ENGINEERING RESEARCH INSTITUTE
UNIVERSITY OF MICHIGAN
ANN ARBOR

PROGRESS REPORT NO. 1

INDEPENDENT ENGINEERING SURVEYS AND EVALUATIONS ON THE TREATMENT AND HANDLING OF FISSION PRODUCTS FROM RADIOACTIVE WASTE SOLUTIONS OF CHEMICAL PROCESSING PLANTS

J. G. Lewis
M. E. Weech
H. A. Ohlgren
R. R. White

Project 2378

THE DOW CHEMICAL COMPANY
MIDLAND, MICHIGAN

July 15, 1955

DISTRIBUTION LIST

	Copy Number
A. W. Hanson	1 through 4
R. G. Folsom	5 and 6
R. R. White	7
H. A. Ohlgren	8 through 15

ENGINEERING RESEARCH INSTITUTE
UNIVERSITY OF MICHIGAN

July 15, 1955

Mr. A. W. Hanson, Director
Nuclear Research Department
The Dow Chemical Company
Midland, Michigan

Dear Mr. Hanson:

Accompanying this letter are some of the thoughts and objectives which we have in mind for the study which we are conducting for you of the packaging of fission products under your Special Authorization S-831.

We have prepared this description of ground rules under which we propose to operate because it appears to us to be important to have a clear understanding of these basic ground rules if an economic analysis of the cost of packaging fission products is to have meaning. We have proceeded on the assumption that you would wish to have a fission product packaging plant located adjacent to some existing or future facility for the chemical processing of irradiated nuclear fuels.

Most existing fuel processing plants are located in quite remote areas, and it has therefore seemed to us that a fission product packaging plant would have to provide for itself or rely upon the existing fuels processing plant for many of the facilities which do not ordinarily require separate consideration when an additional plant is installed in an existing processing complex. We are taking the point of view in these proposed ground rules that you will wish us to rely upon the existing general facilities of the fuel processing plant where possible. We are presently planning, as noted in the accompanying description of ground rules, to omit from the capital requirements the provision of such items as purchase or lease of land, provision of access roads or railroads, steam and electrical generating facilities, a separate well and pumping station, a standpipe for fire protection, heavy maintenance shops, sewage disposal plant, garages, and a cafeteria or other central food supply.

It is possible that additional considerations, such as transportation service for employees to and from the nearest population center, will have to be considered. We are assuming that existing transportation facilities can be employed.

A rather detailed listing of items of cost to be considered is listed in III. PHILOSOPHY OF ENGINEERING DESIGN. This section is intended to define the limits of cost items which will be considered in our estimates, and is not meant to imply that all costs will be estimated in detail.

Additional technical questions arise in the matter of the objectives of the fission product packaging plant. We are planning to proceed with the examination of various processes in accordance with the following priority of objectives: (1) To package fission products, (2) To reduce waste volume, (3) To remove long-lived nuclides sufficiently to permit discarding of remaining fission products.

On certain questions reflecting to a greater degree your specific policies, we will forward for your review further details as these become available.

We are proceeding with conceptual designs of alternative processing methods and would appreciate your comments on these suggested ground rules for economic analysis. If you find any suggested procedures with which you do not agree, we would appreciate meeting with you at an early date in order to clarify our understanding of your wishes.

Very truly yours,

J. G. Lewis

J. G. Lewis
Project Supervisor

jb
Enc.
2378

PREFACE

This volume is a report on the progress achieved to date in studies of the treatment and handling of fission products from radioactive waste solutions of chemical plants processing nuclear fuels.

The staff, faculty, and students of the University have contributed information and efforts to the preparation of this report. The authors are indebted to Mr. D. H. Stewart for supplying information and suggestions used in the preparation of this report.

Many of the techniques described here for the separation and treatment of the fission products were originated and developed in Oak Ridge National Laboratory, Argonne National Laboratory, The National Reactor Testing Station, the Hanford Works, and Brookhaven National Laboratory.

TABLE OF CONTENTS

I. ABSTRACT

This is a progress report on the results of preliminary examination of various possible methods of packaging fission products and an outline of proposed objectives and procedures for the completion of technical and economic feasibility studies for the packaging of fission products.

The objectives of the work described may be summarized under three categories as follows:

A. To produce a package of a desired fission product suitable for use in irradiation procedures.

B. To reduce the storage cost of the fission products remaining after the desired materials have been extracted.

C. To remove long-lived fission products from aqueous wastes to a degree which would permit accelerated disposal of the remaining wastes to the environment.

The studies upon which this progress report is based have consisted of examination of the following alternative or supplementary methods of preparing fission products in the form of packages of solid salts.

A. Co-precipitation for selective removal of cesium and probably other alkali metal ions.

B. Ion exchange for the removal of cesium and probably other alkali metal ions.

C. Co-crystallization of cesium and possibly other alkali metal ions.

D. Complexing and solvent extraction for the selective removal of alkali and alkaline-earth ions.

E. Fluidized bed evaporation of aqueous and acid solvents from solutions of separate or gross fission products.

F. The leaching of soluble oxides, such as cesium oxide, from a dried mass of oxides of the fission products resulting from the evaporation of a solution of gross fission products.

Paralleling examination of the work of others, some experimentation is being conducted on the solvent extraction method of separating soluble fission products. A chart is included in this report illustrating preliminary distribution coefficient data for the extraction of barium as the nitrate, and of barium, strontium, and cesium as the nitrates

from aqueous solution. Capital and operating costs of the required facilities will be estimated and unitized costs reported for the resulting fission products. A description of the operation of the several processes to be considered will be provided.

The philosophy and ground rules of engineering design have been examined for plant-scale methods of conducting the above operations. It is proposed to locate a fission product processing plant adjacent to an existing fuel processing plant, probably at Hanford, Washington, or Arco, Idaho. The remoteness of such locations necessitates consideration of supplying general services to such a plant. In general, it is proposed to rely upon the existing fuels processing plant for the capital facilities required for general services and to exclude such capital requirements from any estimates prepared under this program. Operating cost estimates will include estimated allowances for those services required to operate the plant, such as steam, power, cooling water, drinking water, fire protection water, and sanitary sewage disposal. Comparisons will be drawn for each alternative processing method or methods for the Arco and for the Hanford locations.

It will be assumed that the supply of gross fission products to the processing plant will cost the packaging processor nothing, except for transportation and handling costs. If methods are developed of storage of greatly reduced cost compared with the storage of aqueous solutions, some credit might be taken for such savings. This might be offset by possible charges for the use of the fission products. No estimates of such charges and credits will be made.

An alternative possibility, not to be studied in this scope of work, is that of a private industry building a fuel processing plant integral with fission product packaging facilities.

Some preliminary discussions will be presented of factors likely to affect the sales prices of packaged fission products.

II. INTRODUCTION

A. General

The objectives of this program are to prepare technical and economic evaluations of several methods for the preparation of sources of radiation consisting of packaged fission products. The fission products of longest half-life consistent with moderately large specific activities are cesium^{137} and strontium^{90}. Accordingly, studies are to be conducted for the separation of cesium and strontium from fuel processing wastes in relatively pure form, and also for the preparation of gross fission products packaged in a dry state. Consideration will be given to the reduction of storage costs of undesired fission products by means of reduction of the volume of these wastes, and their storage in the dry state. Attention is also to be devoted to the problem of removing cesium and strontium from active wastes to a sufficient degree to permit remaining wastes to decay to a level safe for discharge to the environment in a much shorter period of time than would be required with the long-lived wastes present. The possibilities will be reviewed of recovery for re-use of chemicals, such as nitric acid, likely to be liberated in processing operations.

It is planned to consider the treatment of the wastes from a specific fuels processing plant. It is thought that the separation of fission products from Purex waste might offer fewer problems than the processing of wastes from sources likely to be complicated by aluminum chemistry. Variables affecting the optimum source of fission products are presently thought to be the following:

1. Concentration of the gross fission products in the waste solutions.

2. Percentage abundance of cesium and strontium in the gross fission products as affected by the length of reactor operation and cooling period.

3. The total quantities of fission products available.

The relative merits of the alternative packaging processes will be considered for alternative sources of wastes offering significant differences in the variables just mentioned. Some thought will be given to the packaging of fission products from pyrometallurgical processing of fuels. However, it is believed that the aqueous process discussed here will probably serve for the packaging of such wastes until methods could be devised to take advantage of the properties of the availability of the fission products in slag form.

B. Methods of Fission Product Packaging

Methods of fission product packaging to be studied are the following:

1. The use of a precipitation to remove cesium from a solution with a subsequent separation and packaging of cesium compounds in dry form.

2. The use of an ion exchange process for the selective removal of cesium from solutions resulting from fuel processing.

3. The addition of certain materials to fuel processing solutions which will permit the cesium to be crystallized from the solution as a complex salt.

4. The use of complexing and solvent extraction methods for the isolation of relatively pure fractions of some of the various fission products, such as compounds of the separated alkali and alkaline-earth elements.

5. The methods of separation of fission products described above have as their chief objective that of the removal of relatively pure fractions of given chemical elements whose radioactive emissions form desirable sources of radiation. In addition to this approach, it is planned to study the conversion of an aqueous waste stream from fuel processing into the form of a bed of solids by removing all volatile substances by the means of a heating process. The resultant product would consist of a dried mixture of compounds of the gross fission products and structural materials contained in the fuel processing waste stream. Such an operation might require the use of a fluidized bed pyrolyzer or a molten salt evaporator.

6. The removal of soluble oxides by leaching the bed of fission product oxides resulting from an evaporation step such as described in 5., above.

C. Work to be Done on this Project

1. Technical and Economical Feasibility Studies

The technical aspects of each of the methods of packaging fission products, mentioned above, will be studied and evaluated. Estimates will be derived for the capital and operating costs to be anticipated for the packaging of fission products by each of the above methods. The estimates of cost to be derived will be based upon certain assumptions detailed further in Section III, PHILOSOPHY OF ENGINEERING DESIGN.

2. Experimental Work

Experimental work will be conducted upon a process being developed at the University of Michigan involving the complexing and the solvent extraction of certain selected fission products. Experimentation will be conducted with the objective of obtaining the conditions of pH, concentration, and temperature in order to attain a desired distribution coefficient of the several fission products which it is desired to extract from the aqueous solution of fuel processing waste. This work will parallel technical and economic evaluations upon this process.

Some preliminary results obtained appear in Figure 1 for the extraction of barium from aqueous solution by means of a complexing agent. Aqueous solutions of cold barium were made up with Ba^{140} added as a tracer. A solution of thenoyl trifluoroacetone (TTA) in methyl isobutyl ketone was placed in contact with the aqueous solution and agitated. The pH of the aqueous solution was continually observed and sodium hydroxide added by increments to the agitated mixture of aqueous and organic phases. At pre-selected values of pH, samples of 100 microliters were taken from each phase and the Ba^{140} counted in each sample. The ratio of concentrations of Ba^{140} in the two phases was taken as the distribution coefficient.

It can be seen from Figure 1 that the distribution ratio of barium in the organic phase to barium in the aqueous phase increases with increasing pH for given initial concentrations of barium and of TTA. From Figure 2, it can be seen that the distribution ratios for strontium and cesium vary with pH in a manner similar to that shown for barium, but reach maximum values at different values of pH than does the distribution coefficient for barium. The existence of a different distribution coefficient for each ion at a given value of pH would provide a basis for the separation of different ions in solution by means of a solvent extraction process.

Experimental work is now continuing on the determination of the distribution coefficients of strontium, barium, cesium, and rubidium in the presence of ions likely to interfere with extraction of desired elements from a solution of fission products. The extraction of very dilute solutions of the ions of interest will also be investigated.

Methods of correlating the data of Figures 1 and 2, as well as of other data to be obtained are being studied. Consideration will be given to the effects of competitive complexing reactions and to relative solubilities and dissociation reactions of the reacting substances.

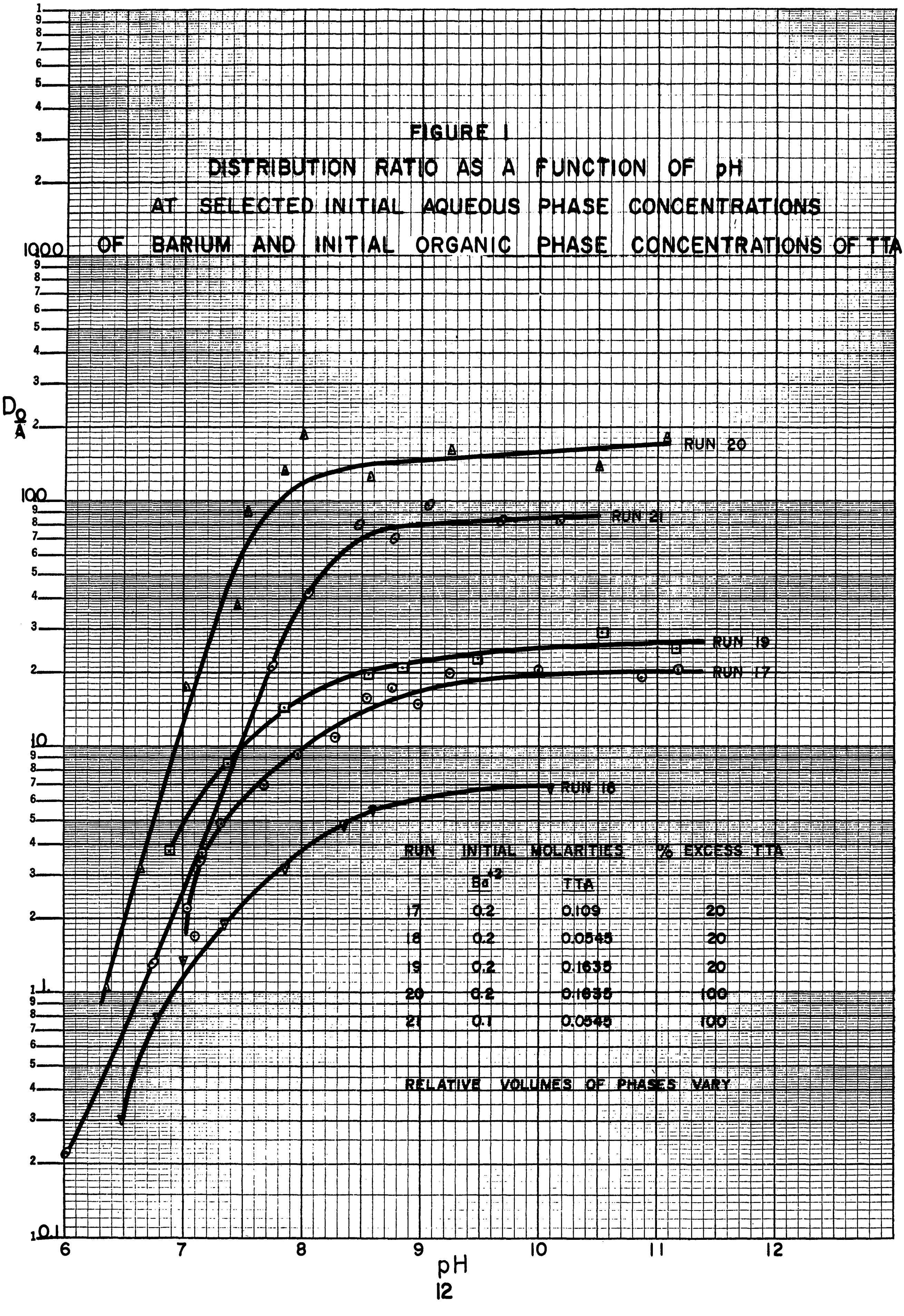

FIGURE 1
DISTRIBUTION RATIO AS A FUNCTION OF pH AT SELECTED INITIAL AQUEOUS PHASE CONCENTRATIONS OF BARIUM AND INITIAL ORGANIC PHASE CONCENTRATIONS OF TTA

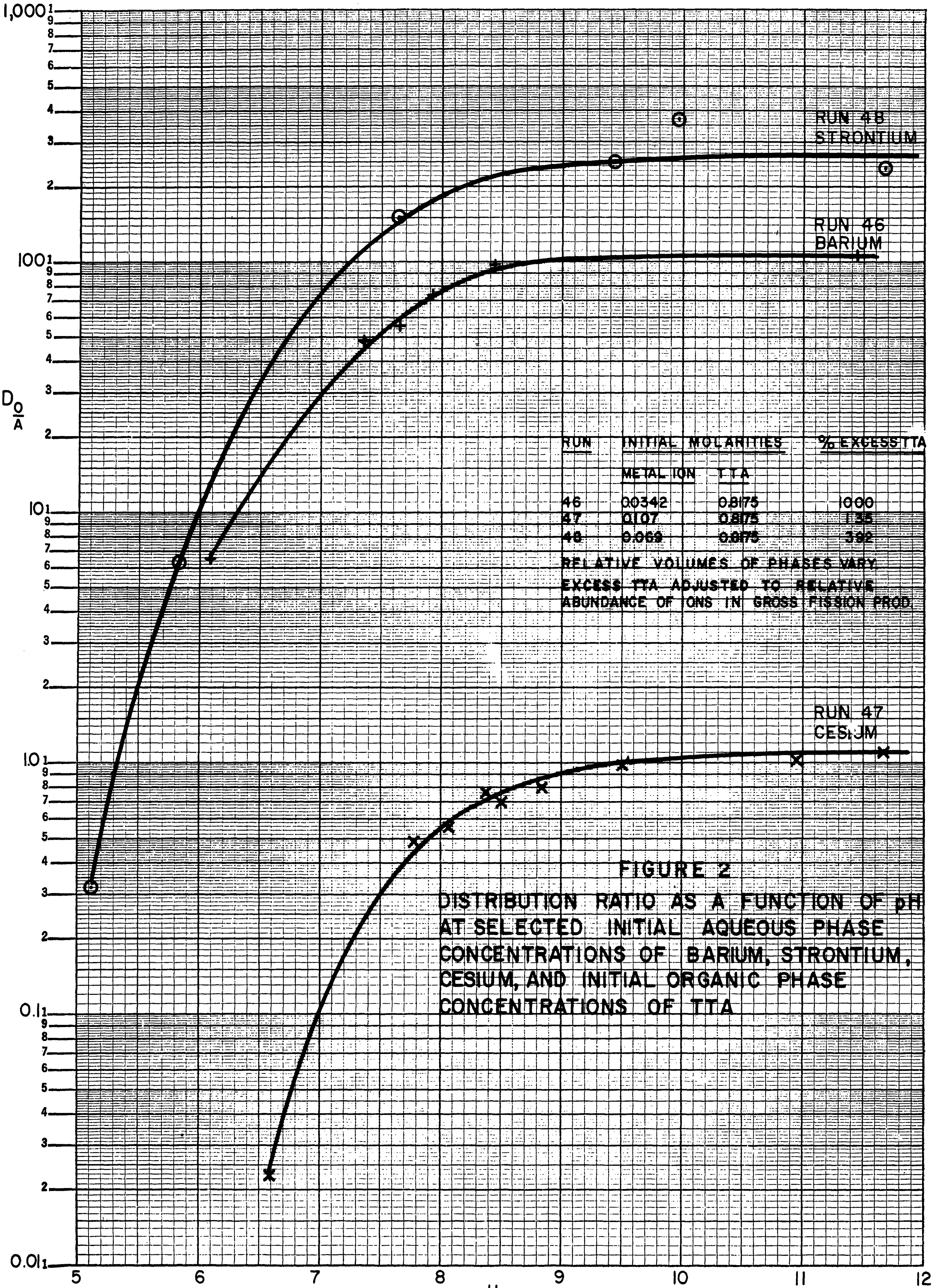

FIGURE 2
DISTRIBUTION RATIO AS A FUNCTION OF pH AT SELECTED INITIAL AQUEOUS PHASE CONCENTRATIONS OF BARIUM, STRONTIUM, CESIUM, AND INITIAL ORGANIC PHASE CONCENTRATIONS OF TTA

III. PHILOSOPHY OF ENGINEERING DESIGN

The philosophy of engineering design adopted for a given plant to package fission products will exert a controlling effect upon the capital and operating requirements of the operation. The elements of a design philosophy which seem most important are:

A. The location of the processing plant, both with respect to the physical requirements of maintaining a remote versus a conveniently located plant and with respect to the nature of the wastes to be processed.

B. The provision for capital facilities to furnish general services.

C. The objectives of the processing operation. That is, whether the plant is to produce packaged fission products, or whether the plant is to produce packaged fission products and an effluent waste stream substantially free of all long-lived fission products.

We are suggesting the following set of understandings for the resolution of the above questions:

A. A chemical separations plant will be constructed which will be located sufficiently close to a nuclear fuel processing plant that waste fuel processing solutions may be transferred to and from the fission product packaging plant by means of pipelines.

B. The fission product packaging plant will depend upon the adjacent fuel processing plant for many general services, and the following assumptions will be made concerning the capital and operating requirements of such a plant. An alternative possibility is the erection of a new fuels processing plant with the fission product plant operating integrally with the fuels plant.

   The items of cost given below are for the purpose of defining the limits of costs to be considered.

   1. A tract of land of adequate size for the operation of this plant may be purchased or leased in the desired site adjacent to the fuel processing plant. No land or site-clearance costs will be considered in the capital cost.

   2. No roads or railroads will be provided beyond the plant limits. Roads and railroads will be provided within the plant limits.

   3. Utilities - Electrical power at not less than 2300 volts will be available at the plant limits. Steam will be available from the

fuels processing plant. Water may be obtained from the fuels processing plant. Drinking water will be available from the fuels processing plant. Adequate water for fire protection will be available from the fuels processing plant. Compressed air for instruments in general plant use will be provided by a compressor station. Fuel will consist of bottled gas for laboratory use.

4. Disposal of contaminated organic wastes may be achieved by storage. Disposal of decontaminated waste solutions may be achieved by dumping to the surrounding earth. Sanitary waste disposal will be by a treatment plant existing at the fuels processing plant. Non-radioactive refuse may be disposed of by burning.

5. Storage facilities will be provided for raw materials and finished products, and shipping and receiving facilities will be provided.

6. Office space and facilities will be provided. Laboratories for the analysis and control of process variables will be provided. A clock house and wash and change facilities will be provided. An allowance for a watchman's security system will be made. An allowance will be made for fire protection alarm and fighting systems other than standpipe, pumping stations, and fire truck. A cafeteria or kitchen for the employees will be assumed to exist in the existing fuels processing plant. A maintenance and repair shop will be provided for routine maintenance of the process. Heavy maintenance will depend upon shop facilities in the existing fuels processing plant.

7. Flowsheets, item lists, and capital cost estimates for the process will include the study of the following categories: vessels, mechanical equipment, instruments, piping, electrical insulation, painting, equipment foundations, and structures.

8. Construction costs will include provision for supervision, insurance, taxes, permits, medical expenses, temporary building construction, temporary utilities and equipment, welding shop, clock house, watchmen's equipment, transportation and communication, construction equipment and tools, preliminary operations, testing and inspection, and start-up operations.

9. Engineering - Estimates will be made of the necessary categories of engineering required for the construction of a given plant, such as chemical process engineering, laboratory work, flow diagrams, layouts, construction engineering, construction detailing, process detailing, piping layout, piping detailing, instrument layout, instrument piping detailing, electrical engineering, electrical layouts and details, insulation specification,

foundation design, foundation detailing, structural design, structural detailing, specification writing, blueprinting and photostating, travel expense, operations and construction manuals, and engineering overhead.

10. Contractor's overhead will be considered, as well as contractor's fee or profit.

C. The primary objective in this study will be that of producing packages of long-lived fission products for use or sale. A secondary objective of this work will be that of reducing storage costs of undesired fission products, probably by packaging these materials in the dry state so that they may be stored in a much smaller volume than would be possible if these materials were in aqueous solutions. The least important objective of these studies will be to arrive at a means of removing long-lived nuclides from the gross fission products to a degree which would permit the discharge of the remaining fission products in dilute aqueous solution to the environment after a relatively short cooling period.

IV. APPROACHES TO COST ESTIMATING

In estimating the capital requirements for a fission product separation and packaging plant, the requirements of the various plants will first be laid out in the form of flowsheets, together with appropriate material and energy balances. All items of equipment will then be examined for appropriateness of layout. Capital costs will be estimated for the fabrication and installation of each separate item of processing equipment, and estimates will be arrived at for all items, including building, office space, laboratory space, shipping, receiving, maintenance, and other required facilities by applying unitized costs to the total requirements for each kind of facility.

Operating costs will be taken to include depreciation according to an estimated life to be agreed upon later for all buildings and equipment. Other items considered in operating costs will be: payroll, maintenance, utilities, and raw materials. Each of these items will be estimated by an examination of the requirements of each individual process studied.

Variables affecting the sales price of packaged fission products will be considered, but no estimate of sales prices will be presented. Present considerations indicate that the following variables will be important in arriving at a sales price:

A. Demand for large sources of radiation for chemical, biological, etc., experimentation and for radiation processing of materials.

B. Alternative present sources of radiation, such as Co^{60} and other nuclides produced as a result of reactor irradiation, accelerators, and radium.

C. Present and future resources of fission products, as affected by past reactor operation and by the anticipated increase in nuclear power reactor operation.

D. Alternative sources of thermal energy, such as heat and electrical power.

E. Specific activity of the packaged fission products, as affecting bulkiness and dose rate, and applicability to various processing operations.

F. Freight rates for shipping heavy shielded containers.

V. PLANNING AND SCHEDULING

For the fission product separation and packaging plants under consideration, it will be necessary to base estimates of capital and operating costs upon the time schedules estimated for research, development, engineering, procurement, construction, and plant start-up. It will be assumed in arriving at estimates of capital cost that an orderly manner of step-wise completion of the various phases of work involved would be employed. It will be assumed that a certain amount of time will be available for necessary research and development in advance of the commencement of engineering activities, and that engineering, procurement, construction, and plant start-up may be overlapped in such a manner that each of the groups involved in the coordinated effort will be working under optimum arrangements.

No allowance will be made in capital cost estimates for the completion of these activities on a highly accelerated, or crash, basis.

VI. DESCRIPTION OF PROCESSES

In this section are described the general features of operation of the several fission product separation and packaging operations listed above. These processes will be examined and evaluated as the work unfolds on the technical and economic analyses to be conducted.

A. Co-precipitation of Cesium

Sodium or potassium ferrocyanide and nickel nitrate or sulfate are added to the aqueous waste streams from the fuels processing plant. A precipitate is formed of nickel ferrocyanide. The nickel compound should be present in the concentration of 0.005 molar for a cesium content of 0.03 grams per liter. During this precipitation reaction, the temperature must be kept below 70° C. It is reported that more than 99% of the cesium is carried down with the precipitate in this operation. The precipitate is centrifuged from the supernatant liquor and the resulting sludge is calcined at 600° C. The calcined sludge is then leached with water and the cesium taken into solution again. The cesium could then be evaporated from the water solution and packaged in the dry state. The path of the rubidium is unknown, and some potassium comes through with the cesium. The recovery of cesium from the original waste stream by this process is greater than 90%. This process was developed at the Hanford Works (4), (5), (6), and is being studied at the National Reactor Testing Station (3).

B. Ion Exchange for the Removal of Cesium

The investigation of $cesium^{137}$ separation from gross fission products by ion exchange techniques has proceeded at the Oak Ridge National Laboratory to runs on cold material and countercurrent ion exchange equipment. In this flowsheet, the irradiated fuel elements (NP or MTR) are dissolved in sodium hydroxide and the resulting solution contains sodium aluminate, cesium, rubidium, and some barium in solution. This solution is fed to a countercurrent ion exchange column in which the cesium is adsorbed and then eluted selectively. It has been claimed that the continuous ion exchange equipment which has been developed solves the problem of radiation damage to the resin. The ion exchange column is U-shaped, and resin is transferred hydraulically in a path countercurrent to the liquid flows. Streams are metered in for approximately one minute; at which time the plug valve in the middle of the U opens and a cylinder pulses the resin from one leg into the other leg. The streams flowing to and from the column and the pulse mechanism are connected through a timer in such a way that the unit operates automatically.

Sodium carbonate is fed in at the top of the left leg of the ion exchange unit in which the resin is continually moved upward. The sodium carbonate converts resin from the ammonium form to the sodium form, ready for re-introduction to the bottom of the left leg. A stream of ammonium carbonate and sodium carbonate leave the leg a little below the point of introduction of sodium carbonate. Somewhat lower down on the leg, ammonium carbonate is introduced. This converts all resin to the ammonium form and elutes cesium, which is carried out with excess ammonium carbonate at a point a little lower in the column. A stream of sodium hydroxide solution is introduced below the cesium take-off in order to wash aluminate from the resin. The alkaline feed solution resulting from the dissolution of aluminum fuel elements is introduced below the sodium hydroxide feed point, and the solution stripped of cesium is removed at the base of the ion exchange leg. Streams of water are introduced at various points in the column in order to maintain hydraulic balance of liquid flows and to separate some reagent streams from others. Cesium recovery of 99.99% is claimed in this unit. However, losses of uranium oxide powder from the original fuel dissolutions are thought to be excessive. A topical report on this process has not yet been written. Some cognate data can be found in work by Blanco, et al. (2), and Higgins, et al. (7).

C. Co-crystallization of Cesium from Fuel Processing Solutions

An additional process developed at Oak Ridge National Laboratory has for its objective the removal of cesium from fuel processing wastes by a co-crystallization process. This process involves caustic dissolution of the fuel elements or acid dissolution plus an acidity adjustment, followed by a treatment with an ammonium alum. The cesium replaces the ammonium ion in the alum, and the cesium-ammonium alum can be crystallized out of solution. Rubidium may be separated from cesium by fractional crystallization of the alum. The mixture of crystals and mother liquor from the crystallizer are separated by jetting the mother liquor from the crystals. $Cesium^{137}$ recovery is on the order of 85% to 90%. A topical report on this process has not been written. Mr. Rupp of Oak Ridge National Laboratory expects to deliver a paper on the process at the Geneva Conference in August, 1955.

D. Complexing and Solvent Extraction

A method is being developed at the Engineering Research Institute, University of Michigan, by M. E. Weech which has as its objective the removal of relatively pure chemical species from a stream of gross fission products in aqueous solution. This method has as its basis the use of a complexing agent, or agents, at selected values of pH, which will combine with selected chemical elements, and the resulting complexes may be separated from the remainder of the aqueous solution by solvent extraction using an organic solvent. Some general information on such processes is given by Martell and Calvin (9).

The chief objective here is to isolate selected alkaline-earth and alkali metal ions in pure form so that compounds of these materials may be packaged in the dry state.

The equipment required for such a process would probably consist of a series of pulse-plate columns for conducting solvent extraction and subsequent stripping operations, together with equipment for drying and packaging the resulting isolated fission products.

Some work of this nature has been done on rare earth compounds by Topp and Weaver (11).

E. Packaging of Gross Fission Products

One of the serious problems encountered in the past in producing packages of dried gross fission products has been that of removing the water and other volatile materials from the gross fission products. Difficulties have been encountered in the plugging of evaporators and in the high rates of heat release which result in some concentrated solutions or solid beds of gross fission products. A solution to this problem which is contemplated for examination in this series of studies is a method under development at Argonne National Laboratories (1), and further described by Jonke, et al. (8) of Argonne National Laboratories, employing a new means for the drying of gross fission products. This procedure involves the use of a fluidized bed technique for the removal of moisture and volatile compounds.

Equipment which has been set up at Argonne for other purposes involves the use of a fluidized bed, about 6 or 8 inches in diameter, into which heated air is blown through the diffusion plate at the bottom of the equipment. An aqueous solution of the material to be dried is fed in at a point partway up the fluidized bed through special spray nozzles, which employ some air as well as pressure of the aqueous solution for the spraying of the solution into the bed. Fine particulates are removed from the exit air stream at the top of the equipment by means of sintered stainless steel filters. These filters may be blown back with compressed air periodically in order to free them from particulates. Problems of development remain in this method, particularly in starting up the operation. A sand bed has been used in start-up operations to provide a means of initiating fluidization. Some problems are also encountered in maintaining a desired range of particle sizes in the bed.

An alternative method for the dehydration and denitration of solutions of fission products is to evaporate the solution until a fused mass of nitrates remains. These might be cast into containers and allowed to denitrate by fission product heat. This method would be particularly applicable to the packaging of wastes which have not cooled for a long period.

F. Water Leaching of the Oxides of the Gross Fission Products

A method of reducing the volume of aqueous solutions containing fission products is that of evaporation to the dry state, as described under E., above. Studies have been conducted at the National Reactor Testing Station by the American Cyanamid Company as described by Stewart (10), which indicate the feasibility of leaching certain water soluble oxides from the oxides of the gross fission products. Cesium oxide would be water soluble and this method might therefore provide a simple means of removing and packaging cesium.

An aqueous solution of salts of gross fission products and of structural materials might be converted to a mass of solid oxides by an evaporation and heating process. These operations could be conducted in a calciner or a fluidized bed. The solid oxides could then be contacted with water in an extraction vessel and the solids separated. The resulting solution of water-soluble oxides of fission products could then be dried and packaged, or else subjected to further separation procedures in order to isolate compounds of specific fission products. It seems likely that only soluble hydroxides could be dissolved by this method, such as $Ba(OH)_2$ and CsOH. $Sr(OH)_2$ might not come out by such methods.

VII. DEVELOPMENT PROGRAMS

It is anticipated that the application of each of the processes for the separation and packaging of fission products will require some further research and development. The areas requiring further investigation will be outlined for each process, and some estimate of probable scope of research activities and most promising areas of development will be outlined.

BIBLIOGRAPHY

1. Argonne National Laboratory, Chemical Engineering Division, Summary Report for October, November, December, 1953, ANL-5213, Feb. 1, 1954.

2. Blanco, R. E., Higgins, I. R., Kibbey, A. H., Oak Ridge National Laboratory, "Separation of Fission Products from Aluminum Waste Solutions by Ion Exchange Scavenging," ORNL-301.

3. Burgus, W. H., and Grimmett, E. S., "A Technical and Economic Appraisal of the Potential Production of $Cesium^{137}$ at ICPP," PPC-66, Feb. 16, 1955.

4. Burns, R. E., Brandt, R. L., and Clifford, W. E., "Removal of Cesium from Uranium Metal Recovery Wastes," HW-31442.

5. Burns, R. E., Clifford, W. E., and Brandt, R. L., "Recovery of Cesium from Purex Wastes," HW-31444, April 13, 1954.

6. Coppinger, E. A., "Rough Draft - Cost Survey of Cesium Recovery from Purex Waste Streams," HW-30503, Jan. 11, 1954.

7. Higgins, I. R., and Roberts, J. T., "A Countercurrent Solid-Liquid Contactor for Continuous Ion Exchange," C.E.P. Symp. Ser. 50, 87 (1954). (Unclassified.)

8. Jonke, A. A., Levitz, N. M., Petkus, E. J., Taecker, R. G., Argonne National Laboratory, "Fluidized Bed Process for the Production of Uranium Tetrafluoride (Green Salt) from Uranyl Nitrate, Interim Report," ANL-5363.

9. Martell, A. E., and Calvin, M., "Chemistry of the Metal Chelate Compounds," Prentice-Hall, New York, (1952). (Unclassified.)

10. Stewart, D. H., "A Proposal for Pyrolysis as a Process for Waste Disposal at ICPP," IDO-15065.

11. Topp, A. C., Weaver, B., Oak Ridge National Laboratory, "Distribution of Rare Earth Nitrates between Tributyl Phosphate and Nitric Acid," ORNL-1811. (Unclassified.)