottoms (A1).

Contamination control requires that the vapors leaving the storage system be essentially free of activity. Although storage tanks are of massive construction, they have limited resistance to pressures much above or below atmospheric pressure. The tanks, therefore, should be designed to provide adequate relief for both positive and negative pressures.

b. Storage Tanks—Low-Level Waste. If many, very large tanks are to be installed at a site, it is sometimes more economical to build low-level tanks of the same general type of construction as the high-level tanks previously described. Usually, however, cooling provisions will not be necessary. In general, low-level, waste-tank construction should be such as to insure containment of the materials of the chemical nature under consideration, for long periods of time. Provisions should be made for withdrawal of the waste at a later date for possible disposal to the environment. Means should be provided for leak monitoring, and radiation and liquid-level measurement. Usually the tanks will be installed underground, although if the levels of radioactivity are low and the tanks are in a remote location, above-ground storage can be considered. Low-level waste tanks have been constructed of mild steel and of prestressed concrete.

3. *Concentration of Liquid Wastes*

a. Evaporation. Principles. The limits to radioactive decontamination of liquids by evaporation, are determined by the carry-over of activity by entrainment, splashing, and foaming, and the carry-over of volatile radioactivity. Volatiles will be considered in the section on gaseous wastes.

The mechanism of liquid carry-over from boiling liquids in evaporation, has been studied by Vorkauf (V1), O'Connell and Pettyjohn (O1), and by Manowitz *et al.* (M2). A brief review of the theory follows:

Carry-over can be caused by: (1) entrainment of small liquid droplets by the moving vapor; (2) splashing of the violently boiling liquid directly into the vapor vent line; and (3) foaming of the solution to the extent that foam is carried over into the vapor vent line.

In general, entrainment contributes less to carry-over contamination than does splashing or foaming. Entrainment results when liquid droplets, formed at the surface of the boiling liquid, or formed by bubble breakage, are thrown into the vapor spaces above the boiling liquid and are carried by the vapor stream if their rate of fall is less than the vapor velocity.

O'Connell and Pettyjohn noted that entrainment concentration in the overhead, decreases with increase in heat flux at constant boiling point up to a certain heat flux, but that continued increase in the heat flux caused the solution to boil so violently that liquid is splashed overhead. Both the entrainment rate and the vaporization rate increase as the heat flux increases. However, at low vaporization rates the entrainment rate increases less rapidly than the vaporization rate, so that the concentration of entrainment in the vapor decreases. O'Connell *et al.*, and Manowitz *et al.*, concluded that in a given evaporator there will be an optimum heat flux to give a minimum carry-over. Splashing can be largely eliminated by proper evaporator design; foam carry-over, however, is quite unpredictable. The specific solution to be evaporated must be examined for foaming tendencies in each case. Foaming can be controlled usually by one or more methods, such as dilution, use of a water spray, use of antifoam agents, reduction of the boil-up rate, or installation of a steam-coil foam-breaker in the vapor space. Even moderate foaming, such as is always encountered in the evaporation of dilute radioactive waste, leads to relatively high entrainment carry-over, probably by the mechanism of bubble breakage. Whatever mechanism projects liquid droplets into the vapor space, may also project any suspended solids present. Although suspended solids, such as rust, normally are not considered noxious contaminants, in this case they are likely to have adsorbed considerable amounts of radioactivity, and if carried over into the condensate, will impart radioactive contamination to it by subsequent desorption.

Liquid droplets or foam bubbles containing steam thrown into the vapor space, have an initial size distribution. This distribution may be considerably altered by conditions in the vapor space, e.g., droplets may evaporate until the droplet, surface-tension forces are equalized by the vapor pressure. The size distribution of suspended solids depends upon the size distribution in which they are initially present in the solution.

All particulate matter in the vapor space above the boiling liquid may be considered to belong to one of three groups:

(1) Particles whose rate of fall is so small that they are carried out of the evaporator by the vapor stream.

(2) Particles whose rate of fall is roughly equivalent to the vapor velocity in the evaporator vapor space. It would be expected that particles in this size range would build up to some equilibrium concentration within the vapor space of the evaporator.

(3) Particles whose rate of fall is greater than the vapor velocity. These particles will fall back into the boiling liquid if sufficient vapor space is provided within the evaporator for them to reach the end of their trajectory, or if a suitable baffle is so placed as to deflect the particles back down into the boiling liquid. It is understood that if these particles reach a path of sufficiently high velocity, they may leave the evaporator (splashing). It is readily apparent, then, that even when foaming and splashing conditions are kept under control, the vapor stream leaving any evaporator can and does carry along foam bubbles and entrained liquid and solid particulate matter, and that the concentration, amount, and particle-size distribution of such carry-over, will vary from evaporator to evaporator, and will depend upon the specific liquid being evaporated.

The mechanisms by which entrained particulate matter is removed from a vapor stream have been discussed by Rodebush *et al.* (R3, R2), and by Langmuir and Blodgett (L1). They are direct interception, gravity, inertia, diffusion, and electrostatic forces. In general, the entrainment removal efficiency of devices depending upon impaction and inertial effects, such as cyclones, bubble-cap or Raschig-ring columns, will be good for average particle diameters 100 μ or more, but poor for particles of 10 μ or less in diameter. To remove submicron particles efficiently from vapor streams, filtering media such as Fiberglas or fine-pore filter paper have been used. Here the removal mechanism is predominantly one of diffusion, although for some filter material, electrostatic forces may play an important part (R3).

In general, if particulate matter is in the size and density range where it can be removed by inertial effects, one can expect to find an increase in filtering efficiency with an increase in vapor velocity. On the other hand, a decrease in filtering efficiency with an increase in vapor velocity, usually indicates that diffusion is the primary mechanism of particle removal. Increasing the vapor velocity would have little effect on the filtering efficiency due to direct interception.

Manowitz *et al.*, propose the following equations for a submerged coil evaporator approximately 29 inches in diameter with approximately 2 feet of free board: (D.F. = Decontamination Factor)

$$\text{D.F.} = 4 \times 10^{3}\, G^{0.75} \text{ (low boil-up rates to splash point)} \qquad (1)$$
$$\text{D.F.} = 1 \times 10^{10}\, G^{-3.5} \text{ (above splash point)} \qquad (2)$$

where G = boil up rate, lb./(hr.)/(sq. ft.) of disengaging area

For the case of an evaporator with a restriction or baffle, the coefficients in the above equations would be higher, and the exponents would remain about the same.

Foaming would also alter the values of the coefficients and would alter the splash point. If the foam is such that all bubbles break at the surface, Eq. (1) should hold up to the splash point, except that the coefficient should be much lower. If hollow-foam bubbles are readily carried into the vapor space, the splash point should occur at lower boil-up rates than occur in the same equipment without foaming. The splash point should be a function of the height of the vapor space.

For the special case, involving the evaporation of a solution of radiochemicals in the presence of suspended solids, upon which the activities in the solution are readily adsorbed and desorbed (in general, fission products will behave this way), the following equations should be of interest:

$$\text{D.F.} = 42\, G^{.9} \text{ (up to the splash point)} \qquad (3)$$
$$\text{D.F.} = 2.9 \times 10^{5}\, G^{-1.3} \text{ (beyond the splash point)} \qquad (4)$$

It should be noted that these decontamination factors are appreciably lower than those for liquid entrainment. The decontamination factors alone are defined as the ratio of fission product activity in the still pot to the fission product activity in the condensed vapor. The above equations apply simultaneously with Eqs. (1) and (2), but only to that portion of the activity which is strongly adsorbed on the suspended solids.

Few data are available on the efficiency of de-entrainment devices. Cyclone separators, bubble-cap columns, or packed columns would not be expected to give additional decontamination to a stream leaving an evaporator by more than a factor of ten, since these devices will remove only particulates down to the 5–10 micron range. Fiberglas filters, however, will give greater decontamination factors. Manowitz *et al.*, propose the following equation to predict the behavior of unbonded 20 μ diameter Fiberglas filters, packing density $5\#/\text{ft.}^3$

$$\text{D.F.} = 940 \left(\frac{h}{u}\right)^{0.65} \qquad (3)$$

where h = filter height (ft.)
and u = vapor velocity through the filter (ft./sec.)

Metallic wool, steam filters have also demonstrated good performance, but they would not be expected to yield as high a D.F. as a glass wool fiber unless the metallic wool could be obtained in fibers 20 μ in diameter or less, or unless fine glass wool fibers were interwoven in the metallic wool.

Evaporators are used extensively for evaporating liquid wastes at AEC production and research sites.

b. Ion Exchange. (1) *Principles.* When an aqueous solution containing cation M^{+n} is brought into contact with an ion exchange resin, M^{+n} ions in the solution exchange with those cations originally held on the resin by its free acid groups. This may be indicated as a reversible reaction (R_nM being the resin anion and H^+ its original cation)

$$n\,RH + M^{+n} \rightarrow R_nM + n\,H^+$$

The final equilibrium concentrations of M^{+n} and H^+ depend, as in any chemical reaction, principally upon the activities of the two cations and upon the respective affinities of each for the resin. In general, the cation-resin bond strength increases with increasing charge on the cation, and decreases with increasing radius of the hydrated ion. A typical series, from strong to weak, is: Th, La, Ce, Rare Earths, Y, Ba, Cs, Sr, K, NH_4, Na, H.

To free an adsorbed cation from its resin-bound state requires its physical replacement by another cation. If the relative affinity for the resin of the supplanting cation and/or its concentration in the solution phase, is sufficiently greater than that of the ion originally combined with the resin, replacement will be nearly complete. The effective concentration of an ion may be lowered by complex formation. Thus, any cation may be effectively replaced from its resin compound by a relatively dilute solution of a second cation containing a compound which will complex the other cation. In a similar manner, anion exchange resins can be used to remove fission product anions from dilute aqueous solutions. The ion exchange process is, in many cases, strongly pH and temperature dependent.

Colloids, for example, oxides with no ionic charge, are removed by ion exchange processes. Zirconium oxides tend toward sal formation, and in this colloidal form are difficult to remove from solution.

(2) *Applications.* The following important parameters should be considered in deciding whether or not to apply ion exchange processes:

(*a*) *Solids content.* Since extraneous solids will only tend to blanket the ion exchange surface, ion exchange processes should be applied only to aqueous solutions whose total solids content is less than 1%.

(*b*) *Chemical nature of waste.* Since ion exchange is a chemical process, the chemical nature of the feed must be controllable and constant. Changes in pH and the presence of complexing agents will markedly affect the efficiency of the process. The presence of trivalent salts of large, hydrated-ion radius (Al^{+3}) as noncontaminants, and the presence of contaminants in colloidal form (Zr) are undesirable.

(*c*) *D.F. desired.* In general, decontamination factors in the order of 100 can be obtained with mixed ion exchange beds (both anion and cation resins). Under optimum conditions D.F.'s as high as 10^5 can be obtained.

(*d*) *Engineering characteristics.* Consideration must be given to the need for regeneration and what to do with the regenerant. The use of ion exchange introduces a solids handling problem. Higgins (H3) has devised an ingenious contactor which makes the operation mechanically feasible for remote control operations. In principle, the contactor permits continuous counter-current extraction of the fission products. The only moving parts are the valves and the resin bed itself. Eventually, however, the resin itself must be disposed of. For high-level wastes, radiation effects on the resin must be considered. In general, these will result in lowered resin capacity, although in some cases radiation will cause physical breakdown of the resin.

Walters *et al.* (W1), have proposed an electro-deionization process for radioactive waste treatment. The process consists of partial deionization in a multi-compartment, permselective, membrane cell, and is similar to the production of potable water from sea water. This first step is followed by final deionization of the partially demineralized effluent in a multi-compartment, permselective, membrane cell employing mixed anion exchange and cation exchange granules in the deionization compartment. This process would permit higher volume reductions than for chemically regenerated ion exchangers.

c. Precipitation. (1) *Principles.* Several different mechanisms have been proposed to explain the carrying of trace elements by precipitation. These mechanisms are not mutually exclusive, and one, two, or more may be operative in any particular instance. The simplest ideal type of carrying arises when a trace element is incorporated into the crystal lattice of the carrier precipitate, a solid solution being formed. This will occur, in general, when the precipitated salt of the trace element and of the carrier are isomorphous, and the ions of the two elements are not greatly different in size. For good carrying (high D.F.), it is desirable that most of the trace element should go into the precipitate with only a small proportion remaining in solution. This condition can be realized, in general, if the compound of the carrier which is precipitated is more soluble in the aqueous solution than is the corresponding compound of the trace element. For example, lead sulfate is somewhat more soluble in water than is barium sulfate, and consequently lead sulfate is a good carrier for barium. Similarly, if complex-forming anions are present in the solution and the carrier element forms complexes in solution, but the trace element does not, the carrying will be enhanced.

Many precipitates, especially when freshly formed, have large surface areas and so appreciable amounts of trace elements may be carried by adsorption. It appears, as a rough general rule, that highly charged positive ions are more readily adsorbed than those of lower charge.

Frequently the carrying can be improved by having an excess of the anion present. Because of this effect the order of addition is important and leads to a precipitation technique called "reverse strike." In this technique the solution of carrier and trace element are added to a solution of the precipitating agent, as for example, adding a solution containing rare earths as trace element, and ferric ion as a carrier to a caustic solution. The precipitate is always formed in the presence of hydroxyl ion so that adsorption of the trace element is favored.

If carrying is principally by adsorption, other conditions, such as concentrations, rate of mixing, temperature, and presence of interfering substances, may affect the efficiency of the process.

(2) *Applications.* The following important parameters should be considered in deciding whether or not to apply precipitation processes:

(*a*) *Solids content.* Selective precipitants may sometimes be found for the removal of radioactive constituents even in the presence of large amounts of inert salts. Therefore, precipitation methods should be considered for decontaminating systems with high solids content.

(*b*) *Chemical nature of waste.* Precipitation processes will be sensitive to changes in the chemical nature of the feed.

(*c*) *D.F. desired.* In general, D.F.'s of 10 to 100 can be obtained with single stage precipitation processes. Multiple stage processes will give D.F.'s in excess of 1000 and D.F.'s in the order of 10^5 can be obtained in the precipitation process is combined with an acid adsorption or ion exchange process.

(*d*) *Engineering characteristics.* In a highly radioactive system, filtration and solids handling are complex operations.

Rupp (R5) mentions the following precipitation methods for mixed and specific fission products:

(1) Ferrous sulfide, ferrous hydroxide, calcium phosphate precipitation of Cs, rare earths, Sr, and Ru.

(2) Precipitation of Cs with silicotungstic acid.

(3) Ferrocyanide precipitation of Cs.

(4) Alkaline carbonate precipitation of alkaline earths and rare earths.

(5) Sulfide precipitation of Hg, Ru, and Te.

(6) MnO_2 precipitation of Ru.

(7) Selective precipitation of Ru with ferric hydroxide at pH 2.5.

(8) Tetraphenyl arsonium nitrate (TPAN) precipitation of technitium.

(9) Rare earth oxalate precipitation.

Ruthenium precipitation techniques are useful in head-end treatments to solvent extraction processes. Scavenge cakes resulting from such operations often constitute major activity fractions of high-level wastes.

Selective Cs and Sr precipitation methods are useful in trying to separate these long-lived elements from the rest of the fission products, in order to permit more rapid disposal of the short-lived residue.

Glueckauf and Healy (G3) have developed a process for Cs and Sr removal which involves precipitation on anionic resins in hydroxide form. This method of precipitation was found to give a good separation of Cs from the rest of the fission products, but the strontium was largely retained, at least in the presence of large amounts of iron contained in the fission product solutions.

4. *Dispersal of Liquid Wastes to the Environment*

The practice of dispersing some liquid waste directly to the ground has been followed at Hanford, Oak Ridge, and at Chalk River. As pointed out by Brown *et al.* (B5), the factors requiring evaluation in considering the feasibility of disposal of radioactive wastes to the ground include:

(1) The chemical and radiochemical content of the waste.

(2) The effectiveness of retention of 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 liquid disposal.

(4) The natural rate and direction of movement of the ground water from the disposal site to public waterways, and possible changes in these characteristics from the overall 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 concentration (and total quantities) in public waters of the radio elements concerned.

Ground disposal has been practiced in a variety of subsurface sumps ranging from open trenches and ponds to subterranean caverns. The various fission products are selectively adsorbed by soils. In general, the order of retention will be: Pu, Rare Earths, Sr, Cs, Ru, NO_3^-. At Oak Ridge, ruthenium and nitrates were found in sampling wells 85 feet distant from injection points six weeks after the introduction of wastes. Approximately 2×10^8 gallons of active wastes containing several hundred thousand curies of fission products, have been stored in such sites as

Hanford in the first decade of operation (B5). As a result of their pile accident, the Chalk River plant disposed of over a million gallons of liquid waste containing 10^4 curies of long-lived fission products to the ground (M3). At Oak Ridge, thousands of curies of fission products have been disposed of in open waste pits (S2). The specific experiences at these locations, however, cannot be applied to other locations without field and laboratory evaluation of local problems.

At any particular site, surface disposal (up to 1000 feet depth), and deep disposal (10,000–15,000 feet) should be considered. If present on the site, salt domes would be good disposal areas. Cavities can be made in salt either by solution or by mining. In general, no ground water movement occurs through salt, which is also plastic and therefore probably freer from earthquake hazards than most other minerals.

Shale cavities have a wide distribution throughout the country and are low in excavation cost. However, they have many porous zones and little is known of their stability.

Deep disposal may be feasible, but an extensive study should be carried out to insure that conditions in areas under consideration are known to be static; however, the expenses for study, drilling, and monitoring, are likely to be prohibitive.

Although ocean disposal is feasible for small amounts of relatively low-level waste, the transportation expense and the unknown factors involved in dispersing a contaminant to a system from which it may later be extracted by marine life, are such as to make this practice inadvisable.

B. GASEOUS WASTES

1. *Stack Dispersal*

The stack is frequently the most effective way of disposing of gaseous wastes. A properly designed stack can make the difference between a safe plant and a plant in which a combination of weather and plant operations may cause costly atmospheric pollution. As has been emphasized earlier, it is imperative that a complete climatological picture be obtained for the site. The meteorologist, from a knowledge of atmospheric diffusion, the terrain, the climatological picture, and the nature and concentration of the radioactive contaminant, determines what the effective stack height should be. The effective stack height is the stack height plus the height above which the effluent plume rises from the stack, owing to the stack draft velocity and/or the buoyancy of the effluent. The design and choice of height for the stack itself, however, is an engineering problem.

A rule of thumb frequently used to estimate effective stack height for low wind speeds, is the result of the investigations of O'Gara and Fleming

(O2). It states that each degree Fahrenheit of smoke temperature above ambient air, is equivalent to 2½ ft. of extra stack height.

The atomic energy industry encounters situations in which the effects of both smoke temperature and velocity must be considered, and for this reason a more exact approach is necessary. When meteorological parameters, such as vertical temperature gradient and wind speed, are taken into consideration as well, the problem becomes quite complicated. The only attempt to take all of these parameters into account is that of Bosanquet *et al.* (B4).

A simpler approach than that of Bosenquet is afforded by an equation empirically derived by Davidson (D1) from the wind tunnel experiments of Bryant (B6). It does not consider the effect of the vertical gradient of air temperature, but this does not eliminate it in favor of the Bosenquet formula, since the effect of air stability on effective stack height is not definitely known for all conditions. The Bryant-Davidson expression is

$$h = d\left(\frac{V_s}{U}\right)^{1.4}\left(1 + \frac{T}{T_s}\right)$$

where h is the rise of the plume above the stack, d is the stack diameter (ft.), V_s is the stack draft velocity (ft./sec.), U is the mean wind speed (ft./sec.) and T and T_s are the excess and absolute temperature of the stack gas in similar units, respectively.

In engineering terminology, the stack draft often is expressed in pounds per second, since this quantity is invariable for different atmospheric pressures and effluent densities. The effluent velocity (V_2) varies considerably with such values, however, for any fixed discharge rate and stack diameter.

Assuming a specific volume of 12.4 cu. ft./lb. of air at 30 in. Hg and 32°F., it can be shown that

$$V_s = \frac{0.962DT_s}{d^2P} \cong \frac{DT_s}{d^2P}$$

where D is the discharge rate in pounds per second, and P is atmospheric pressure in inches of mercury. This expression is valid when the effluent is mostly air, as in the case of most atomic energy plants.

2. *Equipment for Particulate Removal*

Requirements for air and gas cleaning equipment in the atomic energy field differ from those for conventional equipment in the following respect: (a) cleaning efficiencies are much more severe; (b) equipment life is of prime importance and should be designed for as long as possible; (c) minimum maintenance is necessary; (d) consideration must be made for disposal of the equipment itself or for the components thereof.

The problems to which particulate removal equipment must be applied, include the treatment of reactor cooling gases, the treatment of cell ventilation air, the treatment of dissolver exit gases, and the treatment of incinerator flue gases.

Reactor cooling gases should be treated both at inlets, to decrease particulate matter exposed to neutron fluxes, and at outlets to remove particulates resulting from corrosion, erosion, or fuel element rupture. Operating temperatures sometimes prove a limitation for this application. The treatment of dissolver exit-gases imposes acid fume resistance as a condition in the treatment equipment.

The limit to the effectiveness of a gas cleaning device is its efficiency in the removal of aerosols. Neither electrical nor thermal precipitation has proven practical for the rapid removal of aerosols, and filtration seems to be the best method of removal. Aerosol filters consist of loosely aggregated fibers, and in order to avoid excessive resistance to flow of gas, the mesh of the fiber must be larger than the size of the particle to be removed. There is therefore no screening action; the removal of the particle depends entirely upon a chance collision of the particle with a fiber of the filter. Once having collided, the particle adheres by adsorptive forces.

Very large particles can be precipitated by centrifugal action, as in a cyclone separator. For smaller particles whose diameter is in the order of several microns, the centrifugal action is no longer effective since the inertia of the particle is not sufficient to overcome the resistance of the air. Thus, the air flows around the fibers of a filter in stream lines, and the particles are carried around with these stream lines. There is a range of particle sizes for which a higher velocity will improve the operation of the filter, since the inertial effects will carry the particles across the stream lines into collision with the fibers of the filter. For particles whose diameter is smaller than 1 micron, no inertial effects exist, but the kinetic diffusion becomes of greater importance in such smaller particles. Very small particles (0.01 μ) are precipitated very rapidly by diffusion. The process is analogous to the condensation of a vapor on a cold surface. The particles most difficult to remove by filtration, are those in the range of 0.1 to 1.0 microns, i.e., smokes. In order to obtain efficient filtration without excessive resistance, the filter must contain fibers of small diameter, approaching that of the particles themselves.

The high efficiency, asbestos-cellulose filter (C.W.S. filter) is one of the best developed for aerosol removal. This filter, which shows a decontamination factor of nearly 5×10^3 for 0.3 μ particles, improves in performance within a short time after the beginning of service, because of particulate deposition. Usually a glass wool pre-filter is used before the CWS filter, to protect it and to increase its life. One of the major weaknesses of the cellulose-asbestos filter is its inability to withstand more than small amounts of corrosive gases, and its limitation to temperatures below 100°C.

Glass fiber filters have been used for handling dissolver exit gases. This application has involved the use of chemically resistant glass in various diameters from 25 μ to 1 μ placed in layers of varying density and thickness. Studies by Blasewitz and Judson (B3) give performance and life characteristics for pilot studies.

A summary of the operational characteristics of many types of air cleaning equipment has been made by Silverman (S1) and is reproduced in Table II.

3. *Equipment for Vapor Removal*

Radioactive gases created by neutron flux activation, such as A^{41}, and certain radioactive fission product gases (Xe, Kr), are not easily removed by conventional approaches. The noble gases may be condensed and adsorbed on activated charcoal at extremely low temperatures. The cost of such systems per cubic foot of treated air, is so high that the method is feasible only for small volumes. Another approach for such volumes is compression and storage of the gases in chambers, for times of sufficient length to permit these isotopes to decay.

Methods have been developed for the removal of I^{131} from dissolver exit gases. Scrubbing with spray or baffle-plate scrubbers and utilizing caustic adsorbents, yields efficiencies of only 60 to 80%, and removes some oxides of nitrogen simultaneously, through neutralization by part of the caustic. Use of a silver reductor unit, composed of a tower packed with saddles that are coated with silver nitrate and maintained at an elevated temperature, gives the highest removal of iodine compounds (S1). Tests with this reactor have resulted in iodine removal with efficiencies greater than 99.99%.

C. DISPOSAL OF SOLID WASTES

1. *Collection and Packaging*

Solid wastes can be categorized into three classes—(1) contaminated equipment, (2) contaminated nonburnable trash, and (3) contaminated burnable trash.

Occasionally contaminated equipment, such as stainless steel tanks or pumps, which must be disposed of, can be transferred to the burial ground without secondary packaging. More frequently, however, the equipment is first packaged in a metal-lined, airtight wooden box, or in a plastic bag. This is necessary in order to prevent the leakage of contamination when

TABLE II

OPERATIONAL CHARACTERISTICS OF AIR CLEANING EQUIPMENT

Type of equipment	Particle size range, mass median, microns	Per cent efficiency for size in column 2	Velocity, ft./min.	Pressure loss, inches of water	Approximate Cost/CFM ($)	Current Application in U. S. Atomic Energy Program
Simple settling chambers	50	60 to 80	25 to 75	0.2 to 0.5	0.05	Rarely used
Cyclones, large diameter	5	40 to 85	2000 to 3500 (entry)	0.5 to 2.5	0.10 to 0.25	Precleaners in mining, ore handling, and machining operations
Cyclones, small diameter	5	40 to 95	2500 to 3500 (entry)	2 to 4.5	0.25 to 0.50	Same as above
Mechanical centrifugal collectors	5	20 to 85	2500 to 4000	—	0.20 to 0.35	Same as large cyclone application
Baffle chambers	5	10 to 40	1000 to 1500	0.5 to 1.0	0.05	Incorporated in chip traps for metal turning
Spray washers	5	20 to 40	200 to 500	0.1 to 0.2	0.10 to 0.20	Rarely used; occasionally as cooling for hot gases
Wet filters	gases and 0.1–25 μ mists	90 to 99	100	1 to 5	0.09 to 0.10	Used on laboratory hoods and chemical separation operations
Packed towers	gases and soluble particles	90	200 to 500	1 to 10	0.40 to 0.80	Gas absorption and precleaning for acid mists
Cyclone scrubber	5 μ 5	40 to 80	2000 to 3500 (entry)	1 to 5	0.25 to 0.40	Pyrophoric materials in machining and casting operations, mining and ore handling; roughing for incinerators

Inertial scrubbers, power driven	8 to 10	90 to 95	—	3 to 5 HP per 1000 cfm	0.15 to 0.25	Pyrophoric materials in machining and casting operations, mining and ore handling
Venturi scrubber	1	99 for H_2SO_4 mist; SiO_2, oil smoke, etc., 60 to 70	12,000 to 24,000 at throat	6 to 30	0.50 to 3.00	Incorporated in air cleaning train of incinerators
Viscous air conditioning filters	10 to 25	70 to 85	300 to 500	0.03 to 0.15	0.004 to 0.006	General ventilation, air
Dry-spun glass filters	5	85 to 90	30 to 35	0.1 to 0.3	0.02 to 0.04	General ventilation air; precleaning from chemical and metallurgical hoods
Packed beds of graded fibers 1 to 20 μ, 40 inches deep	1	99.90 to 99.99	20	10 to 30	1.0 to 5.0	Dissolver, exit gas cleaning
High efficiency cellulose-asbestos filters	1	99.95 to 99.99	5 through medial, 250 at face	1.0 to 2.0	0.04 to 0.06	Final cleaning for hoods, glove boxes, reactor air and incinerators
All-glass web filters	1	99.95 to 99.99	5 through medial, 250 at face	1.0 to 2.0	0.07 to 0.10	Final cleaning for hoods, glove boxes, reactor air and incinerators
Conventional fabric filters	1	90 to 99.9	3 to 5	5 to 7	0.30 to 1.00	Dust and fumes in feed materials production
Reverse-jet fabric filters	1	90 to 99.9	15 to 50	2 to 5	0.50 to 1.00	Dust and fumes in feed materials production
Single-stage electrostatic precipitator	1	90 to 99; 90 to 95 on metallurgical fumes	200 to 400	0.25 to 0.75	0.50 to 2.00	Final cleanup for chemical and metallurgical hoods; uranium machining
Two-stage electrostatic precipitator	1 to 5	85 to 99	200 to 400	0.25 to 0.50	0.25 to 0.50	Not widely used for decontamination

the equipment is transferred to the burial pits. Transportation is by means of railroad truck, as will be described later.

Contaminated nonburnable trash, is put into convenient containers that can be sealed. These can be boxes, cartons, or drums. Frequently 50-gallon steel drums are used. Drums are sometimes filled with an annular layer of concrete to provide some shielding for very active trash contained in the void space of the drum.

Burnable trash is sometimes packaged in cardboard containers but more frequently is compressed and baled. It is often more economical to bury baled burnable trash than to incinerate it.

2. *Incineration*

Because of the complexity of solids-handling equipment, incineration will be considered for low-level waste only, and not for high-level, burnable trash.

Because of the hazards of handling radioactivity, special consideration should be given to the following features of incinerator design: (a) packaging, (b) feed device, (c) combustion chamber, (d) fly ash and particulate removal, (e) ash removal.

For the convenience of the operator, waste should be packaged in uniform-sized packages designed for ease of handling. Asphalt-lined bags should be avoided because of the evolution of excessive particulate matter from their incineration. Fiberboard cartons or drums are generally satisfactory.

The feed chute must lead through some system of interlocked, double doors to prevent blowback of radioactive ash or fumes from the furnace.

The primary requisite of the combustion chamber is the insurance of complete combustion. If air is used to support combustion, the air supply should be drawn from somewhere outside the building to eliminate the possibility of radioactive dust being dispersed through the building in the event of a puff resulting from momentarily accelerated combustion. Remote ignition should be provided within the combustion chamber.

Both large and fine particulates of fly ash must be removed from the flue gas. A rough scrubber, such as a Pease-Anthony, will suffice for the large particulates, although the scrub solution, of course, constitutes a liquid waste. Fine particulates can be removed by Fiberglas filters or CWS-type filters.

The ashes must be removed into a system in which dusting will be minimized. Collection in sealed receivers or collection under water may be used.

Decontamination factors of about 10^9 are frequently required for incinerators. This degree of decontamination can be obtained with the equipment described above. However, the necessary storage of filters and scrub solutions is a disadvantage of this method.

Incinerators are not widely used by the AEC.

3. *Burial on Site*

In many cases, the most economical method of disposing of solid wastes is by burial on site. For large atomic energy installations, or possibly for regional burial grounds, the following criteria should be observed.

The estimated life of the burial ground should be at least 100 years. Obviously, it will be advantageous if the area can be located in an empty section of the country. At least one square mile of burial area plus one-half square mile of fringe area, should be sufficient to meet the 100-year limitation.

The geology of the area should be known in as much detail as possible. There should be as few faults as possible, with no history of seismic disturbances. Limestone formations should be avoided. The land should be flat with possibly one hill for air dispersion, and there should be tree and shrub coverage to keep dust down.

Low rainfall in the area is preferable, as is a low ground water table. Ground water and surface water hydrology must be carefully studied. Such a study would probably result in a well-survey system, which could also be used to monitor the burial grounds.

The Oak Ridge National Laboratory has been burying solid wastes on site for many years. Trenches 10 × 200 × 15 feet deep, are dug. Packaged trash and equipment, can be transferred into the trenches from the backs of trucks, by a crane. The trenches are then backfilled with 3 to 4 feet of earth. Occasionally, auger holes 1 foot in diameter and 15 feet deep are dug, for the disposal of especially radioactive pieces of equipment. The use of an auger hole reduces the back-scatter of radiation to the bulldozer operator. The area is gridded and recorded, and this procedure is followed both for permanent marker purposes and for the occasional recovery of valuable equipment after burial.

A description of how the Hanford plant discards "hot" equipment, was published in the May, 1955 issue of *Nucleonics*. The techniques involved are as follows:

A remotely-operated crane in the separations plant lifts the "hot" piece of equipment with attached piping from a cell, places it in a metal-lined wooden box on a railroad flat car, and bolts down a lid. With the hot equipment inside the box, no one can stay within 600 feet of it for more than one-half hour. Ten empty cars separate the locomotive from the 6-ton piece of radioactive "junk" on the trip from the separations plant to the burial ground. Each burial is preceded by a dry run, during which the box is hauled empty from the separations plant to the burial ground, to be sure it will pass all gate posts and power lines safely. If, on the appointed day, the winds blow faster than 10 miles per hour, the operation is postponed. At the burial ground the locomotive stops at a concrete siding at the head of a long trench perpendicular to the rail line. A steel cable attached to the box and looped along the train is then attached to a tractor. The tractor drags the box off the flat car and down a ramp into a trench. The box is then buried by a crane operator who dumps a pile of dirt and rocks into the trench, working from behind a pile of dirt and rocks, and using mirrors at the end of his long boom to see what he is doing.

This description clearly indicates the problems involved in burying high-level solid wastes.

IV. Recovery of Fission Products from Radiochemical Wastes

A. Processes for Recovery of Specific Fission Products

By far the best solution to many of the waste disposal problems discussed, would be waste utilization. The fission products may turn out to be valuable by-products. Quite a bit of research is now going on in the finding of uses for intense radiation fields (M1, B1). Certain of the fission products, particularly Cs^{137} and Sr^{90}, would have advantageous characteristics for such use. Most of the work in developing processes for the recovery of specific isotopes from fission product wastes, has been done by Rupp and his co-workers at the Oak Ridge National Laboratory. The following is a description of the processes that Rupp has selected for a fission product recovery pilot plant to be located at Oak Ridge.

Simplified flow sheets are given in Figs. 2 and 3 for two of the basic types of waste that will be encountered. The Purex type of waste is the simplest of all of the wastes to process, being a nitric acid solution of fission products, corrosion products, and a small amount of other impurities. Advantage is taken of the fact that one can evaporate this waste, and thereby achieve a greatly increased concentration of material, before actual chemical separation of the constituents is started.

The evaporator used for this purpose contains steam coils for heating the liquid and additional heat is injected continuously as steam to supply the additional water required for the distillation of nitric acid, which distills at less than 6 *N*. The limits to which evaporation can be carried, probably are determined by the amount of corrosion products that accumulate in the solution, and the degree of contamination of the condensate from the evaporator. The concentrated waste solution is stored in a tank equipped with cooling coils, and is passed on to the next main

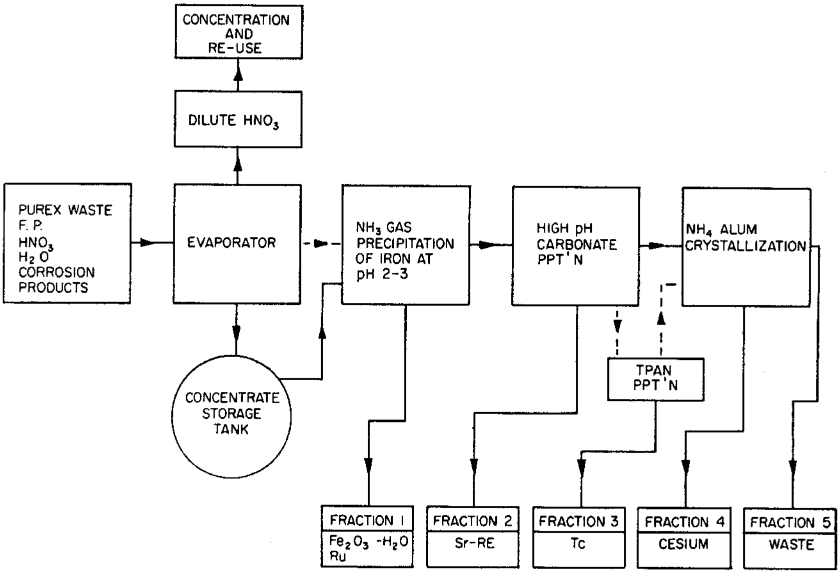
CONCENTRATION AND RE-USE
DILUTE HNO_3
PUREX WASTE
F. P.
HNO_3
H_2O
CORROSION PRODUCTS
EVAPORATOR
NH_3 GAS PRECIPITATION OF IRON AT pH 2-3
HIGH pH CARBONATE PPT'N
NH_4 ALUM CRYSTALLIZATION
CONCENTRATE STORAGE TANK
TPAN PPT'N
FRACTION 1
Fe_2O_3 -H_2O
Ru
FRACTION 2
Sr-RE
FRACTION 3
Tc
FRACTION 4
CESIUM
FRACTION 5
WASTE

FIG. 2.

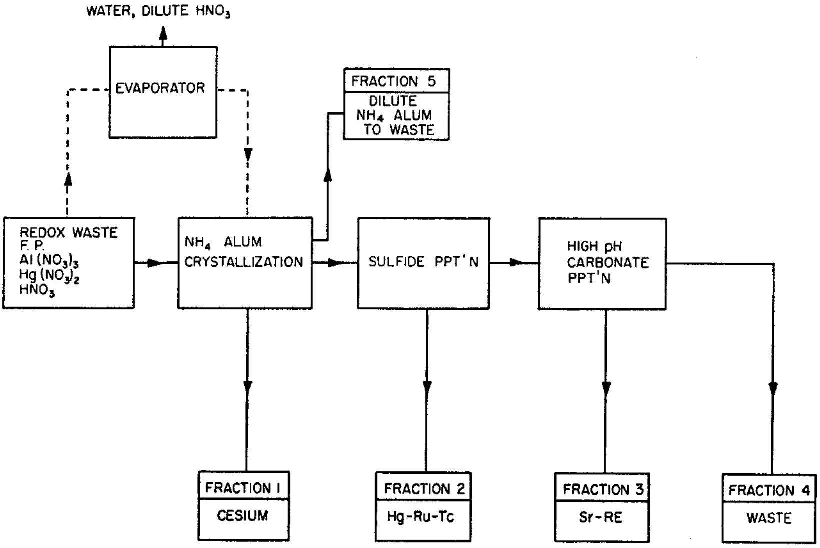
WATER, DILUTE HNO_3
EVAPORATOR
FRACTION 5
DILUTE NH_4 ALUM TO WASTE
REDOX WASTE
F. P.
$Al(NO_3)_3$
$Hg(NO_3)_2$
HNO_3
NH_4 ALUM CRYSTALLIZATION
SULFIDE PPT'N
HIGH pH CARBONATE PPT'N
FRACTION 1
CESIUM
FRACTION 2
Hg-Ru-Tc
FRACTION 3
Sr-RE
FRACTION 4
WASTE

FIG. 3.

step, which is the precipitation of iron at pH 2.5. The precipitation is very similar to the precipitation of iron obtained by the hydrolysis of urea; however, it is more easily accomplished by injecting ammonia gas into the solution, which is vigorously agitated. Most of the iron and ruthenium are brought down at this point, allowing the rare earths, alkaline earths and cesium to pass into the filtrate. A carbonate precipitation is made on the filtrate, bringing down the strontium and rare earths together as fraction no. 2. The solution now contains only cesium, which is taken out in the alum crystallization process as fraction no. 4. If it is desired to remove technetium, a special precipitation with tetraphenyl arsonium nitrate is made and the crude technetium accumulated as fraction no. 3. The slightly contaminated waste that is discharged from the alum crystallizers, is treated before discharge from the plant.

It will be noted from the flow sheets, that the chief difference between the two types of waste processing, is that the Redox waste goes to the alum crystallization first for removal of most of the aluminum in the form of alum. If sulfuric acid is used as the means of sulfate addition in forming the alum crystals, nitric acid is actually generated during the crystallization and it may be feasible to recycle material to the evaporator and remove nitric acid, bringing the concentrated alum back to the crystallizer for another cycle. In this way it may be possible to remove most of the aluminum as the sulfate and the nitrate ion as nitric acid. However, in practice it appears that enough of the aluminum is removed in the first crystallization; so the waste alum solution is passed on to other crystallizers which remove the final traces of cesium. This aluminum waste is then passed on to further treatment to remove traces of strontium and rare earths before discharging from the plant (Fig. 4). The cesium product in this process is accumulated as fraction no. 1 to be concentrated by further fractional crystallization.

Redox-type waste contains considerable mercury, which must be removed. Advantage is taken of the presence of mercury to use it as a carrier for the ruthenium and technetium when this group is precipitated as the sulfides. This involves fairly corrosive chemical solutions, but they can be handled in equipment fabricated with special grades of stainless steel. The filtrate contains only the alkaline and rare earths which are then precipitated as carbonates, the same as the Purex-type procedure. The waste from this step is treated separately.

In the finishing stages of cesium processing, the cesium fraction that is brought to the final crystallization step, represents a large concentration of cesium over that found in the original waste. This is because the cesium can be progressively accumulated in the bed of crystals. By decanting off the supernatant liquor after one crystallization, another batch of raw material containing cesium can be put into the crystallizer and the previous crystals redissolved. Additional ammonium alum is added if necessary, to saturate the solution at 80°C., and a new crop of crystals is grown, this time including all the cesium that was brought down during the previous crystallizations, plus the newly added cesium. As successive crystallizations are made, the crop of crystals becomes progressively richer in cesium.

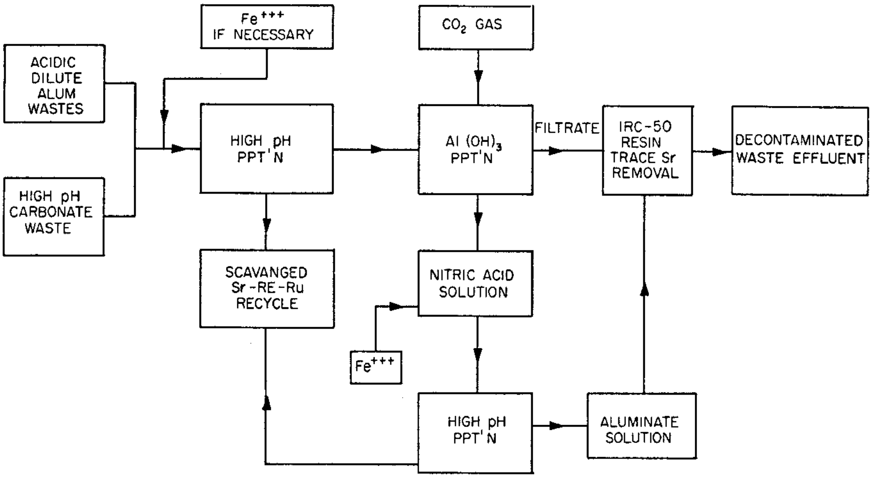

FIG. 4.

This procedure can be followed until the cesium losses in the supernatant liquor become higher than desired. In the semi-works equipment designed for this process, several crystallizers are used, some of which handle the supernatant liquor for recrystallization and recovery of small amounts of cesium that have passed the first crystallization. Others are used to concentrate the cesium by dissolving the alum crystals in water, and then growing smaller batches of crystals to get the cesium into progressively smaller volumes of liquid. If enough cesium is accumulated, pure cesium aluminum alum crystallizes. In this process, Cs^{137} comes down almost radiochemically pure after one crystallization; only insignificant amounts of Ce^{144}, Sr^{90}, and Ru^{106} superficially contaminate the first batches of crystals. However, some inactive fission products, such as rubidium, accompany the cesium and must be removed by selective crystallization near the end of the refining process. At all stages of the process, the various fractions can be recycled to an appropriate previous point in the crystallization cycle, to keep losses at a minimum.

It may seem that crystallization would be a hard procedure to carry out by remote control, but in actual practice with large batches of cesium in 50-gallon crystallizers, this has not been the case. The crystals are of large size, like granulated sugar, and the supernatant liquor can be easily decanted through a screened dip-leg. The remaining crystals are very easily dissolved by adding water and increasing the temperature to 80°C. Speaking in terms of decontamination factors, it is easily possible to obtain a cesium decontamination of 10^2 per crystallization step. Therefore, two successive crystallizations are usually enough to achieve the required decontamination. It has been found that in achieving decontamination factors larger than this, the limiting factor is often not the process itself, but rather the contamination that is picked up from the equipment.

The concentrated cesium alum produced by the crystallization process contains ammonium and aluminum sulfate, which must be removed. The next step is to precipitate the aluminum as the hydroxide, bringing it down in the granular form by the ammonia gas technique. The filtrate contains cesium sulfate and ammonium sulfate and could be transported to a source fabrication plant as a highly concentrated product at this stage.

In the preparation of the sources, the next step is to remove the sulfate by passage of the solution at high flow rates through a bed of anion exchange resin; good control is necessary at this point to avoid too much radiation damage to the highly sensitive anion exchange resin. It has been found possible to do this at the 1000-curie level with an acceptable amount of resin damage. This is the only point in the process where an organic material is used, and work is being continued to find a suitable substitute procedure. The effluent from the column contains cesium hydroxide and ammonium hydroxide; the latter is removed by distillation. The cesium hydroxide is neutralized with hydrochloric acid, and the cesium chloride solution is taken to dryness at 460°C. to remove all traces of water. The dried material is then pressed in a hydraulic press at 20,000 psi to produce pellets which are very compact and hard.

The finishing stages for strontium separation are shown in Fig. 4. The main separation of the rare earths from the alkaline earths is made by ammonia gas precipitation of the rare earths as hydroxides in a carbonate-free medium. The alkaline earths pass into the filtrate and are removed in the next step as the carbonates. Since the separation of rare earth hydroxides and the only moderately soluble alkaline earth hydroxides is not clean, a re-precipitation step is required. The alkaline carbonates are then passed to packaging, either as the dried carbonates, or are first converted to sulfates, oxides, or fluorides for subsequent packaging in multiple-walled, weld-sealed, containers for storage. The filtrate containing some strontium is returned to the plant, waste effluent, decontamination process.

The plant waste streams are kept to a minimum by appropriate recycling, but the waste acidic alum and alkaline carbonate filtrates are further decontaminated, principally from Sr^{90}, by the procedure shown in Fig. 4. This is essentially a high pH scavenge with $Fe(OH)_3$; and is followed by trace Sr^{90} cleanup, by passing the alkaline waste through a bed of IRC-50 resin, which is especially suited for removing alkaline earths from highly alkaline media. Since only trace amounts of radioactivity are present at this stage, the problem of radiation damage to the resin is not serious.

B. PROCESSES FOR THE RECOVERY OF MIXED FISSION PRODUCTS

If a cheap way could be found in which to concentrate mixed fission products and package them in a stable form, these might serve as economical radiation sources. Relatively little work has been done in this area.

One possibility is a process being developed by Hatch and his co-workers (H2). The method involves the adsorption of fission—product ions on montmorillonite clay, and the fixation of the adsorbed ions by heating the clay to a high temperature. The capacity of the clay is found to be about 1 milliequivalent per gram for cation exchange, and a high degree of fixation has been demonstrated. One limitation of the process is that the feed must be free from multivalent inert ions to keep from blanketing the clay with inert material. Investigations are now centering on methods for removing aluminum ions from raw wastes.

Another possibility lies in the use of equipment being developed by Manowitz and co-workers (H2). The equipment, called a continuous calciner, converts aqueous slurries to compact fused salts. One source of fission products of high specific activity would be the MnO_2 scavenge cake used in separations processes, to remove most of the fission products in a head-end treatment prior to a solvent extraction process. MnO_2 may be dissolved in fused caustic in a continuous calciner to form a highly concentrated radiation source.

V. Other Waste Problems

There are occasions when radiochemical wastes are amenable to treatment by biological methods. This may be because the waste contains sewage and must be biologically treated, or because the waste contains organic matter (i.e., laundry wastes) to which a biological treatment may be applicable. The effectiveness of activated sludge in concentrating radioisotopes from liquid wastes has been studied by Kaufman (K1), Christiansen (C1), and others. The use of microorganisms in oxidation ponds has been studied by Gloyna (G1). The removal of radioactivity from liquid streams by trickling filters has been studied by Dobbins (D2) and others.

Occasionally, also, the contaminant of prime concern in a waste stream is the chemical content rather than the radiochemical content. For instance, at uranium (feed) production centers, the fluoride concentration in the waste is the most serious hazard. Liquid wastes of this nature are usually discharged to lagoons or holding ponds rather than to an environmental stream.

VI. Future Problems

Future problems associated with separations and waste treatment processes, will be the result of two major changes in reactor development. These are the use of new materials of construction for fuel cladding or fuels themselves, and the increase in specific activity of processed fuels.

Fuel composition may change from uranium to plutonium, and cladding from aluminum to zirconium to stainless steel. In some cases blankets, moderators, and coolants must be processed, and these will introduce thorium, beryllium, NaK, and bismuth to the chemical process. These changes in materials will present new chemical and corrosion problems in waste treatment processes and waste storage procedures.

An increase in concentration of fission products in the chemical process will introduce many new problems and will magnify many old ones. Much higher decontamination factors will be required in order that process solutions may be safely discharged or re-used. A considerable amount of heat will be generated in the fuel, in the fuel solutions throughout the separations process, and even in the waste streams. The heat generation problem may be complicated by activity adsorbing on walls of containers or on suspended particles, and creating local hot spots within the system.

Accompanying the heat generation problem will be the problem of radiation damage to the fluids in the separation and waste systems, and perhaps to parts of the containers of the systems. Radiation damage to the fluids of the system may include pH changes, oxidation reductions, or gassing. Radiation damage to containers may include embrittling or fracture of gasket and packing material. Corrosion of container walls may be accelerated by radiation decomposition products.

Fission product recovery may alleviate some waste storage problems but will not nullify the need for low-level waste processing. Indeed, any fission product extraction process will, by its inevitable inefficiency, generate low-level wastes. Thus, continued research and development in the field of low-cost waste processing techniques, is probably justifiable.

Another lucrative field for research and development should be that of chemical reagent recovery. The recovery of aluminum nitrate and nitric acid from solvent extraction wastes is now under study. Future separation processes will result in wastes containing bismuth, zirconium, and other critical materials, whose recovery should prove economically feasible.

The development of cheap transportable packages may change the economics of shipping low-level wastes to a regional disposal site.

Several processes intended to reduce the volume of high-level waste and to convert it to a form in which containment is more assured, are now in the development stage. One of these involves the adsorption of fission products on clay, and the subsequent fixing of the activity on the clay by firing (H2). A second involves the conversion of aqueous waste solutions to anhydrous melts by calcining (H2). A third involves the incorporation of wastes into concrete. These processes are presently in the pilot plant stage and it is difficult to assess their final applicability until they have been further developed and tested for a longer period of time.

To summarize, the problems of radiochemical waste disposal are many and varied. Adequate solutions for many of these problems have been applied by the AEC at its own sites, but these solutions may not be adequate for new commercial separation plants. Waste disposal is a field in which ingenuity and imagination can pay off by appreciably improving the feasibility of widespread atomic power.

REFERENCES

A1. Anderson, C. R., and Rohrmann, C. A., Geneva Conference Paper No. 552 (1955).

B1. Ballantine, D. S., *Modern Plastics* **32,** 131 (1954).

B2. Beers, N. R., *Nucleonics* **4,** No. 4, 28–38 (1949).

B3. Blasewitz, A. G., and Judson, B. F., *Air Repair* **4,** 223 (1955).

B4. Bosanquet, C. H., Carey, W. F., and Halton, E. M., *Inst. Mech. Engrs. (London) Proc.* **162,** 355–367 (1950).

B5. Brown, R. E., Parker, H. M., and Smith, J. M., Geneva Conference Paper No. 565 (1955).

B6. Bryant, L. W., Experiments in a Wind Tunnel, National Physical Laboratory (Great Britain), 1949.

C1. Christiansen, C., *U.S. Atomic Energy Comm.* Wash. **275,** Sanitary Engineering Conf., April, 1954, p. 198.

C2. Church, P. E., and Gosline, C. A., *Bull. Am. Meteorol. Soc.* **29** (2), 68–73 (1948).

D1. Davidson, W. F., Ind. Wastes, *Ind. Hyg. Foundation Amer. Trans. Bull.* 38–55 (1949).

D2. Dobbins, W. E., *U.S. Atomic Energy Comm.* Wash. **275,** San. Eng. Conf., Apr. 1954, p. 38.

D3. Dwyer, O. E., Geneva Conference Paper No. 550 (1955).

F1. Ferguson, D. E., Geneva Conference Paper No. 551 (1955).

G1. Gloyna, E. F., and Steel, E. W., *U.S. Atomic Energy Comm.* Wash. **275,** San. Eng. Conf., Apr. 1954, p. 10.

G2. Glueckauf, E., Geneva Conference Paper No. 398 (1955).

G3. Glueckauf, E., and Healy, T. V., Geneva Conference Paper No. 415 (1955).

H1. Hatch, L. P., *Am. Scientist* **41,** No. 3, 410 (1953).

H2. Hatch, L. P., Regan, W. H., Manowitz, B., and Hittman, F., Geneva Conference Paper No. 553 (1955).

H3. Higgins, I., AECD 3809 Mar., 1955. Dev. of the Continuous Ion Exchange Contactor.

I1. Recommendations of the International Commission on Radiological Protection, ICRP/54/4. *Brit. Inst. Radiol.*, London, 1954.

K1. Kaufman, W. J., Klein, G., Greenberg, A. E., *U.S. Atomic Energy Comm.* Wash. **275,** San. Eng. Conf., Apr. 1954, p. 1.

L1. Langmuir, I., and Blodgett, K., OSRD 3460 (1944). Smokes and Filters.

M1. Manowitz, B., *Chem. Eng. Progr. Symposium Ser.* **50,** 201 (1954).

M2. Manowitz, B., Bretton, R. H., and Horrigan, R. V., *Chem. Eng. Progr.* **51,** 313 (1955).

M3. Mawson, C. A., Geneva Conference Paper No. 12 (1955).

N1. *Natl. Bur. Standards (U.S.) Handbook* **52** (1953).

O1. O'Connell, H. E., and Pettyjohn, E. S., *Trans. Am. Inst. Chem. Engrs.* **42,** 795 (1946).

O2. O'Gara, P. J., and Fleming, E. P., American Smelting and Refining Co. unpublished data.

R1. Reid, D. G., Geneva Conference Paper No. 543 (1955).

R2. Rodebush, W. H., *U.S. Atomic Energy Comm.* AEC Handbook on Aerosols, pp. 60–64.

R3. Rodebush, W. H., Holly, C. E., Lloyd, B. A., OSRD 2050 Filter Penetration by Aerosols (1943).

R4. Rodger, W. A., The Handling of Radioactive Wastes. Talk given before the Natl. Ind. Conf. Board (October 1955).

R5. Rupp, A. F., Oak Ridge National Laboratory, Fission Product Processing. Talk given at Oak Ridge Natl. Lab.

S1. Silverman, L., Geneva Conference Paper No. 571 (1955).

S2. Struckness, E. G., Morton, R. J., and Straub, C. P., Geneva Conference Paper No. 554 (1955).

V1. Vorkauf, H., *Forchungsheft* **341** (1931).

W1. Walters, W. R., Weiser, D. W., Mareck, L. J., *Ind. Eng. Chem.* **47,** 61 (1955).