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A conference report is submitted on a general program concerned with environmental contamination, in anticipation of the beginning of operation of a reactor. The purpose is two-fold, to continue and expand present preventative measures and to develop additional procedures which will allay fear and minimize the probability of legal action. Measures considered include studies of local geology for its influence on the movement of ground water; setting up of a network of observation wells and of water-sampling points for both ground and surface water; study of air-dug effluent as a density and dispersion pattern as affected by meteorological conditions; and airborne radio-activity measurements by continuous monitoring. A system for handling liquid waste products from the reactor, the reactor laboratories, and the "hot" laboratory includes facilities for preliminary treatment, disposal, storage, and neutralization.

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DIVISION: Atomic Energy (44)  SUBJECT HEADINGS: Waste, Radioactive - Disposal
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Waste Processing
1. Brookhaven National Laboratory
Waste Problems
March 27-28, 1950
CONFERENCE ON WASTE PROCESSING

Meeting of U. S. Atomic Energy Commission Waste Processing Committee

March 27-28, 1950

Section 1. BROOKHAVEN NATIONAL LABORATORY WASTE PROBLEMS

J. H. Hayner, Permanent Chairman
B. Manowitz, Host Chairman

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BROOKHAVEN NATIONAL LABORATORY
Upton, N. Y.
The continued advance of the national atomic energy program is, to a large extent, dependent upon the hazard-free and controlled ultimate disposal of radioactive wastes. Any radiochemical laboratory or reactor operation and development will produce these waste materials in varying amounts and compositions, constituting a hazard and possible impediment to the optimum development of nuclear energy.

The need for intensive and extended research and development of processes, equipment, and systems has resulted in the establishment of various projects in practically every area.

The Atomic Energy Commission has been concerned with the integration of these projects and with the exchange of information so that there will be constructive planning with a minimum of duplication of effort in future programs. In this connection, on November 10, 1949, at the direction of L.R. Hafstad, Director of Reactor Development, a Waste Processing Committee was established. This group is made up of individuals from each operating contractor's organization where waste processing research and development are under way. The Committee meets periodically for the purpose of defining the activity of each area, the results to date, and any proposed research and development.

The first meeting of the Committee was held at the Atomic Energy Commission offices in Washington. It was decided at this meeting that future on-site meetings would be divided into two technical sessions: one devoted to a discussion of the waste problems of the site and to the local research and development work leading to a solution of those problems; and the other devoted to a seminar on a waste processing method or operation of common interest to representatives of all sites.

This report summarizes the first session of the March 27-28, 1950, meeting of the Committee at Brookhaven National Laboratory.

B. Manovitz
Host Chairman
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The Geology Group started work in March, 1946, on its phase of the general program of environmental studies at Brookhaven National Laboratory. The work is being done by employees of the Water Resources Division, U.S. Geological Survey, who have been assigned to the Laboratory. Most of the scientific direction has come through Survey channels, rather than from the Laboratory, while the Laboratory has handled virtually all of the contract negotiations for well drilling and related major expenditures. The members of the Group, however, have been given the status of Laboratory employees for all such routine, but important, purposes as signing purchase or work orders and use of the Laboratory's many facilities and services.

The purpose and scope of the program have never been set down, and deliberately so, since the whole question of environmental contamination is new, complex, and probably full of unexpected problems. It was early decided, however, at a conference which included virtually all of the interested people, that one need not be concerned, at least until further word, with airborne contamination which might get into surface or ground water, since airborne contamination serious enough to do this would be a more serious problem in its own right. The Geology Group has, therefore, centered its attention on the direct contamination of the surface or ground water.

Since the nuclear reactor and the waste storage connected with the reactor are apparently to be about as nearly foolproof as science and money can make them, one has no idea as to what particular event one should be on guard against; a few, but not all, of the Laboratory people concerned, feel that more important than the danger of a real spill is the possibility of fear, or of legal action, on the part of the public or of public officials in this county. The only safeguard against this is to have always available and always up to date impartial, simple, and clear-cut evidence that the water supply of this area has not been contaminated by the Laboratory. That is the first goal which the Group has set.

What has been done so far is this: A good start has been made on getting together an adequate picture of the local geology insofar as it influences the movement of ground water in this area. Also, it is known where this picture is weak and will have to be strengthened. The major part has been completed, at least as far as expense is concerned, of a network of observation wells to give the major features of the motion of the ground water; and it is known where this information is weak. A good start has also been made toward setting up a net of water sampling points, for both ground water and surface water. Detailed plans were made for completing this work before the reactor starts up and the need for the work arrives. About 20 surface water and ground water samples have been taken and analyzed in detail, both chemically and radiometrically. A good working picture seems to be available of what the water in tuis area was like before the start of operations. It is important, however, to emphasize as strongly as possible the inadequacy of any ground water.
sampling program to insure that contamination is not present and is not getting by the sampling points. The Survey's experience with ground water contamination of other sorts has shown how erratic the spread of the danger may be, even in apparently homogeneous materials, and how it may pass by wells where it should show up, and how it does put in a sudden appearance where it has no business to be at all.

The use of tracers has been initiated to follow in detail the movement, in space and in time, of the ground water in the area. This has served principally to show the serious inadequacy of the older accepted methods, but it has also led to new and possibly improved methods which should give reliable results. It is hoped to make some major contributions in this direction; well-laid plans and high hopes are on hand, but few results, so far.

One major part of the work has not been started at all; that is the study of the adsorption of the reactor waste products by soil materials. The Survey in Washington has hopes and plans for the start of a general study of this broad and complex field. In the meantime, it would be desirable to have some figures, preliminary and empirical perhaps, on the adsorption effects of our own local materials. Such information is needed, and needed now, for a variety of purposes, not the least of which is as a guide in the water sampling program. The rate of travel of any material in solution in the ground water is markedly affected by the degree and type of its adsorption. With tracers, for example, we have already found some traveling more than twice as fast as others through the same material and at the same time because of differences in their adsorption isotherms. Further, it seems not unlikely that some contaminants will put in an appearance gradually at any given water sampling point, while others may arrive dramatically and with virtually the maximum concentration that they are to attain. For these reasons, and others, the services of a physical chemist were sought shortly after the Group was established here, but so far without success. It is intended to keep on trying.

One last question which has been of concern from time to time is what measures might be taken in case there were a serious contamination of the ground water. Three lines of attack would be possible. The first would be to provide new sources of water supply to replace those which had been hurt, or which were in danger. The second would be to inject into the contaminated region an absorbent which would pick up and hold an appreciable portion of the active material. The third, and most desirable, is the apparent possibility of sinking a group of scavenging wells into the contaminated area and pumping out the hot water. This involves, of course, putting that water somewhere else. With this as one of the reasons, two deep wells have been drilled to bedrock in this area. The details are complex, but we feel confident that it will turn out to be entirely practical to pump large volumes of contaminated water into any one of several deep lying formations as a highly desirable alternative to letting it drift along in the surface formations which supply both the laboratory and the neighboring communities. Such heroic measures would only be necessary in case of a serious accident, but plans, and some of the preparations, could well be made beforehand if further study of methods and policy are favorable.

One lack which is felt here, and which the AEC Waste Processing Committee may be able to help us with, is lack of contact with the Survey
groups or other workers engaged in ground water or surface water studies at the other AEC installations. While all send in monthly reports to Washington, there is no contact with one another, except on rare occasions and usually on a purely personal basis. The AEC could, with real profit, make possible an occasional get-together of the various men and groups working in this field.
PROGRESS OF THE METEOROLOGICAL CONTROL GROUP

M.E. Smith
Brookhaven National Laboratory

Relatively complete summaries of the BNL meteorological program have been presented at prior meetings of the AEC Waste Processing Committee and other groups, and in a number of Laboratory publications. These reports have all necessarily dealt with the planning and construction phases of the program, and, although various segments of meteorological data have been presented, little information concerning the dispersion of the oil-fog effluent has been available. Since January, 1949, members of the Meteorology Group have been able to conduct "smoke runs" on a routine basis. In this same period, full meteorological records from both the 420' and 160' towers have been available. It is now possible to present a number of reasonably firm conclusions regarding the atmosphere waste disposal problem.

Before proceeding to this portion of the discussion, a brief review of the smoke run procedure is in order. The oil-fog effluent is produced by an Army M-1 smoke generator, at a rate which can be varied from 20 to 100 gallons per hour. Smoke is ejected from a 355' long x 20" diameter steel pipe mounted within the tower structure. The effluent trail is photographed from selected sites with cameras operating synchronously at a speed ranging between one frame per second and one frame per five seconds. Significant features of the dispersion pattern are observed visually and by theodolite from appropriate vantage points. Concentrations of the effluent which descend to levels near the ground are measured by photometric densitometers mounted in trucks. Up to the present, attempts to measure effluent concentrations aloft from aircraft have been relatively unsuccessful, owing to a number of technical difficulties. A light-weight densitometer has just been completed for use with balloons, so that reliable measurements of concentrations aloft will be available. Data are processed in conjunction with detailed meteorological data obtained during the same period, thus providing a complete record of all aspects of the dispersion conditions.

The record to date includes 35 processed smoke runs; they were conducted in such fashion that the dispersion conditions typical of the most common meteorological regimes would be determined first. As a consequence, the bulk of the information now available generally covers periods in which rather steady lapse, inversion, or neutral (isothermal) temperature structures maintained. A number of runs have been conducted during periods in which a changeover from lapse to inversion, or vice versa, occurred.

During lapse conditions, when the temperature shows a decrease with height of 0.5°C (or more) per 100 meters, large thermal eddies bring relatively high concentrations of effluent to the ground for brief periods of time. The smoke trail also shows wide horizontal fluctuations, so that any single area is affected by the effluent quite intermittently. The region of maximum concentration is generally on the Laboratory site; few measurable concentrations have been found at distances of 3 kilometers or more.
A neutral or isothermal condition is defined as one in which the vertical temperature gradient lies between $+0.5$ and $-0.5^\circ C$ per 100 meters. High frequency mechanical turbulence dominates the air flow during this condition; the larger, relatively long-period thermal eddies are conspicuous by their absence. Under neutral conditions, which generally occur with moderate winds, effluent is brought to the ground more steadily and over a narrower path than under lapse conditions. The area of maximum concentration is usually near or slightly beyond the site boundary; concentrations have been measured up to 6 kilometers from the source.

The inversion case, in which temperature increases with height at a rate exceeding $0.5^\circ C$ per 100 meters, is particularly interesting owing to the unusual turbulence pattern encountered. Vertical fluctuations of the effluent trail are almost completely suppressed. Although short term horizontal fluctuations are also of small magnitude, significant lateral eddies appear over the period of an hour or more. Thus, the effluent trail assumes the appearance of a narrow meandering ribbon, visible for great distances. No low level concentrations have been measurable under inversion conditions, even at distances of 30 kilometers from the source. Visual observations have indicated that the base of the trail has reached a level no lower than 300-350'. Owing to the geography of the area, it has not yet been possible to follow an inversion trail to greater distances; but it is certainly reasonable to believe that no significant ground level concentrations occur while the inversion persists.

The period of change-over from inversion to lapse, though transitory, is worthy of consideration. When the lapse condition builds up to the level of the inversion trail aloft, appreciable amounts of effluent are brought quickly to the lower levels. The development of normal day-time turbulence dissipates such concentrations within a short period of time.

In the reverse situation, when a lapse condition changes to an inversion, it has been found that the effluent remains entirely aloft after the inversion is fully developed; the residual low level concentrations, usually typical of the neutral case, gradually dissipate.

With reference to the operating levels agreed upon for the Brookhaven area, final conclusions concerning radiation dosage resulting from the reactor operation now await the operation of the reactor itself. Theoretical calculations of the ground level dosage under various conditions have been made, which show the following with reference to operating conditions: Under lapse conditions mean hourly concentrations in the area of maximum will probably exceed the off-site operating level consistently, although the area of maximum will generally be on-site. Under neutral conditions, mean hourly concentrations in the area of maximum should generally equal or exceed the established operating level. During the brief fumigation period following the change-over from inversion to lapse, instantaneous, low level concentrations are expected to reach values in excess of the operating level by at least a factor of ten.

Casual appraisal of these estimates might lead to the belief that it would be necessary to restrict the operation of the reactor a considerable portion of the time. However, a number of additional factors must be assessed. The variability of wind direction and speed, the normal diurnal fluctuation of
temperature structure, and the relatively short half-life of $\text{A}^{41}$ conspire to make it unlikely that a given area will receive maximum low level effluent concentrations for any extended length of time, or on repeated occasions during the 7-day period on which the operating level is calculated. Thus, the early estimates of restriction of reactor operation 5% of the time are still considered reasonable. These periods of restriction are expected to occur when a stagnant synoptic weather situation causes the detailed pattern of dispersion to be persistent or repetitive over several days.

From the point of view of forecasting dispersion conditions, considerable progress has also been made. It has been found that certain synoptic weather factors, for the most part routinely observed and forecast, strongly influence the micrometeorological developments. The most important of these are cloud cover, gradient wind, air-ground temperature relationship, and the state of the ground. Experimental forecasting carried on over the past year has resulted in a preliminary determination of these factors, which are summarized as a guide in the routine forecasting of local wind, temperature, and turbulence.

Templates depicting the ground level patterns of radiation dosage have been prepared for a wide variety of weather conditions. These are currently utilized in a daily practice evaluation and forecast of reactor operating conditions.
AIRBORNE ACTIVITY

The BNL policy is to design facilities so that the disturbance to the off-site radiation background shall not average more than 1/3 of the accepted whole body exposure limit of 300 mr/wk. The function of the meteorology program in predicting dilutions for the reactor cooling air has been described in this report by M. S. Smith. The system of area monitoring stations, which will be described here, has been set up to measure the actual disturbance of the natural background at 10 locations located strategically within 10 miles of the site, as well as at 6 locations within the boundaries of the site. The radiation measurements made at these stations, the data obtained from the smoke runs of the meteorologists, such theoretical material as is available in regard to the dispersion of stack gases, and the gross and micro-meteorological data collected by the Meteorology Group combine to give a very excellent picture of radiation conditions in the area and make it possible to meet the severe requirements of the above-mentioned policy. The area monitoring data and known characteristics of the reactor effluent can be used to correct or extend present knowledge of diffusion theory, as well as to check the conclusions made from the smoke runs. The meteorological analysis can be used for interpolation between the discreet locations of the area monitoring stations. A close liaison is maintained between the Meteorology Group and the Health Physics Division. Further elaboration of operating procedures is to be worked out when reactor operations have commenced.

The area monitoring station equipment includes a variety of G-M counters, an ionization chamber in combination with a vibrating-reed electrometer and chart recorder, a battery-operated ratemeter to provide data during power failures, and a continuously-operated dust collector and counter. All of these operate continuously. The indicators are located on a central display board, which is photographed automatically once every 6 minutes. Data are analyzed periodically. Immediate records of radiation conditions are available from the chart recorders of the ionization chamber and the battery-operated ratemeter. The dust monitor consists of a strip of filter paper drawn, at a constant rate, across a suction head, and then past both alpha and beta-gamma counters. The equipment for the area monitoring stations was developed by the Electronics Division; it is operated by the Health Physics Division.

The naturally occurring background radiation in this area amounts to about 0.01 mr/hr. With a standard Eck and Krabs G-M tube about 6" long x 3/4" diameter, the counting rate is of the order of 30 c/min. Quite a considerable variation in background as the result of rainfall is noted, with peaks as much as twice the normal counting rate. The great bulk of the activity collected by the dust monitor consists of active deposits of radon and thoron. Peaks in this activity of 3 or 4 times the normal amount are noted as a result of temperature inversions. During such periods, the lower layers of air are stable, and vertical mixing does not occur. Accordingly,
the activity released from the soil is not dispersed in the normal fashion. Curves for these effects are given in "Brookhaven National Laboratory Progress Report," July 1 - December 31, 1949 (BNL 39 (AS-3)).

Liquid Waste

Brookhaven National Laboratory has its own sewage system and sewage processing plant, the effluent from which is discharged into a small stream which is part of the headwaters of the Peconic River. The policy in regard to the release of radioactivity from this system is a stringent one dictated by the lack of massive dilution and the details of ecology of this region. This policy, recently approved by the Atomic Energy Commission, calls for release of effluent whose activity, averaged over a 3-month period, shall not exceed $3 \times 10^{-3} \mu\text{c/cc}$. Peak values of 10 times this concentration will be permissible; the total amount is limited to 1.5 c/yr, corresponding to a flow rate of 360,000 gal/day at the maximum allowable average concentration. This concentration control will be applied at the point where the Imhoff waste processing tank discharges to the filter beds. Since decontamination is to be expected in passing through the filter beds, the activity released to the stream is expected to be even lower than indicated by the agreed policy. Some interchange with the ground water is to be expected in the filter beds and along the stream, but this is of secondary importance at these levels because of the adsorptive characteristics of the soil and a number of other factors.

In order to achieve such a policy in a laboratory as extensive as BNL, it is obviously necessary to control the discharge of activity to the waste system at the source, and particularly at locations where large amounts of activity are being manipulated. The daily quota for the Laboratory amounts to 3.4 mc. The severity of the problem is brought out if half of this amount is allocated to the nuclear reactor hot laboratory complex and the other half is divided among various other facilities, such as the cyclotron, chemistry laboratory, Chemistry Department semi-hot laboratory, Biology Department dilution wing, decontamination laundry, Medical Department therapy wing, waste concentration laboratory, metallurgy laboratories, hot shop, and numerous other chemistry, biology, and medical laboratories which use appreciable quantities of radioactive isotopes. With such an allocation, one finds that each laboratory wing or small group of laboratories can release no more than 50-100 µc/day.

The reactor complex has been set up as a self-contained system, with full control of its liquid waste; problems entailed therein are discussed in this report by F.L. Horn. Only diluted and monitored wastes will be discharged to the laboratory waste system. Hold-up tanks and proportional sampling devices are being provided for other critical locations, such as the Chemistry Department semi-hot laboratory, cyclotron chemistry laboratory, and Biology Department dilution wing. The intent here is that the held-up waste will normally be of low enough activity that it may be released after an assay has been made. If, because of accident or poor technique, this is not the case, it will be transported by means of tank truck to the tank farm associated with the nuclear reactor. For some other locations which are less likely to cause difficulty, proportional samplers without hold-up tanks will be provided. This will allow detection of foci of poor technique which, if not detected,
might result in activity levels at the sewage plant which are too high. For other areas where activity levels are very low, no control will be exercised. In all laboratories, containers will be provided for collection of liquids containing appreciable amounts of activity. These will be transported periodically to the tank farm.

Extensive monitoring facilities are being provided at the sewage processing plant. These will consist of four installations:

1. A flow meter and proportional sampler at the exit of the Imhoff tank.

2. A similar installation at the chlorination plant, which is located between the filter beds and the point of discharge to the Peconic River.

3. A flow monitoring station upstream from the point of discharge.

4. A flow monitoring station downstream from the point of discharge at the point where the Peconic River crosses the eastern boundary of the site.

These installations are partially completed; they will be in operation in a few months. The data obtained, in addition to making it possible to know whether or not the agreed policy is being met, will yield other interesting information, such as the loss of activity in the filter beds, the dilution by the flow of the stream, and the amount of activity leaving the boundary of the site in the water of the stream.

Some miscellaneous aspects of area monitoring are analysis of vegetation, soil, and well water. Particular attention will be paid to analysis of water and biological forms downstream along the Peconic River. These phases of the program will be elaborated as the quantities of activity released become appreciable.
The radioactive waste system of the BNL reactor complex is nearly completed. It is designed to handle liquid waste from the reactor, reactor laboratories, and hot laboratory. Radioactive waste from some other groups in the laboratory will also be stored with reactor waste, but will be handled in hot pots.

Types of Waste

The waste has been divided into five types, each of which is handled by a separate system. The designations, definitions, and expected yearly volumes of these wastes are:

A. Accountable metal waste containing uranium and fissionable material. 7,300 gal/yr.
B. High activity chemical waste containing no accountable material. 32,000 gal/yr.
C. Contaminated solvent waste which cannot go through waste system because of volatile solvents or HCl content. 3,900 gal/yr.
D. Low activity chemical waste which does not require shielding. 300,000 gal/yr.
E. Normally uncontaminated waste which is monitored for activity before disposal. 4,000,000 gal/yr.

The volumes given above are rough estimates used in designing the system, and are probably high. The volumes are also for full operation, which will not be attained for 6 to 10 months.

Collection

All waste from drains at reactor faces, floor drains, and sinks will run by gravity to the "F" waste tanks in the hot laboratory. This waste should normally be uncontaminated. After it is monitored, it will be released to sewerage. The east wing reactor laboratories have gravity "F" drains to the hot laboratory. The west wing reactor laboratories have "F" drains to a 2000-gal hold-up tank, from which the waste is pumped periodically to the 8000-gal hot laboratory tanks for monitoring. Should any of the "F" waste be contami-
nated, that 8000-gal tankful will be pumped to the "D" storage tanks.

"D" waste in each of the reactor laboratories will be collected in 5-gal containers. The containers will be trucked to the hot laboratory and emptied into the "D" system.

Another source of waste from the reactor is the canal. The canal will be emptied when a ruptured slug is removed from the reactor. The 80,000 gal of canal water will then be run to a 100,000-gal tank reserved for this purpose.

The hot laboratory cold area will also collect its "D" waste in 5-gal containers to be emptied in the waste treatment area. All "F" waste is conducted by piping to the "F" waste monitoring tanks in the waste treatment area.

The hot area of the hot laboratory is the only area which is completely connected to the waste treatment area by piping. The pipes are all welded stainless steel. The piping is laid in concrete trenches which are painted with Amercoat. If there are any leaks in the piping, the liquid will drain to the waste treatment area. The trenches have 1' thick concrete covers for shielding which fit flush with the floor.

Waste will be collected from three main areas in the hot area: the dissolver cells, the semi-works area, and the semi-hot cells. The trenches from these three areas have a junction point at the neutralization cell in the waste treatment area. The pipes for "A," "B," and spare waste lead into the neutralization cell in the waste treatment area. "D" waste lines go into off-gas separation tanks in the neutralization cell. The gas will go to the scrubber cell, and the "D" waste to the "D" cell. "F" waste will continue through the neutralization cell to a drum trap which catches large articles in the waste. From there, the "F" waste will run into a diversion tank with a baffle. The baffle causes the waste to flow past a G-M tube which is connected to a counter. Should the count on "F" waste go above a predetermined level, a magnetic valve at the bottom of the tank will be actuated, dumping 60 gal to the "D" system. This process continues until the "F" line is flushed out.

All the tanks in the neutralization cell are vented into the scrubber off-gas system, causing a flow of air down all waste lines. The semi-works area has ventilation ducting that passes through the scrubbers. The off-gas from tanks and semi-works is known as "acid off-gas." It contains acid and iodine fumes. The caustic scrubbers will neutralize the acid and scrub iodine from the gas. The gas will then pass through a roughing filter and a Chemical Warfare Service filter. A 14" stainless steel duct will carry the gas to join the off-gas duct from the decontamination area, then to the blower in the hot laboratory fan house, which will send it up the stack.

Room and hood air ventilation for the hot area and waste treatment area is termed "nonacid off-gas." The air will go through ducts to CWS filter units and blowers. A 42" duct will carry the air to the hot laboratory fan house blower, which will send it up the stack.
Treatment

"A" waste is accountable. It will be sampled carefully and neutralized. The volume that is jetted to storage will be recorded. Neutralization will be accomplished by jetting waste to the neutralizer tank, where caustic or carbonate will be run in. The solution will be agitated, cooled, and sampled. Waste will then be jetted to the tank farm for storage. "B" waste will be treated similarly. These operations will all be controlled remotely at a panel board which is separated from the cell by a 4' thick concrete floor.

"D" waste will be sampled and the proper amount of neutralization solution added directly to the "D" tank. "D" waste will then be sampled and pumped to storage at the tank farm.

As mentioned previously, "F" waste will be monitored daily and pumped to the sewer.

Storage

The tank farm is located 1/8 mile north of the waste treatment area. "A," "B," and "D" liquid wastes are stored there. The tank farm is joined to the waste treatment area by means of pipes in a covered concrete trench. "D" waste is stored in two 100,000-gal tanks above ground. A third 100,000-gal tank is reserved for contaminated canal water.

"A" and "B" waste is stored in six 3000-gal stainless steel tanks below ground. Each tank is enclosed in a concrete cubicle. The cubicles have sumps which will be monitored to determine whether the tanks are leaking.

"C" waste which is collected in pots will be stored in the pots awaiting further developments.

In general, the waste treatment area is constructed to handle large volumes of waste from the hot laboratory and reactor. Hot wastes are controlled remotely by the use of steam jets. Collection, classification, neutralization, and storage of radioactive solutions are accomplished. Accountability of uranium and fission product solution is maintained. Sufficient storage capacity of "D" waste is provided for 1 to 2 years, after which concentration of the waste is expected. "A" and "B" waste storage capacities are sufficient for 2 to 3 years.
Figure 1. Nonacid off-gas system.

Figure 2. Acid off-gas system.
Figure 3. "D" and "F" waste systems.

Figure 4. "A" waste system.
Figure 5. "B" and "spare" waste systems.

Figure 6. Blowers and duct connections for filter units used in semi-works ventilation.
Figure 7. Process and instrument piping to tanks in neutralization cell.

Figure 8. Two "F" cell tanks at upper level.
Figure 9. Four caustic scrubbers used in acid off-gas system.

Figure 10. Diversion tank connected between "F" and "D" systems.

Figure 11. Control and instrument panel for neutralization cell.
Figure 12. Tank in "D" cell, from upper level.

Figure 13. Waste lines in bottom of hot cell--"A," "B," "D," "spare," and "F."
PERMANENT DISPOSAL OF RADIOACTIVE WASTES

W. S. Ginell and L. P. Hatch
Brookhaven National Laboratory

An investigation is currently under way at BNL to develop a procedure for the permanent disposal of long-lived radioactive wastes, particularly those which occur as a result of the fission process. In this work, the basic premise is to achieve assimilation of the wastes by highly stable natural materials in the earth rather than resort to such means as the use of storage containers, contamination of the ocean, etc.

It is known that minerals of the montmorillonite group have pronounced cation exchange properties. Furthermore, in the temperate regions of the earth's surface, they are very nearly the end product of the weathering of volcanic ashes, and so they can be considered highly stable natural materials. These factors, along with the known imperviousness of clay beds to the passage of water, suggest the use of clays as disposal media. The disposal process would involve replacement of inactive exchangeable cations in the clay by radioactive cations. The active clay, after suitable processing, could then be disposed of by burial in some selected area, possibly in a deep stratum of clay itself.

Since ion exchange reactions, in general, follow the law of mass action, one would expect a reversal of the exchange reaction in the presence of an excess of other cations. Presumably, there would be some very small movement of liquids containing dissolved solids through the natural clay beds, so that the reverse reaction could occur to some degree. Hence, it would be necessary to treat the radioactive clay in such a manner that the rate of the reverse reaction would be negligibly small. In other words, the radioactive cations would have to be fixed in the clay. The immediate problem, therefore, resolves itself into two major tasks, namely:

1. Determination of the cation exchange capacities of certain clays as functions of the concentration and nature of the cation, temperature of the reactants, particle size of the clays, etc.

2. Development of a process for fixing the radioactive cations in the clay.

Three clays are currently under investigation at BNL: a low volatile matter fuller's earth (Attapulgus Clay Co.), and two types of pure montmorillonite (Filtrol Corp.). Preliminary experiments are being conducted using radioactive strontium 89 (half-life: 28 days) obtained from Oak Ridge in the form of $\text{Sr(NO}_3\text{)}_2$. The material is purified by precipitation of $\text{SrCO}_3$ with $(\text{NH}_4)_2\text{CO}_3$, followed by filtration, washing, and dissolution of the solid in an excess of HCl. The resulting solution is evaporated down to half its volume, to expel CO$_2$, and then neutralized to pH 6.3 with an inactive solution of $\text{Sr(OH)}_2$, using a Beckman pH meter. The resulting stock solution is analyzed for Sr by a gravimetric procedure involving precipitation of $\text{SrSO}_4$ from 50% alcohol. The specific activity of the solution is determined by diluting an aliquot of the stock solution to a convenient volume and counting.
the resulting solution in an annular jacketed thin-wall glass G-M tube.

In the determination of exchange capacities, weighed clay samples, pretreated by drying at 110°C, are transferred to 100-ml glass stoppered flasks. To these are added known volumes of the strontium chloride solution. The flasks are placed on a wrist-action mechanical shaker and agitated. Aliquots are withdrawn at intervals for counting. Knowing the specific activities of the original and final solutions, it is possible to calculate the capacity of the clay in milliequivalents of strontium per gram of clay (me/gm). After equilibrium has been established, the clay is centrifuged, washed, and dried at 110°C. A weighed sample of the strontium-saturated material thus obtained is digested with concentrated HNO₃ to decompose the clay. The resulting solution is then counted. This serves as a check on the previously determined exchange capacity. The figures in Table 1 give the values of the exchange capacities of the clays under investigation in terms of milliequivalents of strontium per gram of oven-dried clay.

<table>
<thead>
<tr>
<th>Table 1</th>
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<tr>
<td>Exchange Capacities</td>
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<td>Clay</td>
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<tr>
<td>Attapulgus Fullers Earth</td>
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<tr>
<td>Filtrol Montmorillonite R-2532</td>
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<td>Filtrol Montmorillonite R-2472</td>
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1. 10 gm clay + 225 ml of 0.05M SrCl₂.
2. HNO₃ extraction of saturated clay from 1.
3. 0.5M BaCl₂ extraction of saturated clay from 1.

The saturated clays are then subjected to a series of heat treatments designed to determine the amount of activity which can be fixed in the clay. Weighed amounts are placed in Vycor dishes and heated in a muffle furnace for periods of the order of 24 hours. Thus far, temperatures in the range 300-900°C have been tried. After removal from the oven and cooling, the clays are transferred to 100-ml flasks, treated with a large excess of BaCl₂ solution, and shaken for long periods. Aliquots of the supernatant solution are withdrawn at intervals for counting. Tests have shown that barium replacement is 100% complete in the case of unheated saturated clays; therefore, this technique serves as a measure of the degree of fixation achieved by heating. Figures 14, 15, and 16 show the observed fixation as a function of temperature.

The montmorillonite minerals are known to have a crystal lattice whose c dimension is a function of the water content of the mineral. Expansion and contraction of the lattice layers is reversible if the clay is maintained at low temperatures. However, it was found by other workers that heating the clay to 400-500°C causes a loss of swelling capacity when the clay is placed in water. Since the exchangeable cations are located between the lattice layers for the most part, the loss of swelling ability should
also be associated with a lowering of the exchange capacity. This effect can be seen readily in Figure 14 and, to a lesser degree, in Figure 15; a marked decrease in exchange capacity (or increase in percent fixed) is observed between 400 and 500°C. Above this temperature range, the structure of the clay is probably altered by the loss of the elements of water from within the lattice layers. At temperatures around 800°C, the exchangeable cations appear to be fixed to a very great extent. In addition to the extralattice exchangeable cations, some exchange presumably occurs with cations formally within the lattice but which had been exposed as a result of broken crystals. Such cations could also be replaced. If, however, the clay were heated to a temperature at which sintering occurs, the number of such exposed cations would be greatly reduced. It seems probable that this could be one explanation for the relatively slow decrease in exchange capacity in the temperature range 500-900°C. In the case of montmorillonite R-2532 which was heated to 875°C, sintering was actually observed.

Future experiments will be conducted along these lines, using mixed fission products and possibly other clays. We hope to be able to study, by means of X-ray diffraction techniques, the actual changes in the clay structure brought about by heating.