HAO-46

ENGINEERING RESEARCH INSTITUTE

UNIVERSITY OF MICHIGAN

PROPOSAL-REPORT ON

PYROMETALLURGICAL MELTING INVESTIGATION

FOR

CORE FUEL ELEMENTS OF A FAST BREEDER POWER REACTOR

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Project 2300

Consumers Power Company of Jackson, Michigan
For The
Atomic Power Development Associates

March 31, 1955

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UNIVERSITY OF MICHIGAN

March 31, 1955

Mr. R. W. Hartwell, Director
Nuclear Power Development Department
The Detroit Edison Company
2000 Second Avenue
Detroit 26, Michigan

Dear Mr. Hartwell:

Accompanying this letter are four copies of a proposal-report entitled, "Pyrometallurgical Melting Investigation for Core Fuel Elements of a Fast Breeder Power Reactor." This proposal-report has been prepared during the past two months at your request and subject to certain understandings reached with your group. We regard the submission of this report as fulfilling our obligations under the current study contract, which is Engineering Research Institute Project No. 2300 with Consumers Power Company, Jackson, Michigan, for the Atomic Power Development Associates.

In addition to the proposal-report herein presented, we have conducted extensive investigations of reactor fuel processing systems for the past thirteen months. We have studied methods for partial treatment, as well as methods for complete treatment of fuels, paralleling some investigations reported by your group.(1) Our essential conclusions regarding the technical and economic feasibility of partial treatment of your fuels by high temperature methods are reported in HAO-23, report no. 2300-4-F, "Pyro-Extraction Plant and a Pyrometallurgical Process with Aqueous Leg," January, 1955. We have considered in the preparation of this proposal-report your stated objectives of considering partial treatment of core fuels by batch melting methods, as described by Mr. Stanford.(2) Partial treatment as used here refers to reconstitution of the metallic properties of irradiated fuel without meeting any fixed specifications for decontamination or removal of plutonium from the fuel.

We have a keen interest in conducting the research activities described in this proposal, but wish to call your attention to certain qualifications set forth below. In this proposal we are describing certain fields of activity and approaches which we regard as vital to the realization of an industrially operable pyrometallurgical processing plant. We are proposing essentially to conduct studies of container materials and of some of the properties of fuels containing residues of fission products not likely to be removed by currently known pyrometallurgical techniques. The attainment of these objectives we would regard as part of the information required to set up a complete pyrometallurgical process. We do not wish to propose the development of a simple melting process for partial treatment, since we do not regard such a process as a desirable objective, as discussed below.

Because of the scope and magnitude of the problems remaining to be solved before pyrometallurgical processing can be applied to a production plant,

the University of Michigan is not in a position to propose a program of investigation in which both the time table and degrees of progress are predictable. We regard the achievement of a working pyrometallurgical plant as a program requiring long term basic research and development efforts.

As a consequence of our investigations of methods of fuels processing, and as a result of some evaluations which we have placed upon these investigations, we do not believe it to be economically feasible at this time to conduct batch melting as a partial method of fuels processing for enriched core fuels of the type contemplated by APDA However, it is likely that a long term program of development of high temperature processing will result in improvements in technology and reduction of cost. The attainment of improved technology and reduced cost would probably place high temperature processing on a competitive basis of performance and cost compared with other methods of processing.

It is our conclusion that if decisions are made to pursue pyrometallurgical methods for processing fuels, that it is not believed possible to arrive at a total cost for development at the present time, since so many unknowns are apparent in the application of such a process. It is suggested that consideration be given to the following general areas of developmental activity for the realization of a complete pyrometallurgical process. The scope of this proposal, however, does not cover this complete program.

1. Establishment of physical and structural properties for metal recycled to fuel elements.

2. The development of an optimum pyrometallurgical process, considering materials of construction and material balances.

3. The possible integration of remote fuel element refabrication with an optimum pyrometallurgical process.

If a decision of policy is reached by APDA to enter the fuels processing business, then we believe that consideration must be given to a facility performing a complete fuel separation. Otherwise, it does not appear that a private industry can justify any investment in the fuels separation and recovery operation.

If you care to discuss this proposal-report with us, we will be happy to meet with you at your convenience.

Very truly yours,

John G. Lewis

John G. Lewis
Project Supervisor

(1) A. P. Donnell, Letter, "To Members of the Technical and Engineering Committee, " February 18, 1955.

(2) R. E. L. Stanford, "Memorandum to: Members of Technical and Engineering Committee, Atomic Power Development Associates," February 18, 1955.

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I. FOREWORD

The Atomic Power Development Associates have extended the contract between the University of Michigan and Consumers Power Company, Jackson, Michigan, in order to permit the preparation of recommendations for further work in fuel processing. Members of the faculty and staff of the University of Michigan have evaluated required areas of investigation for the recovery of fissionable materials from irradiated enriched fuel elements of the core of a fast breeder power reactor. Previous studies have been performed for the Atomic Power Development Associates relating to the technical and economic feasibility of fuels processing. The following fuels processing methods have been studied.

A. Aqueous chemical separations with conversion to metal of decontaminated salts from core and blanket fuel elements.

B. Bromine trifluoride volatility processing with conversion of separated salts to metals.

C. High temperature processes with melting and inorganic extraction, employing auxiliary aqueous system.

The final engineering study in this group was completed in January, 1955.

The specific requirements and stated needs of the processing system for the power breeder reactor under consideration are believed by APDA to indicate that conventional methods of processing are not suitable for this purpose. It is desired to reconstitute fissionable materials from the spent core fuel so that structural properties in the fuel elements are restored. APDA thinking is that a melting and recasting operation should achieve this end and that fission product de-poisoning is not necessary for the continued operation of the fast reactor.

Once the reconstituted metal is obtained, it must be refabricated by remote means and reloaded into the reactor. It is desired to achieve the lowest possible cost of fuel reconstitution and also to achieve a minimum out-of-reactor fuel inventory.

A method of fuel separation which is attractive, provided that removal of plutonium and of all fission products from core fuel is not required, is melting with or without fluxes.

The purpose of the investigations described herein is to demonstrate on a pilot scale a processing system of recovering fissionable constituents from irradiated core elements. It is hoped that suitable separations can be achieved from structural materials and from alloying materials.

The topics for investigation and outlines of work are presented herein.

II. JUSTIFICATION OF ACTIVITIES

A batch-melting process might be adequate for the reconstitution of irradiated uranium into form suitable for reuse in a fast breeder power reactor. Such a melting process may be simpler than fuels processes in use at the present time. This simplicity would be made possible because the fast reactor permits greater freedom in selecting structural materials and permitting impurities in the fuels. Since July 1, 1954, the pyrometallurgical research and development conducted in various AEC laboratories has been studied intensively by this group. Investigations, correlations of data, and analyses of results have received attention. Efforts have been made to apply the information obtained to the devising of a system suitable for processing the fuels of the APDA reference design reactor. Reports of the data obtained might be applied to separations required for a given reactor design. In order that data may be applied to full-scale reactor separations, it is necessary that separations experiments be conducted on a sufficiently large scale that material balances and solubility data for all pertinent components may be obtained. The variations of the several important components of the fuel with successive extractions conducted after successive irradiations requires consideration. The influence upon separations must be noted of the fuel element configuration, of the cladding material, of the alloying constituents and of the build-up of certain nuclides.

Consequently, if a fuels melting process for core material is to be used, investigations must be conducted in order to predict the properties of the resulting treated fuel.

If APDA decide to purchase fresh fuel from the Government and to return the core and blanket fuel to the Government for processing, it would be unnecessary to conduct the investigations outlined herein.

III. SCOPE OF WORK

The work proposed herein can be separated arbitrarily in two general categories of activity. These categories are, A., Establishment of Specifications for Core Fuel Elements, and B., Fuel Melting Studies. Schematic portrayals of the reprocessing schemes considered and of the required investigations appear in Figures 1 and 2.

A. Establishment of Specifications for Core Fuel Elements

It is proposed to conduct engineering calculations in order to determine the rates of build-up of alloying components and of certain fission product nuclides from the fuels. These materials will build-up because they are not likely to be removed by melting operations upon the irradiated elements Consequently, on subsequent cycles through the reactor, the concentration of these materials in uranium will increase. The objective of these calculations would be to indicate the probable compositions of core fuels after a series of cycles through the reactor. From these calculated analyses, a series of simulated core fuel samples will be made up. These samples will represent the properties of the particular fuel after given numbers of reactor loadings.

It is not within the scope of this proposal to perform the metallurgical tasks required to determine the suitability of these simulated reactor fuels for use during actual reactor operations.

B. Fuel Melting Studies

The series of investigations described below are required to demonstrate the suitability of batch melting processes for the reconstitution of core fuels. Facilities are available at the University which would permit batch melting studies to be conducted. These investigations on melting would be conducted concurrently with the work outlined under A., above, and would be expected to reveal the following information.

1. Heat and energy balances.
2. Material balances for uranium.
3. Distribution of selected fuel alloy constituents.
4. Possibilities of melting of fuel metal from cladding materials.

Initial work will not include extraction studies or core material balances for plutonium or for active fission products. The feasibility of evaluating a plutonium-distribution melting process will be considered further as soon as space becomes available in the University's new Phoenix Memorial Laboratories. It would be desired to undertake studies on plutonium in concentrations which will likely permit extrapolation of the data to full scale operations. If such studies are to be undertaken, it will be necessary to modify and extend the work outlined herein.

A perspective view of a contemplated experimental arrangement for conducting melting studies appears in Figure 3. Figure 4 portrays a possible flow sheet for the conduct of such melting operations. Figure 5 is one preliminary concept of a crucible arrangement for experimental melting studies.

IV. OUTLINES OF INVESTIGATIONS

The investigations which are considered in this proposal are as follows.

A. Selection of Crucible Materials and Sizes

Crucible materials which would be examined for suitability in this application are graphite, metal-lined graphite, graphite lined with carbides of the transition metals, oxide refractories, sulfide refractories, or carbide refractories. The selections of materials to be used are contingent upon mutual solubilities of uranium, fission products, plutonium, etc., and the materials of construction.

B. Melting Experiments

Melting experiments would be conducted in order to determine the corrosion resistance of crucible materials and the solubility of uranium in the crucibles.

C. Preparation of Samples

Representative samples of uranium alloys would be prepared. These would be made up of cold uranium and alloying elements in proportions required to represent fission products in irradiated fuel. Radio tracers would be added to the alloys if required.

Zirconium, niobium, molybdenum, and ruthenium are not ordinarily removed completely by melting or extraction processes. These materials would be added in non-active form to the uranium in order to form alloys. Various different compositions of alloys would be prepared which would best represent irradiated fuels. This series of samples would represent irradiated fuel after successive cycles through the reactor and separation facilities, considering the amounts of these materials removed in separations. Evaluation of the physical and nuclear properties of these samples would be conducted by others.

D. Flux Materials to be Considered

Various flux materials would be considered for use in melting operations if it becomes necessary to use extraction techniques in order to separate alloying constituents, structural materials, or fission products. Such extractants might be fused salts such as lithium fluoride, magnesium fluoride, calcium fluoride, sodium fluoride, zirconium fluoride; or molten metals such as magnesium, calcium, magnesium alloys, silver, cerium, or lanthanum. In each case consideration would be given to:

1. The effectiveness of extraction to be anticipated.
2. The cost of inventory of extractant and operating cost of using material.

3. The physical limitations upon equipment required for the use of a given extractant.

E. Melting Experiments with Flux

If it were necessary to use fluxes in order to separate fission products which might impart undesirable physical or nuclear properties to the fuels, then extractants would be selected with a given purpose in mind. The transition metals promise to be the most difficult to remove and it is likely that the nature of flux selected would be controlled by the necessity of removal of these materials.

F. Analytical Work

Analytical work of a radiochemical nature would be held at a minimum during the course of these investigations as described above. It will be necessary to ascertain the composition of the simulated irradiated fuels and to obtain crucible and uranium balances during the course of the melting. If fluxes or extractants were employed, and it were necessary to obtain accurately the relative distributions of the various fission products, then it would be necessary to add very considerable sums to the estimates given for the conduct of the required analytical work.

G. Resolution and Tabulation of Data

It will be necessary to review and test the data for consistency and conformity to required boundary conditions.

H. The Application of the Results to Hot Operations

Descriptions and estimates will be formulated of probable systems for full-scale fuel separation based upon the data and other information obtained from the foregoing studies. Attempts would be made to arrive at estimates of capital and operating costs required.

I. Plutonium Analyses

Plutonium handling is not contemplated in this work. The possiblity of studying the distribution of plutonium would depend upon the availability of facilities in the Michigan Memorial Phoenix Laboratory.

V. PROJECT EXECUTION

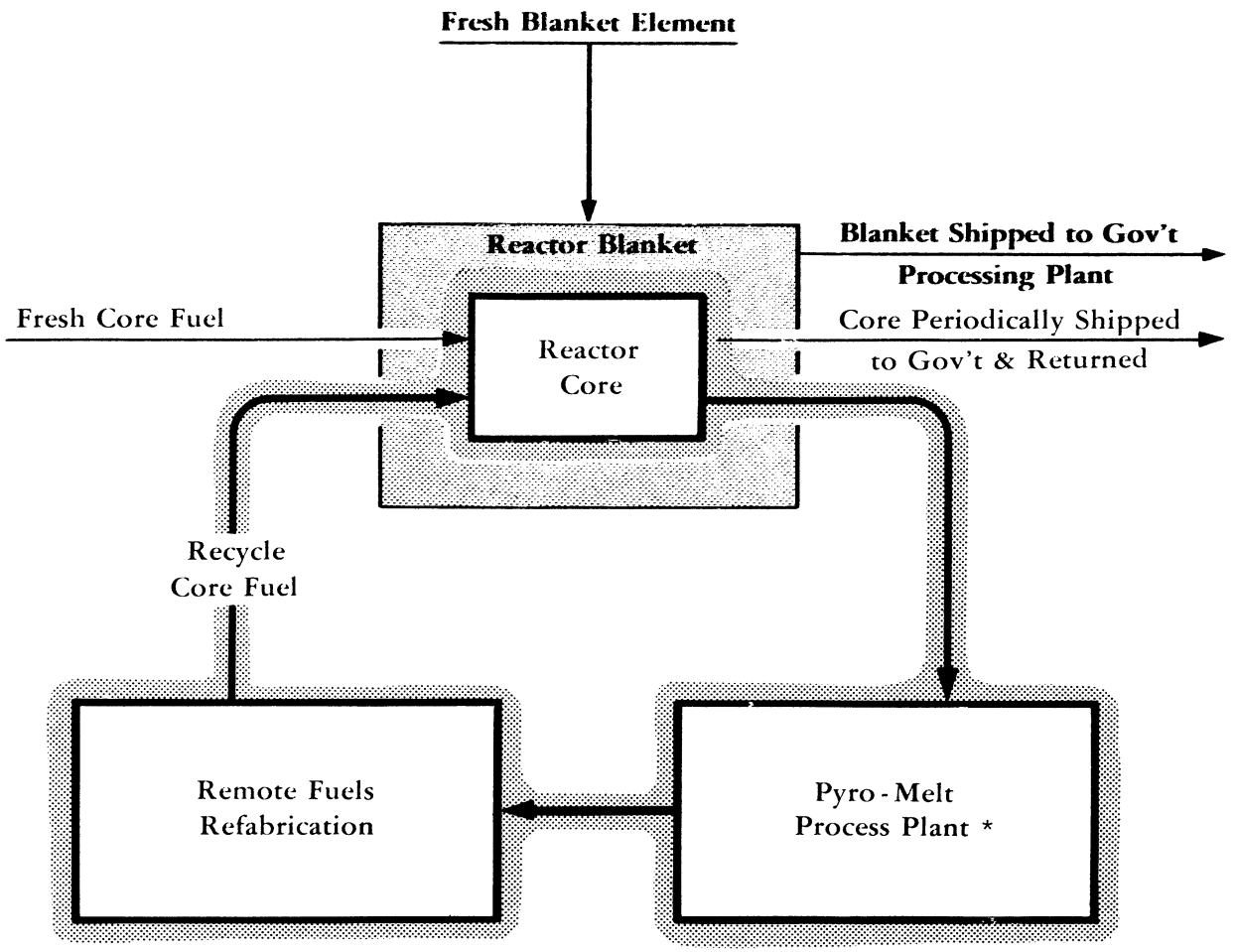
It is proposed that the work described above would be executed under a contractual arrangement between the Atomic Power Development Associates and the Engineering Research Institute of the University of Michigan. Under such an arrangement, project supervision would be the responsibility of Marx E. Weech. The faculty of the University and the staff of the Engineering Research Institute would be available to fulfill the needs for specialized and expert manpower and services. Undergraduate and graduate students would be called upon to assist in the prosecution of the work and in the formulation of the results. Professor W. W. Meinke, of the Department of Chemistry, would be available to assist in the establishment of the analytical work required. Professor H. A. Ohlgren would be available as advisor to the project. Professor David Ragone, of the Department of Chemical and Metallurgical Engineering, would be available to assist in technical features of metallurgical operations.

VI. SCHEDULES AND ESTIMATES OF COST

It is proposed that the time of one year be allowed for the completion of the foregoing work. During the course of this year, it is estimated that a total of $76,300 will be required to sustain the work. This amount will be the total of direct and indirect costs and would include the work described above. A tabulation of the costs is described below.

Total Direct and Indirect Payroll	$50,000
Equipment and Materials	21,300
Travel	5,000
Total	$76,300

REPROCESSING OF CORE FUELS FROM APDA POWER BREEDER REACTOR



Required Investigations:

To determine effectiveness of Pyro-Melt process and number of allowable Recycles before replacement with fresh fuel.

* Objectives of Pyro-Melt Plant:

1. To depoison fuels
2. To minimize fuels inventory out of reactor
3. To reconstitute metallurgical and structural properties of fuel

Figure 1

REQUIRED FIELDS OF INVESTIGATION & FUNCTIONS FOR PYRO-METALLURGICAL MELTING PROCESS

EXPERIMENTAL SIZE 10 KG / DAY

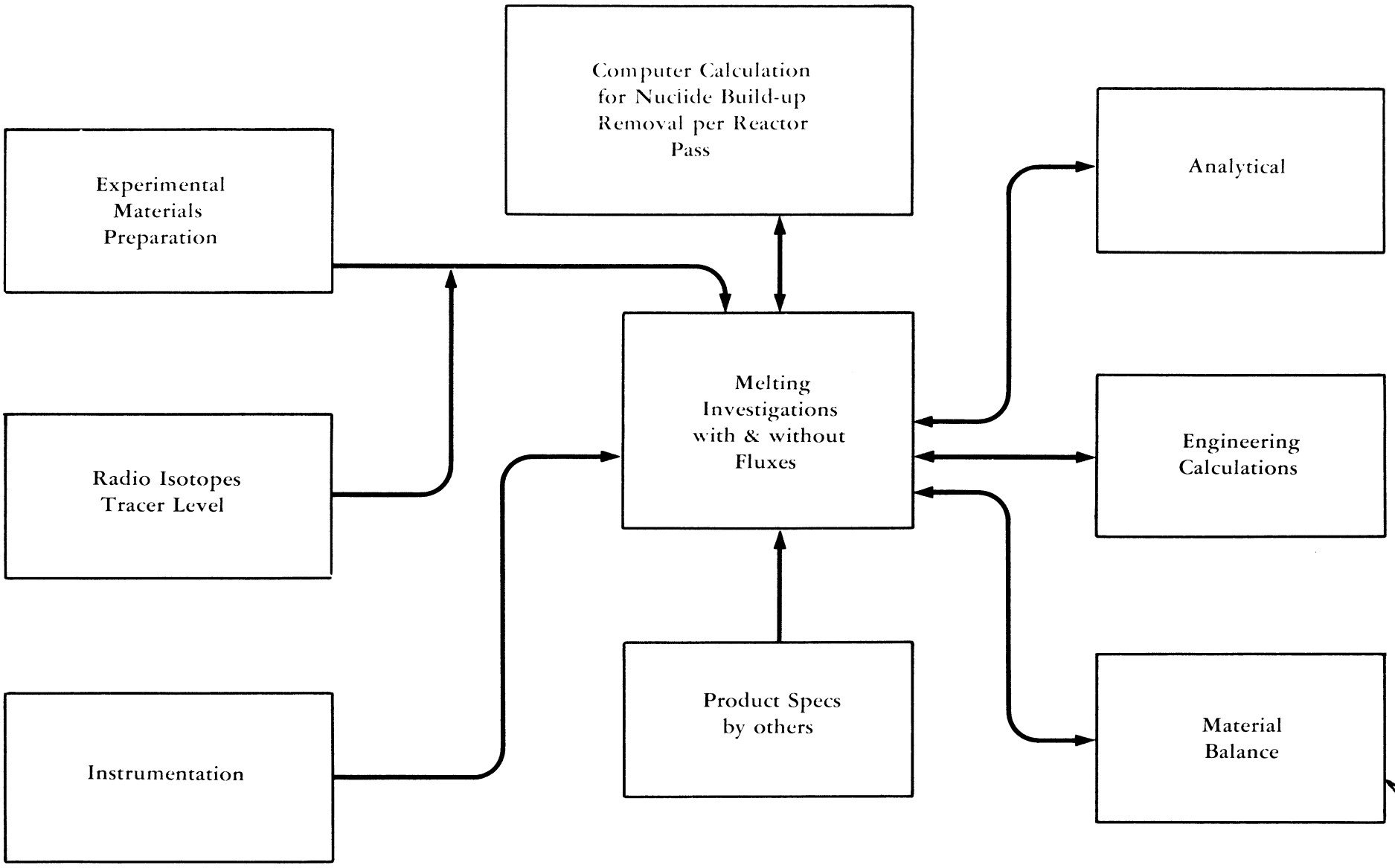


Figure 2

SECRET

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PERSPECTIVE OF EXPERIMENTAL SET-UP

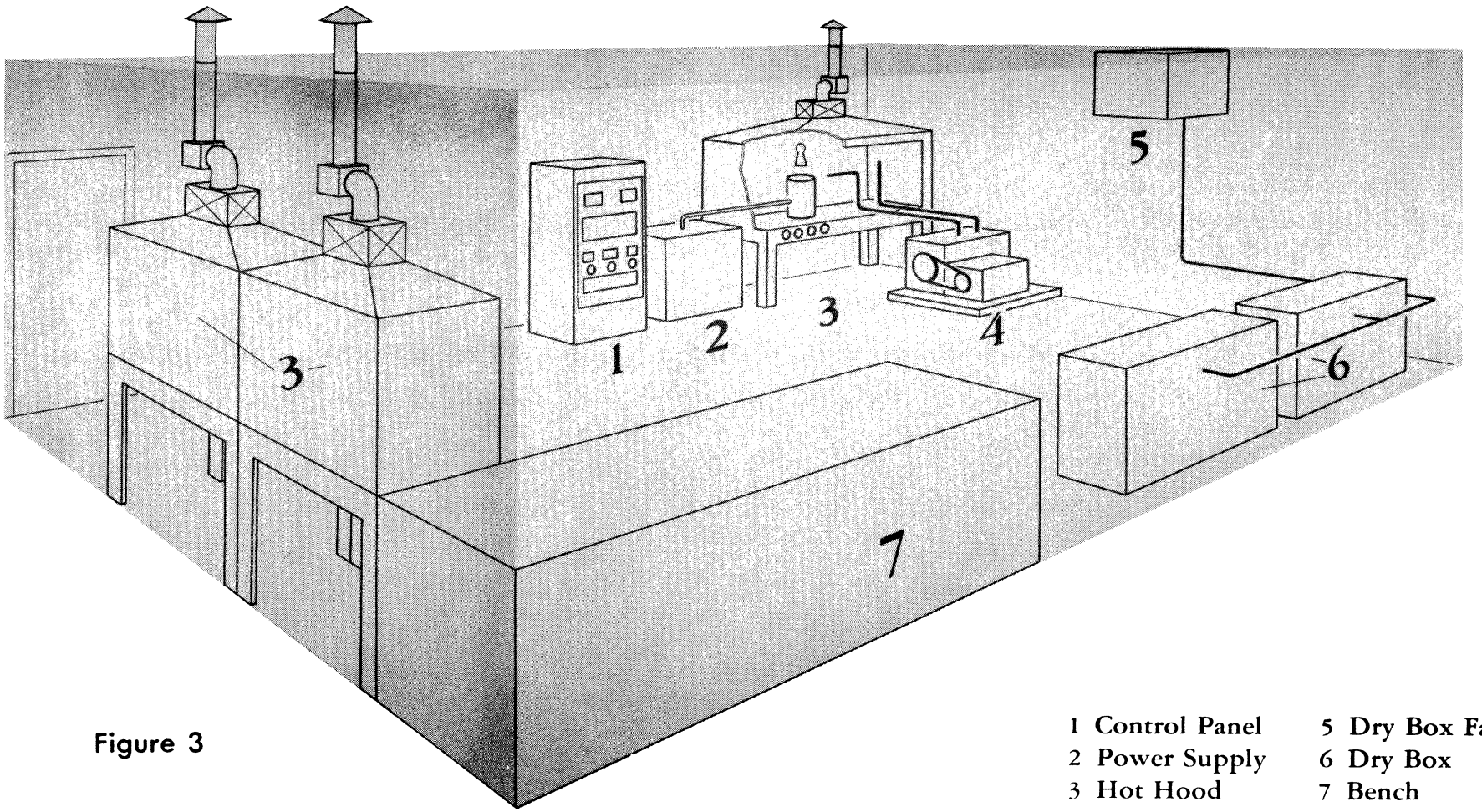
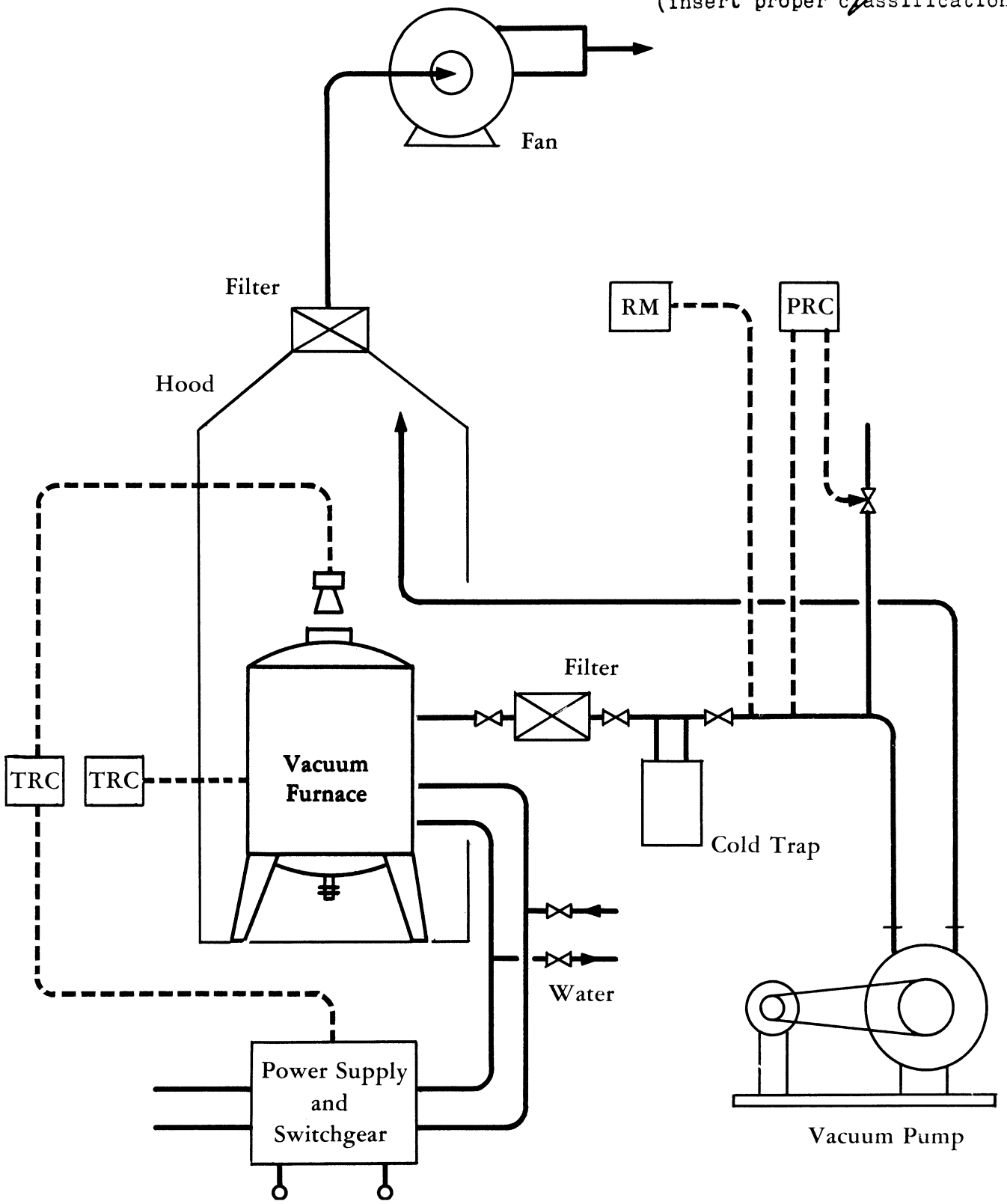


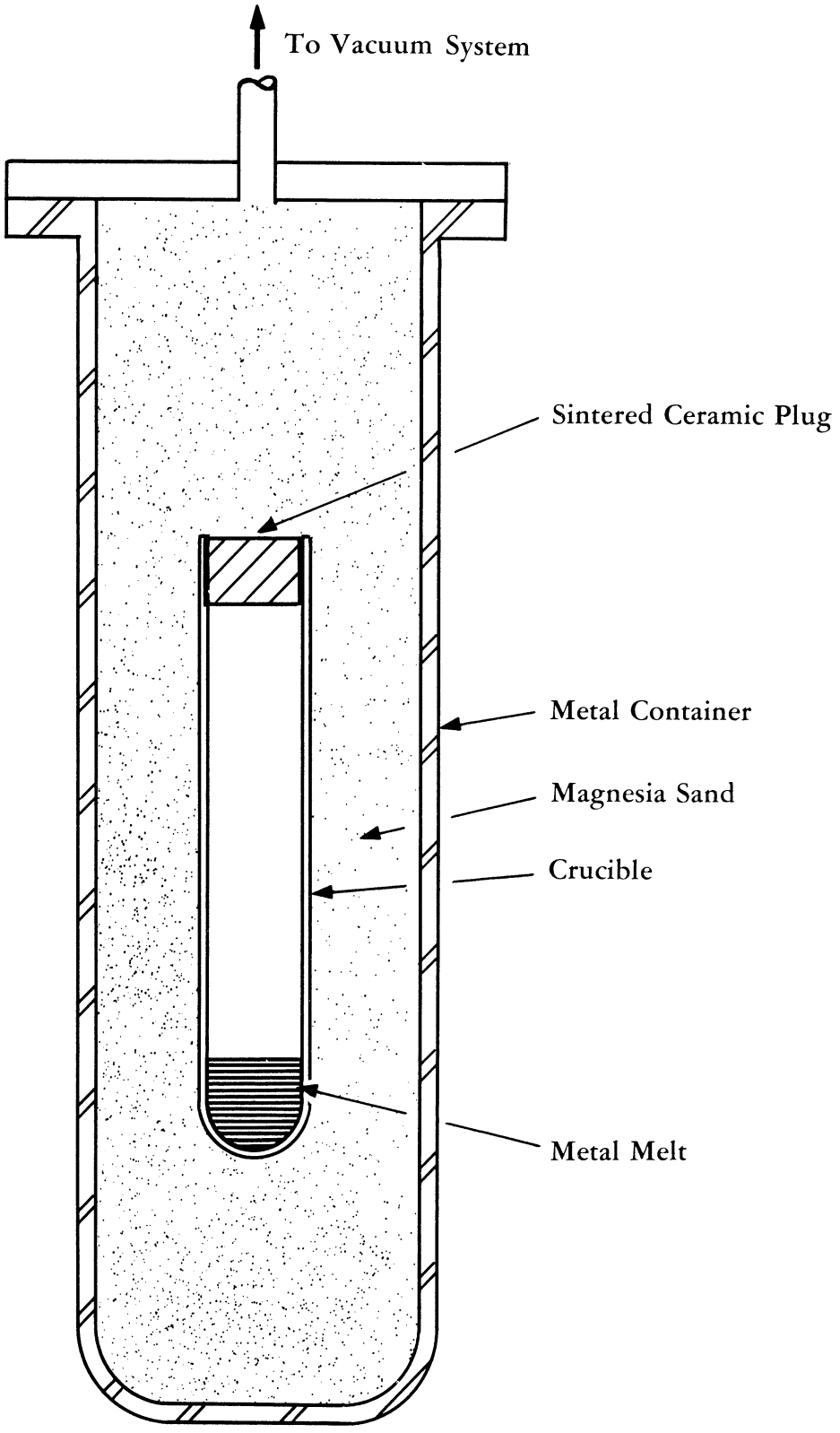
Figure 3

1 Control Panel
2 Power Supply
3 Hot Hood
4 Vacuum Pump
5 Dry Box Fan
6 Dry Box
7 Bench



FLOW SHEET FOR HIGH TEMPERATURE PROCESSING

Figure 4



CONCEPTION OF MELTING FURNACE

Figure 5