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PROJECT HOPE

A CHEMICAL REPROCESSING PLANT FOR A NUCLEAR POWER ECONOMY

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J. Halperin
R. J. Klotzbach
J. R. McWherter
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R. W. Stoughton
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RESEARCH AND DEVELOPMENT REPORT

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OPERATED BY
CARBIDE AND CARBON CHEMICALS COMPANY
A DIVISION OF UNION CARBIDE AND CARBON CORPORATION
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Contract No. W-7405-eng-26

PROJECT HOPE

A CHEMICAL REPROCESSING PLANT FOR A NUCLEAR POWER ECONOMY

by

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E. P. Wigner, Chairman

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JAN 21 1954

OAK RIDGE NATIONAL LABORATORY
Operated by
CARBIDE AND CARBON CHEMICALS COMPANY
A Division of Union Carbide and Carbon Corporation
Post Office Box P
Oak Ridge, Tennessee

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December 19, 1953

Mr. A. M. Weinberg
Research Director
Oak Ridge National Laboratory
P. O. Box P
Oak Ridge, Tennessee

Dear Mr. Weinberg:

Enclosed you will find the summary report of Project Hope entitled "A Chemical Reprocessing Plant for a Nuclear Power Economy." Although it would not be correct to claim that the design of the Hope plant is complete in an engineering sense, we believe that in its present state the Hope plant demonstrates that it is possible to reprocess U^{233} for $1/gram. It should be pointed out that this charge for reprocessing is then lower than the anticipated refabrication costs of fuel elements which are given as $1.50 to $2.50 per gram. The chemical costs are firmer at this time than are the metallurgical costs.

The main purpose of this letter is to bring to your attention some suggestions which came out of the Hope study but which are much broader in scope than the remainder of the report since they really apply to all AEC engineering work.

1) Do not hand over the conceptual design of a group of engineers and scientists to even the most competent contractor for detailing and construction. The stories which result from such a procedure are amusing, and they grow in volume and flavor as they go around and around the Project. However, we have enough stories of this nature and their original cost is high.

No design is entirely ready before construction is finished and, as a rule, some problems of operation and maintenance remain unsolved even then. If the contractor has to solve such a problem he will do it without an intimate knowledge of the ideas and concepts of those who are responsible for the preliminary design. He will overcome the difficulty by adding some auxiliary equipment to that originally contemplated. This may necessitate the addition of further equipment and so on. It may also introduce elements into the design which make

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impossible the functioning or servicing of the originally planned equipment in the way initially planned. Very often those responsible for the conceptual design can resolve such minor problems without increased cost or complexity.

The group which receives the conceptual design can have no pride in its principles, in the purposefulness of the manner in which it overcomes problems. Its pride only too often turns toward building something big and intricate rather than something purposeful and inexpensive. As long as the principles of nuclear engineering are not common knowledge, it seems more appropriate to increase and change continuously the group responsible for the design by the inclusion of more engineering and drafting talent rather than to make an abrupt change of responsibility from this design group to an architect engineer.

2) Do not keep cost figures secret. Only if the costs of installations become generally known will there be a universal and wholehearted effort to reduce them. The importance of economy is obvious in commercial undertakings. When a large fraction of the national income is spent on military preparations, its significance is equally large in defense work. We cannot increase the fraction of the national income spent on defense by a factor of four, and it would be hard to increase the national income on short notice. One often feels that the effectiveness of the sums spent in our defense effort could be increased by a large factor.

3) If you make a person responsible for the design of part of an installation, no matter how large or how small, give him a tentative budget which you consider reasonable. Otherwise, he may err only too easily on the costly and apparently safe side. It is only natural, and also desirable, for everyone to think that he designs the most important part of an installation. He knows that the cost of his part is only a small fraction of the whole. It is natural for him to spurn economies and novel ideas which will reduce the total cost only by a small percentage but make the design of his part less safe or less easy. One cannot economize in the large; one must be economical everywhere!

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4) Allot ample time to the conceptual design and to the basic decisions. It is natural for an administrator to feel that nothing happens on his project as long as only some "longhairs" think about it. It is hardly surprising if he grows impatient that nothing tangible happens. However, the time spent by a competent group on the basic design is time well spent. Many of us are familiar with cases when a few weeks' delay in the startup of construction would have advanced the completion date a few months.

5) See to it that the basic design be given not only ample time but also ample talent. We try to acquaint the future engineer in our engineering schools with the laws of nature which underlie his designs. There is less accumulated experience in nuclear engineering than in almost any other branch of engineering, and in many cases all one has on which to base his design is some knowledge of the laws of nature. The laws of nature relevant to nuclear engineering are taught in our schools only in an inadequate way, and it will be necessary, in many cases, to equip the basic design groups with several "longhairs". This is often embarrassing because the "longhairs" working habits and ideas differ from those of the engineer and even the project administrator. It often leads to friction. All this may have to be contended with if the job is to be successful and is not to become what future engineers will consider to be a monstrosity.

In addition to the specialists, i.e., chemical engineers, it will be wise to have on the job - at least in consulting capacity - a senior physicist and chemist with a real interest in the problem and engineers with varying backgrounds and areas of interest.

Sincerely yours,

Eugene P. Wigner
Chairman, Project Hope

EPW/cb

[redacted]

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I. Summary of Results

A rather embarrassing situation exists at the present time in that the cost of recovering a gram of enriched fissionable material from a spent fuel element is very often as large as the initial cost of producing a new gram of fissionable material in the diffusion plant. It is evident that if we are ever going to establish a nuclear power economy based on heterogeneous reactors which use metallic fuel elements, it will be absolutely necessary to drive the costs of reprocessing down from the present high levels to such a point that the chemical costs represent only a small fraction of the total cost of power.

We feel fairly certain that a very substantial fraction of the cost of reprocessing at the present time is due to a combination of factors which is an outgrowth or holdover from our weapons' economy and which will not be present in a power economy. In fact, if one examines in detail the design of all existing chemical plants he finds that a very substantial fraction of the capital and operating expenses can be charged to need for SF accountability, preoccupation with criticality considerations, and uneconomic though well-engineered waste storage systems.

Since we felt at the beginning of the summer that there were enough unknown factors in chemical plant design that it should be possible to markedly reduce the cost of reprocessing by adopting a rational point of view, we set ourselves the problem of designing a completely rationalized reprocessing plant. This work has come to be known as Project Hope.

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In order to have a definite chemical system to study, we have chosen a reactor which is probably not very realistic as a power producer, but one which has the advantage that we know both the design details of the fuel element and the cost and design data for the chemical processing plant in which this fuel element is presently handled. We have adopted an MTR type fuel element which is composed of enriched uranium aluminum alloy, but we have assumed that the U^{235} is replaced by U^{233}. Since any thermal reactor power economy will surely depend on U^{233} and not on U^{235} and since it is clear that physical operations with U^{233} are closely allied to the problems encountered in handling Pu, this assumption probably makes the study more realistic.

Before one can make an economic study it is necessary to choose a fuel processing rate. It is obvious from already existing studies that if one assumes a very large plant for study the unit cost will be low. In order to avoid this pitfall, we have chosen a plant which processes 5.5 kg equivalent of U^{233} per day. This is intermediate in size between the minimum and maximum rates for which the existing Arco plant was designed. For reference, these rates are about 1 and 8 kg per day.

This throughput is a convenient choice because it represents the reprocessing requirements of a power system which operates with a heat generation of a million kilowatts producing 200,000 kilowatts of electricity. The reactor required to produce this amount of power is more or less an average one and does not suffer from the criticism of being so large that the economic considerations involved in it will obviously

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lead to a low power cost. As an initial goal at which to shoot we assume that we are willing to pay 7 mills per kwh for power. Further, we arbitrarily assign about 0.9 mills per kwh for reprocessing. This turns out to be $1 per gram allocated to reprocessing for the assumptions of the study.

The chemical process we have chosen is solvent extraction using 1.5% TBP in Amsco. We chose this system over the Hexone system presently used at Arco because it is more flexible and does not require the precise concentration adjustment typical of Hexone. Laboratory studies and pilot experience show that adequate decontamination can be obtained with TBP in one extraction cycle.

The novel features of the Hope plant are continuous dissolving; relaxed, yet safe, criticality criteria; sampling only for process control; a realistic approach to waste storage; and a new operating-maintenance philosophy.

In the proposed design the processing plant is a concrete-lined pit which has a capacity of about 100,000 gallons. It is located outdoors and the top of the pit is formed of rolling concrete shielding blocks. The plant is operated dry but when maintenance is necessary the cells can be flooded with water. We have developed tools for the simple maintenance jobs which we know will be necessary and, in general, are very enthusiastic about the simplicity of design which is possible because of the maintenance philosophy.

[redacted]

Figures 1 and 2 summarize the economic results of this study for the most conservative assumptions which were examined; i.e., two cycles of extraction. For this case the processing cost is 96.1¢ per gram of U^{233} fed to the reactor; the total capital cost of the plant has been estimated to be $2,853,000. A physical plant has been designed to the point where we can make valid cost estimates for it. An artist's drawing of this plant is shown in Figure 3. It is clear from even the most cursory examination that this plant is simple in concept and small in size.

We believe that for fuel elements other than the MTR it will be no easier or harder to reach the same goal. It should be pointed out that $1 per gram is still high since no account has been taken of blanket processing, but we feel certain that routine process improvements will remedy this situation.

In summary, we can say that we are deeply convinced that it is possible to reprocess enriched fuel elements for about $1 per gram by being realistic in the plant design. We do not think that our choice of the MTR type fuel element abrogates the general applicability of this study to other fuel systems because it seems to be a general rule that the inert material is the controlling factor in the chemical process. Although by trading aluminum for zirconium or stainless steel one introduces new problems, at the same time you simplify some of the problems peculiar to the Al system. We hope that this study has broad general application to all of the chemical reprocessing problems of the AEC.

FIGURE 1

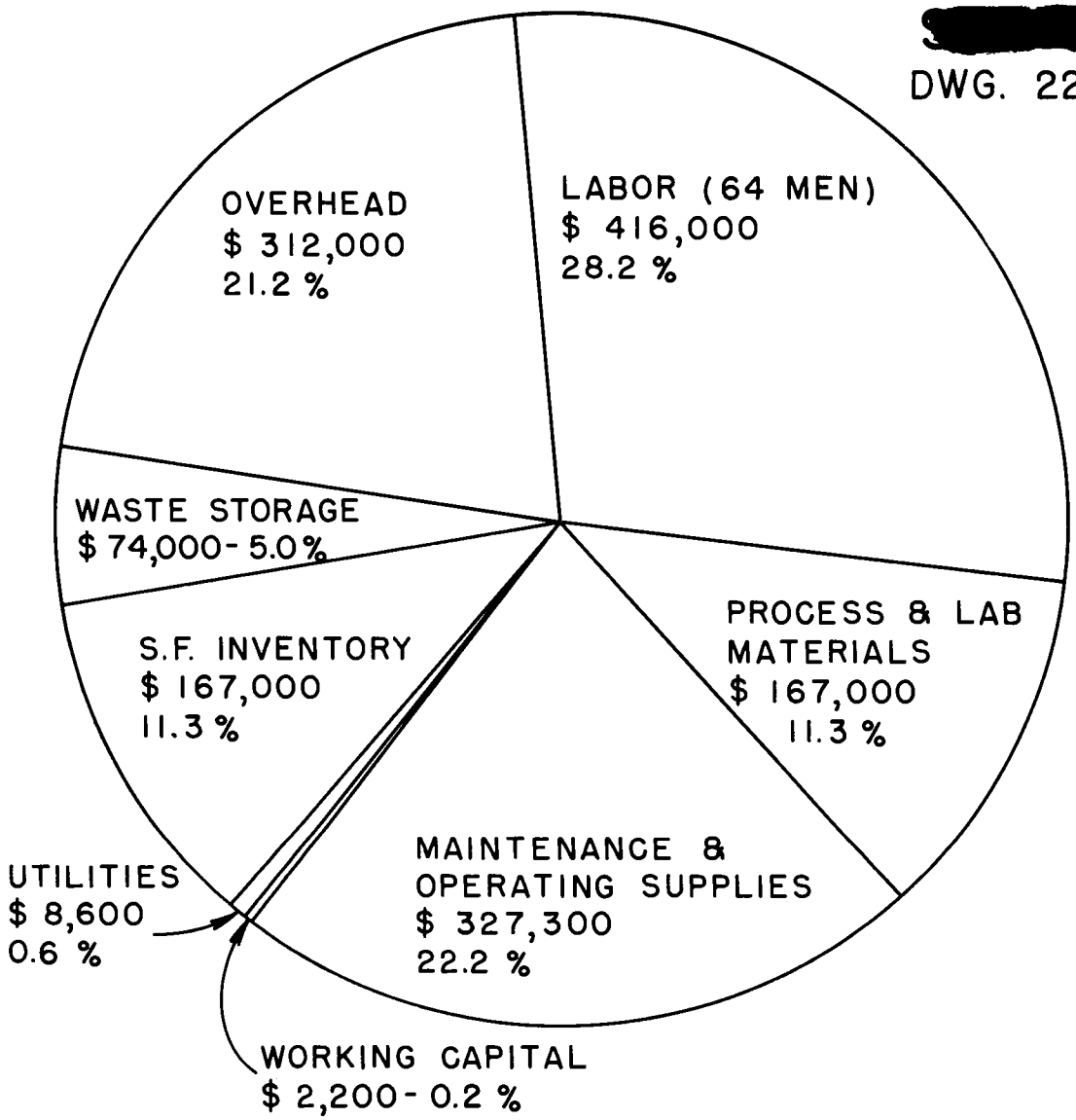
HOPE PROJECT COST BREAKDOWN

PROCESSING COST IS $0.961/gm U^{233} FED TO THE REACTOR

5.5 Kg U^{233}/DAY, 365 DAYS/YEAR

[redacted]

DWG. 22207



ANNUAL OPERATING COSTS

ANNUAL OPERATING COST = $ 1,475,000

$ 0.734 / gm U^{233} FED TO THE REACTOR

FIGURE 2

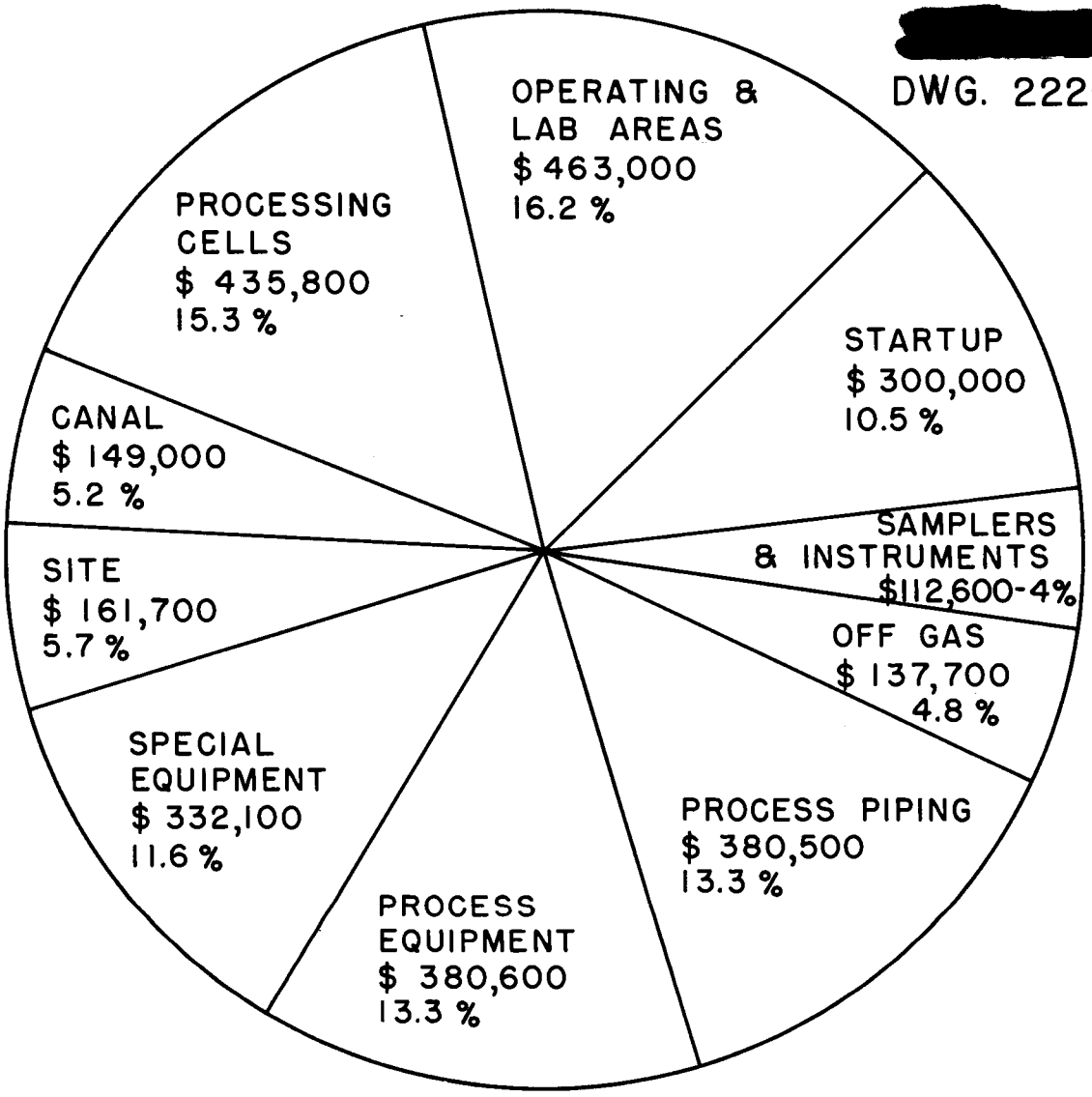
HOPE PROJECT COST BREAKDOWN

PROCESSING COST IS \$ 0.961/gm U^{233} FED TO THE REACTOR

5.5 Kg U^{233}/DAY, 365 DAYS/YEAR

[redacted]

DWG. 22206



TOTAL CAPITAL COSTS

TOTAL CAPITAL COST = \$ 2,853,000

ANNUAL CAPITAL COST = \$ 465,500

ANNUAL COST IS \$ 0.227/gm U^{233} FED TO THE REACTOR

Operating & Capital cost
Annual [illegible] 416 000

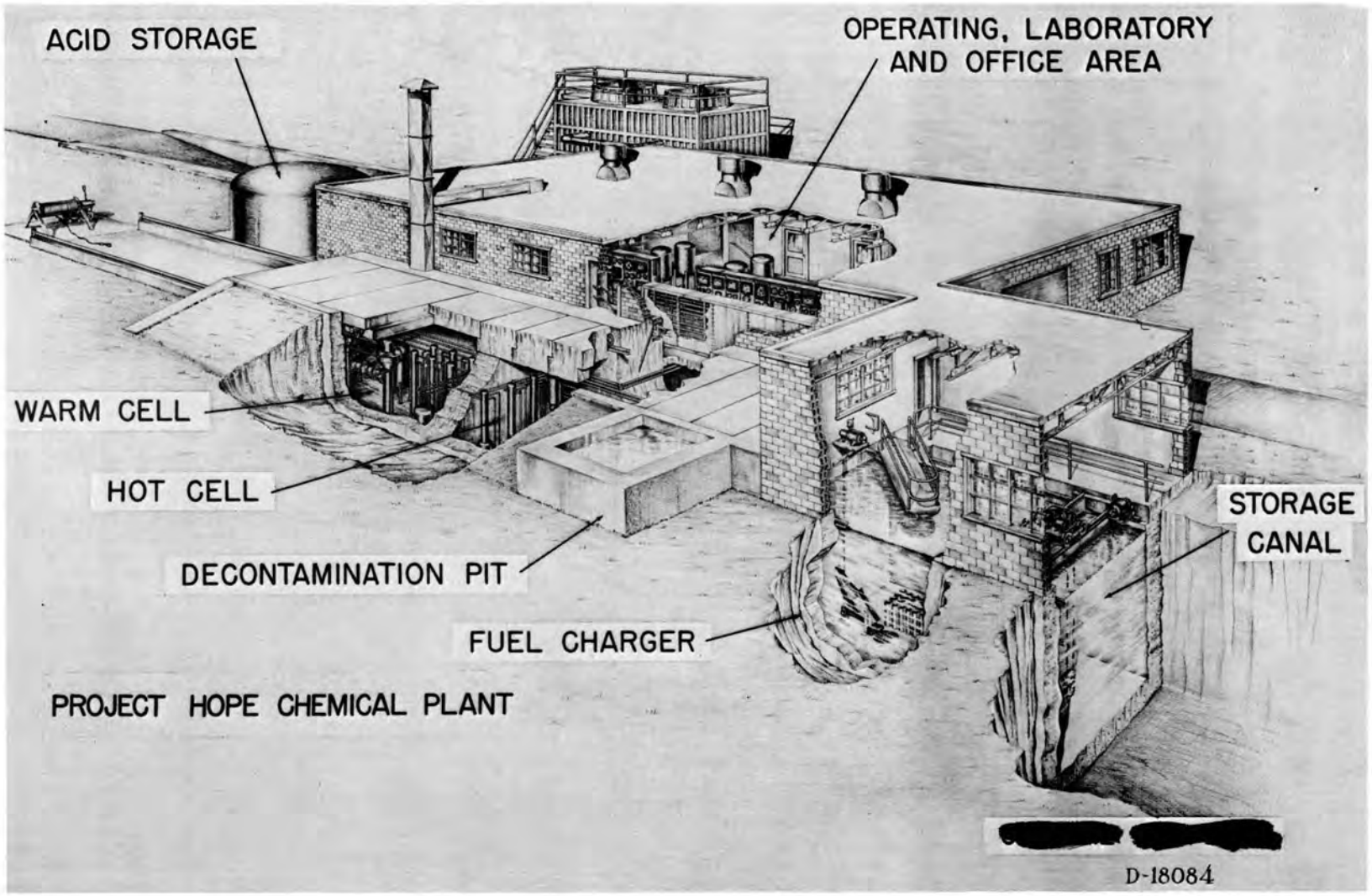

ACID STORAGE
OPERATING, LABORATORY AND OFFICE AREA
WARM CELL
HOT CELL
DECONTAMINATION PIT
FUEL CHARGER
STORAGE CANAL
PROJECT HOPE CHEMICAL PLANT
[redacted]
D-18084

FIGURE 3

[redacted]

II. Purpose of Study

A. Origin of the Problem

It is well known that the fuel of nuclear reactors deteriorates with use, partly because the radiation affects its mechanical properties and partly because of the accumulation of fission products and the depletion of the fissionable material. It is therefore necessary to withdraw the fuel from time to time from the reactor, recover the unspent fissionable material contained therein and purify it by separation from fission products. This purification will be an integral part of the operation of every reactor producing power and the cost of purification one of the expenses of the operation of power reactors. For this reason the cost of the purification has been reviewed recently both at ORNL and at ARCO. These reviews show that the cost of purification with the present chemical plants of the AEC can be as high, or higher, than the cost of producing an equal amount of fissionable material. Since this latter cost is higher than the normal price of the electricity that the fissionable material can produce, we are facing a serious difficulty in the production of economic nuclear power. There are several ways out of this difficulty.

1. Use nuclear power only in locations where the price of electricity is high enough so that it may cover the high cost of fuel purification.

2. Use those types of reactors which either (a) use cheap fuel originally and burn up a sufficiently large fraction of the original fuel charge to make reprocessing of the fuel unnecessary or (b) for which the fuel processing is naturally so simple that its cost can be reduced considerably.

[redacted]

3. Invent a new fuel purification process.

4. Reduce the cost of the present fuel purification process.

An example for 1 is the package reactor, examples for 2a and 2b are the high burnup natural uranium reactor and the homogeneous reactor. The present study aims at exploring the fourth possibility since it is necessary to know the potentialities of existing processes before one can decide whether or not the third alternative, above, is the proper course to steer.

B. Desirable Price of Purification

The average price of electricity may be estimated at 0.7¢/kwh. In order to produce 1 kwh of electricity, it is necessary to produce about 4 kwh of heat. This amount of heat is produced when

$$4 \times \frac{3.6 \times 10^{3} \times 10^{10} \text{ erg.} \times 235 \times 1.6 \times 10^{-24} \text{gm.}}{186 \times 10^{6} \times 1.6 \times 10^{-12} \text{ erg.}} = 1.82 \times 10^{-4} \text{ gm.}$$

of fissionable material undergoes fission (186 Mev is taken as the fraction of the energy of fission which appears as heat within the reactor). We shall assume that the material has to be reprocessed when 20% of it has undergone fission.[1] Hence, the amount of fissionable material that needs reprocessing per kwh of electricity produced is about 9.1×10^{-4} gm. If we allot to the chemical purification process .09¢, i.e., about 13% of the

[1] As will be brought out later, we are considering the reprocessing of MTR-type fuel elements which have been irradiated satisfactorily to a burn-up of greater than 80%.

price of the power we arrive at the figure of $1/ gm. fissionable material in fuel elements before irradiation as a reasonable cost of purification.

In our opinion, the above price is rather on the high side from an economic point of view. Most attractive power reactors have two distinct separation problems, that of the core and that of the blanket. It would be more reasonable for the two separations together to require 13% of the income. In those reactors which have no separate blanket, the purification of the core usually involves more complicated chemistry. It is our impression, therefore, that a further reduction of the price of processing is a continuing problem. In spite of this we have adopted the $1/gm. price as the aim of this first study.

C. Specification of the Problem

It would have been very difficult to tackle the problem of purification in general and the results of such a general study would have been, in our opinion, very unconvincing. It was decided, therefore, to study the purification of a well-defined fuel element at a well-defined rate. Every fuel element will have chemical and metallurgical characteristics which render the purification of its fissionable material easy, others which render it difficult. The designer of the purification plant will endeavor to take advantage of the former characteristics. We have done this in our problem. Naturally, different characteristics will have to be exploited when a purification system for an alternate fuel element is considered.

[redacted]

The MTR fuel element, with the usual U^{235} replaced by U^{233}, was chosen as the subject of this study. We made this choice because we have excellent cost data and adequate chemical information for this fuel element. Hence the problem is well defined. The physical composition of the MTR fuel element at the receiving end of the purification process is expected to be as follows:

4700	gm	Al
134	gm	U^{233}
33.6	gm	fission products
10	gm	AlSi alloy

Its dimensions are, roughly, 8 x 8 x 60 cm.

The price of purification decreases rather rapidly with increasing level of operation. In order to avoid the just criticism which could be leveled at us for choosing an excessively high reprocessing rate and thereby obtaining a very low unit cost, we have elected to consider a plant of roughly the same capacity as the existing Arco plant. We have arbitrarily assumed that we are processing fuel for a reactor which has an average power production of 250,000 kw, i.e., a heat output of 10^6 kw. This then gives, per day

$$1.82 \times 10^{-4} \times 24 \times 0.25 \times 10^6 = 1100 \text{ gm}$$

of fission products. As a result, we shall have to process, on the average, $\frac{1100}{33.6} = 33$ fuel elements with the total composition 155 kg Al,

[redacted]

[redacted]

4.4 kg U, 1.1 kg fission products, and some AlSi. The task which we undertook is, therefore, to show that the processing of fuel elements with this total composition and in the form of the MTR elements at the price of $1/gm fissionable material is possible. It may be well to anticipate here that we have at least convinced ourselves of this possibility. We have considered only methods and processes which are either already demonstrated or constitute only reasonable extrapolations from demonstrated processes and methods. Naturally, in order to account for temporary shutdowns, it was necessary to design for a capacity somewhat in excess of the daily amounts given above and we have chosen a safety factor of 1.25 when considering the size of equipment and when specifying processing rates.

The uranium content given above disregards the formation of U^{234} and is valid, therefore, only during the early part of the operation. As the operation proceeds, more and more U^{234} will be formed and this will give rise to the formation of even heavier isotopes. Under the favorable assumption that the fresh uranium from the blanket is pure U^{233}, and the unfavorable assumption that the U^{234} does not undergo fission in the reactor, the 4.4 kg U assumed above will be replaced, eventually, by 10.7 kg of this metal with the composition 38.5% U^{233}, 21.5% U^{234}, 3% U^{235}, 37% U^{236} and some Np and even Pu. This result assumes that the core's uranium is never discarded or sent through an isotope separation unit.

[redacted]

[redacted]

It also assumes that one wishes to maintain the concentration of fissionable material ($U^{233} + U^{235}$) in the fuel element at a constant level and that the permissible burnup of the fissionable material is not altered by the presence of inactive uranium. We based our considerations on the composition which prevails at the early part of the operation. However, we would not anticipate serious additional problems if the uranium content were increased to the higher figure of 10.7 kg/day.

The average power level of the associated reactor plant assumed above was chosen as a compromise, considering the desirability of a high figure, keeping in mind, however, that relatively few places in the U. S. can be expected to provide a market for very large amounts of additional power. Naturally, our final cost figure would have come out lower had we considered a purification unit of larger capacity.

D. Comparison with Earlier Cost Estimates

It is always difficult to compare cost estimates of different groups because of the varying conservatism and optimism of different groups. We have endeavored to make our estimates comparable with earlier cost estimates by making similar basic assumptions and using for equipment the costs actually paid for similar equipment at the construction of the ARCO plant. The price of chemicals was ascertained from manufacturers. The basic assumptions were:

[redacted]

[redacted]

1) Amortization of the plant and equipment in 6-2/3 years at an annual charge of 16% (amortization rate 15% and interest rate on one-half the investment at an average of 2%).

2) Inventory charge on the fissionable material received from the reactor at 4.5% per annum and at an assumed purchase price of $24/gm. Since we are concerned here with a non-weapons economy we will pay interest on the SF material only as if it were money.

3) An overhead of 75% on all wages and salaries.

4) No charge for the land occupied, which was assumed to be reasonably far from urban habitation.

5) We have assumed that we would pay for power, heat and light at conventional rates and that these would be furnished by the reactor and its facilities which we are servicing.

6) In contrast to earlier custom we consider the waste retainers as an operating expense. They should be built in reasonably large blocks when the need arises. Clearly it would be unreasonable to attempt to construct originally all of the waste retainers which will be needed during the anticipated lifetime of the plant.

7) The expense of fuel transportation was included in operating costs (cf. the end of this section).

8) The cost of extraordinary maintenance is included under "other" in the estimate below. Routine maintenance is included partly under payroll, partly under amortization.

[redacted]

[redacted]

The cost estimate given in IDO-14.062 ACCO for chemical purification is given below in $/gm for a 1 kg/day and a 12 kg/day plant, processing various types of fuel elements. Our plant capacity is about midway between the two.

	Amortization	Payroll	Chemicals	Utilities	Shipment of Fuel	Other	Total
1 kg/day	12.6	1.7	0.38	0.70	0.52	2.4	$18.3/gm
12 kg/day	1.46	0.74	0.37	0.11	0.51	0.99	$ 4.2/gm

One sees in particular the decreasing amortization costs with increasing plant size. The price of fuel and of utilities was left independent of plant size in the aforementioned report.

It would not be fair to compare the cost of our proposed process with the estimates given above. In this regard our assumption that the separation unit is quite close to the reactor so that only nominal fuel shipment charges will accrue is not a decisive factor (cf. 7 above). The principal five reasons that the comparison would be unfair are (a) that we attributed only a monetary value to fissionable material and could greatly relax the accountability requirements, (b) that our safeguards against criticality were based on a more detailed knowledge of the factors determining criticality rather than on rigid formulae heretofore accepted uncritically, (c) that we could make use of the experience which had accrued at ARCO and the other separation plants, (d) that we had the time and desire to pay close attention to cost figures, (e) no bombproof

[redacted]

[redacted]

construction was assumed. In addition to these principal reasons we should mention that the ARCO plant is designed to process five kinds of fuel elements. Although the dissolution system for two of these has not yet been incorporated and is not included in the above cost estimates, the greater adaptability of the ARCO dissolution system also contributed to its cost. It certainly adds to the price of the chemicals. It should be mentioned, on the other hand, that the waste disposal system which underlies the ARCO estimates is not yet complete and we have the impression that we have faced this problem more squarely (cf. 6 above). Similarly, we have tried to incorporate some process improvements which appear desirable in view of recent plant experience and which, naturally, add to our price. Some of these improvements, principally in the field of waste storage, will probably be necessary also at ARCO and will have to come out of future operating costs.

E. Tentative Allocation of Funds

We started our work with a tentative allocation of the funds which are available for the purification process, and the reader may gain in perspective by our giving such an allocation at the outset. The funds to be allocated amount to $5,500/day since the amount of fuel originally present in the elements to be processed was 5,500 gm. For the sake of comparison we give here, also, the final results of the study.

[redacted]

[redacted]

	Tentative Allocation		Final Result	
Amortization + interest	$1,750/day	32 ¢/gm	$1,145/day	20.8¢/gm
Waste retainers	200	3.7	200	3.7
Fuel inventory	450	8.2	450	8.3
Payroll	2,000	36	2,720	49.4
Chemicals	525	9.6	440	8.0
Other	575	10.5	140	2.5
TOTAL	$5,500/day	100 ¢/gm	$5,095/day	92.7¢/gm

It should be reiterated here that the comparison of these figures with those given for the ARCO type plant would be entirely misleading because the purpose and the whole conception of the two plants are entirely different.

F. Consequences of the Cost Allocations

The amortization cost of $1,750/day demands a plant not exceeding in cost

$$\frac{1750 \times 365}{16\%} = \$4,000,000$$

Since the cost of the plant and its amortization is, traditionally, the most important single item in our cost allocation, a further breakdown of the $4,000,000 available will be given here. For details, the reader is referred to the section on Cost Studies.

[redacted]

[redacted]

Building	$ 750,000
Laboratory, shops, offices	400,000
Site development	150,000
Chemicals storage	50,000
Process equipment	1,300,000
Other	100,000
	$2,750,000
Contingencies	550,000
	$3,300,000
Design	330,000
Preoperation and materials for maintenance	370,000
	$4,000,000

The wastes may amount to about 5,500 ℓ = 1,500 gallons per day, although they may be considerably below this figure. Hence the cost of the waste retainers should not exceed

$$\frac{\$200}{5{,}500\ell} = 3.6¢/\ell \quad \text{or} \quad \frac{\$200}{1{,}500 \text{ gallons}} = 13.3¢/\text{gallon}.$$

This also will be discussed below in much detail.

The fuel inventory of $450 demands that the holdup time before processing and the processing time together do not exceed

$$\frac{8.2¢ \times 365 \text{ days}}{\$24 \times 4.5\% \times .8} = 35 \text{ days}.$$

[redacted]

The factor 0.8 in the denominator originates from the fact that the plant receives only 0.8 gm of fissionable material, on which it has to pay interest for every gm of fissionable material in the fuel element before its use. Because of the 35 day restriction, we decided on a 30-day hold-up before processing.

We might remark here, parenthetically, that we consider the $24 price of fissionable material to be too high for the purposes of the present calculation. As long as the price of fissionable material has to be assumed to be higher than its value as fuel (around $12/gm at a $5/ton price for coal) - which will be for some time to come - the power unit which is in the background of our considerations should receive credit for the breeding which it is able to do. As soon as there is no premium on fissionable material beyond its energy content, its price should be reduced to the above figure. On the other hand, the interest rate of 4.5% per annum may be considered by some to be rather low.

A daily payroll of $2000 gives $60,000/month so that with an administrative overhead of 75%, $34,000/month would be available as net pay. This would permit the employment of about 60 people or an average of 15 people per shift. There will be disagreement as to the adequacy of this figure, and we admit that we find it more difficult to judge this than any of the other figures. We are convinced that 72 people are ample for the plant in question, except possible during the first few months of

[redacted]

operation, provided one is willing and able to employ people with the proper qualifications for the jobs required. One may possibly have:

1 chief supervisor	1
1 chief engineer	1
1 chief safety engineer (health physics)	1
1 chief chemist	1
1 shift supervisor per shift	4
1 shift engineer " "	4
1 safety man " "	4
3 analysts " "	12
2 mechanics-pipefitters per shift	8
3 operators " "	12

This would leave 3 men per shift free for other and occasional duties which would involve repairs, cleaning, trouble shooting and some process improvement.

The price of chemicals was obtained by adding the quoted prices (cf. below).

The item "other" in the allocation represents simply the difference between the funds available and those which we could definitely allocate, whereas "other" in the final result is the sum of all the miscellaneous charges which we know will accrue.

G. Specifications and Scope of Present Report

The functions of the purification plant as here considered are as

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follows:

Receiving the spent fuel elements as described under C above.

Returning the unused uranium in the form of uranyl nitrate solution, except for a permissible loss of 1/2%, at an activity level which produces a radiation field of less than 100 mr/8 hrs at a distance of 30 cm from 1 kg uranium.

Disposing of the waste products both liquid and gaseous.

Keeping the radiation in all accessible areas of the plant and its surroundings below 20 mr/8 hrs.

Providing personnel to help in the case of unforeseen difficulties in the rest of the plant and receiving help to a similar extent.

The plant will not be charged for the following services:

Land for plant and waste disposal area. The last item may amount to 1 acre/yr.

Technical assistance as specified above.

Security and guarding.

It was mentioned before that the purpose of the present study is merely to show that purification at the price of $1/gm is possible. Actually such proof can be provided, if at all, only by building and operating a purification plant at this cost. Hence the most that we could hope to achieve is to make it reasonable, in spite of adverse past experience, that the cost of fuel purification can be reduced to

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the above figure. The present study does not, of course, contain a design; the cost of a detailed design was estimated above at $330,000. However, we went through the whole process with all the care that we could muster in the limited time that was available and have every reason to believe that our cost figures are altogether realistic and contain the usual safety factors.

The principal by-products of the present study are a conviction that $1/gm processing can be achieved and the suggestions for further exploratory work which are given under Recommendations. Several of these have applicability not only to the purification of MTR fuel elements but to the problem of purification in general. They may prove ultimately of greater value than the body of the report.

H. Acknowledgments

We are very much indebted to Mr. F. S. Chambers of the du Pont Company and to the Chemistry Group of the Knolls Atomic Power Laboratory for a most useful and informative conference. We received assistance from members of the Chemical Technology Division of ORNL to an entirely unusual degree. We wish to mention in particular the assistance which we received from Messrs. F. L. Culler, C. E. Winters - who was particularly helpful in selecting the subject of our study - F. R. Bruce, F. N. Browder, D. E. Ferguson, E. Frederick, A. T. Gresky, H. E. Goeller, H. K. Jackson, E. O. Nurmi and above all, W. G. Stockdale both in the

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form of information and stimulation. A. C. Martinsen's knowledge of concrete was also most helpful. Finally, the Health Physics Division collaborated with us most wholeheartedly in the very difficult problem of waste disposal. Without reports provided to us by K. Z. Morgan, T. W. Brockett, R. J. Morton, R. F. Myers, O. R. Placak, E. G. Struxness, and many others we would have been greatly delayed in our study. Our thanks are due also to R. Richardson of the U. S. Weather Bureau for his council and for drawing our attention to the asphalt and tar-coated canals of the Bureau of Reclamation. We have used to an extent the underwater maintenance idea proposed and partially developed by the Vitro Corporation; consultation with their engineers gave us assurance that underwater maintenance is possible.

We should add a word to the question of whether we believe that our purification price of $1/gm is a rock bottom price or an intermediate price which we expect to be considerably reduced by further similar studies. It is natural for every group to believe that it went as far in its exploration as it is possible to go with the methods and tools that were at its disposal. We share this belief. However, we fully expect that the normal processes of standardization and of engineering and managerial efficiency will further undercut our price. We also realize that our prices are based on quotations and costs of equipment acquired without the customary bargaining and shopping around process and thus may be somewhat inflated. Finally, improvements of the process or new processes may also entail price reductions, even though

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we consider the solvent extraction process to be an excellent one, almost admirably adapted to the purpose and to remote operation on a considerable scale.

The dependence of the price of purification on the form of the fuel and on the degree of decontamination hardly needs mention. Its dependence on the level of operation was mentioned earlier.

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III. General Principles of the Equipment

A. Properties of Process Solutions from the Point of View of Criticality

The total fuel contained, before its use, 5500 grams of fissionable material and had therefore a total cross section of

$$\frac{5500 \times 0.6 \times 564}{233} = 8000 \text{ cm}^2.$$

If this material is mixed with substances with a total absorption cross section of $(\eta - 1)\ 8000 = 10{,}700 \text{ cm}^2$, its multiplication factor will be below one. The absorption cross section of 1000 grams of water is 20 cm^2; that of 1000 grams of steel is 30 cm^2. Hence, as long as the daily feed is dissolved in at least 500 liters of water its multiplication factor, i.e., the criticality constant of an infinite amount, will be below one. We shall see that the acid process stipulates its dissolution in about 3000 liters of water; the multiplication factor of the solution then will be 0.25 even for unspent fuel, i.e., well below one. The caustic process, which is the second choice at present, contemplates dissolution in about 2000 liters of water. In this case the multiplication factor of the solution of the unspent fuel elements would be about 0.36.

The above figures already show that the danger from criticality of the solution is remote and can materialize,[1] in the course of the process,

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[1] Although prudence dictates that nuclear incidents should be avoided in processing plants, it is worth noting that an accident will not, in general, be catastrophic in a plant such as ours. In unrestrained cylindrical geometry the large negative temperature coefficient of multiplying aqueous systems will surely quench any critical assembly before a significant hazard can develop.

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only if the concentration of the uranium in the liquid increases very substantially. This can happen if (a) some of the uranium plates out, perhaps in the form of UO_2, on the walls of one of the vessels; (b) if a precipitate forms which either increases the uranium concentration in part of the liquid or remains in the vessel to augment the uranium content of the next charge. The alternative (a) is not likely to affect criticality seriously. The reflection coefficient of an 0.4 cm tank wall for fast neutrons is below 0.1 so that of all the neutrons which originate from a uranium film on the wall more than 45% escape immediately. Hence, less than $\eta \times .55 = 1.28$ neutrons will be slowed down in the tank for every fission neutron absorbed in the film. Considering the absorption of the wall and of the water in the tank, this cannot give rise to a chain reaction.

Surely no tank larger than 1000 gallons = 3600 liters will be used in the process. Such a tank would naturally have a height of about 1.65 m. Since the solutions contain about 1.55 grams/liter of uranium, there will be - above 1 cm^2 - $1.55 \times .165 = .255$ gram uranium. The total fission cross section of this uranium is 0.37 cm^2. If all this uranium assembles in L cm near the bottom, possibly as a precipitate, the thermal utilization will be

$$f = \frac{.37}{.37 + .022\ L}$$

The fast escape, under the same condition, will be $\exp(-B^2\tau) = \exp(-33\ B^2)$ where $\tau = 33\ cm^2$ is the age in water, B^2 the buckling in the reactor.

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For this we shall assume $\pi^2/(L+5)^2$ with a 3 cm extrapolation distance on the upper side and a 2 cm extrapolation distance on the lower side. Hence the criticality constant becomes:

$$k = \eta\ f\ \exp(-B^2\tau) = \frac{2.33}{1 + .060\ L} e^{-330/(L+5)^2}$$

The maximum value of this expression is still well below one. We shall make therefore the further assumption that all the uranium in the vessel concentrates vertically in a layer of L cm and that horizontally in an area which is only half that of the base of the tank. Then the criticality constant will be as high as .95 as shown in the following table.

Table 1

L	f	exp	k
15 cm	.69	.46	.74
20	.625	.616	.90
30	.525	.777	.95
40	.455	.86	.91.

The reader familiar with criticality calculations will notice that we have made several approximations which are all in the direction of increasing the criticality constant k. Clearly one could go along the same lines and make increasingly less and less likely assumptions and obtain higher and higher criticality constants. We feel, however, that a gentle stirring with an air sparger (or an impeller if necessary) will render making even more extreme assumptions unnecessary. We believe that

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with this proviso the danger of criticality is zero as long as the tanks are not surrounded by reflecting material such as water. In order to estimate the effects of an infinite water reflector two-group calculations have been done for infinite cylinders. These results are given in detail in Appendix X.

A new problem will arise, however, in underwater operation or during underwater maintenance because the neutrons originating from a surface film of uranium and traveling away from the tank may not be lost with a probability of 90% but have a better chance of returning. As a possible way of guarding against the posibility of the vessel going critical when submerged in water, one could place a sheet of cadmium of the usual thickness (0.045 cm) on the surface of all tanks, which could conceivably accumulate enough uranium in the form of a surface film to become critical. It would not be desirable to increase the wall thickness of the vessel to provide neutron absorption by steel instead of cadmium. Such an increased wall thickness would also increase the neutron reflecting power of the tank wall and in some cases increase the criticality constant by a larger factor than would an equal volume of water.

The concentration and criticality of the solutions in the organic solvent are almost identical to those of the aqueous solutions.

Another procedure which we seriously recommend is to work nitric acid of about 20% concentration through the entire plant once a month instead of the regular feed in order to eliminate the possibility of

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uranium accumulation in parts of the equipment.

In the section entitled "Recommendations," we have advocated development of fissionability detectors, i.e., of instruments which permit the detection of substantial amounts of fissionable material. Such detectors would be a more rational answer to the criticality problem than overcautious design.

It may be worth remarking that no pipe of a diameter below 11 cm, and no sheet of a thickness below 23 cm, can become critical in a water medium by the slow neutron process. The critical mass for UO_2 through the fast process is higher than the total amount of fissionable material that goes through the plant in a week.

B. Properties of the Fuel Elements from the Point of View of Criticality

The unspent fuel element contains 168 grams of fissionable material which has a total cross section of 244 cm^2. If it has to compete with more than 325 cm^2 of other absorbing material, the multiplication factor will be below one. Our storage facility includes 8650 grams of iron and 4700 grams of water per cm height of the fuel element, with aggregate cross sections of 260 and 105 cm^2, respectively. We are convinced that the neutron density in the iron and water is equal to that in the uranium so that the multiplication constant for the storage facility is $2.33 \times 244/617 = .92$. Its criticality constant is, of course, considerably below this number. Considering that the above calculation

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is based on a 40 cm active height, instead of the actual 60 cm, and on unspent fuel elements we consider the fuel storage safe.

The fuel elements cannot become critical in the dissolver because a single row, or in fact a layer of unit thickness, of MTR elements cannot become critical. There is a possibility that some undissolved uranium-aluminum alloy accumulates at the bottom of the dissolver or of the digestor. The probability that a fission neutron emitted by the uranium in the tube reaches thermal energy in the tube is given by the expression $\exp(-B^2 \tau)$. For 5", 7", and 10" tubes, B^2 has the values $(2.40/6.35 + 3)^2$, $(2.40/8.9 + 3)^2$, $(2.40/12.7 + 3)^2 =$ 0.066, 0.0355, and 0.0235. The value of τ is $33/(1 - \rho)^2$ cm^2 where ρ is the volume occupied by the fines. Neglecting this factor, we find that the probabilities for the thermal neutron being formed inside the tube are 0.13, 0.33, and 0.485. The first case is surely safe without any additional precaution. If one uses tubes of 7" or larger diameter, it would be necessary to surround the tube with a cadmium sheet.

The above discussion is not meant to be complete; a similar discussion should go hand in hand with the continuation of the design. The importance of such a procedure is emphasized in the section containing our recommendations. This discussion of criticality was carried out in some detail because we hoped to show in this way that a rational approach to this problem permits one to overcome difficulties

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with very little change in the equipment which would be used for processing non-fissionable materials.

C. Radiation and Heat Production of the Feed

The reduction of the cooling time entails a considerable increase in the radiation and heat production of the feed. The following table gives, for the most important fission products, the disintegration energy, the energy content, and the radiation per day, in Mev, kev, and ev per fission, respectively.

The entry d in the third column indicates that the nucleus in question is the daughter of the preceding one. The decay factor already contains the yield; it is the average number of radioactive nuclei present after 30 days of cooling in a fuel which was in the reactor for the preceding days and in which one nucleus underwent fission. The following columns contain the β and γ energies in Mev per decay; the two succeeding columns contain the average energy content in kev resulting from one fission which occurred in the 30 days preceding the 30-day cooling period. The last two columns contain, in ev, the part of the energy emitted in one day. The last row indicates that our table accounts only for 0.65 fission chains, i.e., only one-third of all. The remaining two-thirds of the fission chains contain, however, only very long-lived and very short-lived nuclei with appreciable yields. The former are too inactive to be considered; the latter ones decay too completely during the cooling period. Nevertheless, we have increased

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Table 2

Nucleus	Half Life	Yield %	Decay Factor	Energy Mev/Decay		Energy Content kev		Energy ev/Day	
				β	γ	β	γ	β	γ
Sr^{89}	53 d	4.6	2.7×10^{-2}	1.5	----	40	-----	690	----
Sr^{90}	25 y	5.2	5.2×10^{-2}	0.54	----	145	-----	11	----
Y^{90}		d		2.2	----				
Y^{91}	57 d	5.9	3.6×10^{-2}	1.6	----	58	-----	700	----
Zr^{95}	65 d	6.4	4.1×10^{-2}	0.4	0.8	32	66	340	710
Nb^{95}	35 d	d	4.3×10^{-2}	2.4	0.8				
Ru^{103}	42 d	3.7	1.9×10^{-2}	0.2	0.55	4	10	60	164
I^{131}	8 d	2.8	8×10^{-4}	0.40	0.55	0.3	0.45	2.8	3.9
Xe^{133}	5.3 d	4.5	2.2×10^{-4}	0.3	0.1	0.07	0.025	10	3.3
Cs^{137}	33 y	6.1	6.1×10^{-2}	0.5	----	39	45	2.2	2.5
Ba^{137}		d		---	.67				
Ba^{140}	13 d	6.1	7×10^{-3}	1.0	0.55	17	21	900	1100
La^{140}		d		1.4	2.5				
Ce^{141}	28 d	5.7	2.1×10^{-2}	0.6	0.2	13	4	320	100
Pr^{143}	14 d	5.4	7.5×10^{-3}	1.0	----	8	----	400	----
Ce^{144}	300 d	5.3	5.3×10^{-2}	0.35	----	185	8	420	18
Pr^{144}		d		3.1	0.15				
Nd^{147}	11 d	2.6	2×10^{-3}	0.7	0.6	1	1	90	75
Pm^{147}	3.5 y	d	2.6×10^{-2}	0.2	----	5	----	3	-----
Total		65%				550	155	3950	2200

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the above sums by 20% for the calculation of the total heat production.

When calculating the energy production, the β energies were divided by three because two-thirds of the β energy is emitted in the form of neutrinos. Furthermore, to compensate for temporary shutdowns, the daily production was assumed to correspond to 1.25×10^6 kw days' heat production. Hence, the total heat produced per day by the fission products going through the plant is

$$1.25 \times 10^6 \text{ kw days } \frac{1/3 \times 3950 + 2200 \text{ ev/day}}{186 \times 10^6 \text{ ev}} \times 1.2 = 28 \text{ kw}.$$

The total energy content of the wastes will increase to

$$10^6 \text{ kw } \frac{1/3 \times 550 + 155 \text{ kev}}{186 \times 10^3 \text{ kev}} \times 1.20 = 2300 \text{ kw}$$

when sufficient time has elapsed to permit the decay products listed in the table to come to equilibrium. One sees that the apparent radioactive life-time of the feed is about 100 days, corresponding to a mean half-life of about 70 days. Naturally, this life-time of the waste solutions increases with increasing age. The temperature increase of the fresh process solution, assuming that its volume is 3200 liters for a day's feed, is

$$\frac{(28/4.2) \text{ kcal/sec}}{3.2 \times 10^3 \text{ kcal/}^\circ\text{C}} = 2 \times 10^{-3}\,^\circ\text{C/sec} = 7.5\,^\circ\text{C/hr}.$$

The heat capacity of the fuel elements alone is much smaller than that of the solution; namely, for 155×1.25 kg Al (the heat production refers

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to 1.25 days' average feed) 155 x 1.25 x 6.4/28 = 46 kcal/^{o}C. Hence the temperature increase of the uncooled fuel elements would be about (28/4.2)/46 = 0.14^{o}C/sec $\approx$ 8°C/min. If the fission products were separated from the Al and present in elemental form, the temperature increase without cooling would be about 370°C/sec.

The total energy output of the wastes, 2300 kw, suffices to evaporate the liquid intake, 5500 liters/day, about 15 times.

One sees that an increased cooling time would reduce the iodine, xenon, and Ba-La activities considerably. This would not reduce the total activity by an order of magnitude but would, nevertheless, reduce the activities of the most obnoxious products. The xenon and partially also the iodine escape during the dissolution process. The Ba-La gives the most penetrating radiation and increases the shielding needed. This is particularly onerous during maintenance when it demands, in the underwater type of maintenance, an increase in the height of the water level by a couple of feet.

We have found two concepts concerning radiation level quite useful. The first of these, the swimming water activity, is the activity of a medium, immersion into which gives 7.5 mr/hr to the object immersed. 7.5 mr/hr in air corresponds to an energy absorption of

$$\frac{7.5 \times 10^{-3} \times 1.60 \times 10^{-12} \times 32.5}{4.8 \times 10^{-10} \times 3600} \quad \frac{\text{erg}}{\text{cm}^3 - \text{sec}} = 2.26 \times 10^{-7} \frac{\text{erg}}{\text{cm}^3 - \text{sec}}$$

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since 1 r corresponds to 1 esu/cm ion pairs and 1 esu ion pairs corresponds, with an energy of 32.5 ev/ion pair, to 0.1 ergs. The energy production is, of course, equal to the energy absorption in a large medium. Hence, in "swimming water activity" air the energy production is the above amount. In "swimming water" the energy production is higher by the density ratio of water to air since the absorption coefficients are about in this proportion and the absorption in a unit volume of air, enclosed in water, is smaller by this factor than the absorption in water. The energy production in "swimming water" is therefore

$$\frac{2.26 \times 10^{-7} \times 1.13}{.1293 \times 10^{-3}} \frac{\text{ergs}}{\text{gm - sec}} = 2.0 \times 10^{-4} \frac{\text{ergs}}{\text{gm - sec}} .$$

We call this last figure, $\approx$ 1 erg/gm hr, swimming water activity in every medium. If we assume an average disintegration energy of one Mev, the number of disintegrations per gram for swimming water activity becomes

$$\frac{2.0 \times 10^{-4}}{1.6 \times 10^{-6}} = 125 \frac{\text{disintegrations}}{\text{gm - sec}} = 0.0034 \frac{\text{curie}}{\text{gm}} .$$

It follows from the energy production of the feed, given as 28 kw = 28×10^{10} erg/sec that the daily feed would have to be dissolved in $28 \times 10^{10}/2.7 \times 10^{-4} = 10^{15}$ cm^3 or one km^3 of water to bring it down to swimming water activity.

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The second concept which we have considered is the "drinking water activity". We have learned again from the Health Physics Division that it would be most dangerous to release fission products even of swimming water activity. The fission products contained in this water, particularly the Sr, might enter the circulation of organisms living in or drinking that water and finally enter human food. The health physicists have determined therefore that the maximum permissible Sr activity of water that is released in such a way that it is accessible to living organisms be

$$8 \times 10^{-7} \frac{\text{curie}}{\text{ml}} = 0.029 \frac{\text{disintegrations}}{\text{ml sec}} .$$

This is what we call drinking water activity. Considering the fraction of Sr present in the original fission product mixture, one sees that drinking water activity is a few thousand times lower than swimming water activity.

It may be useful to give here, finally, the irradiation to which the process vessels and the walls of the waste tank will be exposed. The former depends, of course, on the size of the vessel and will be relatively low in thin pipes. Its value at the surface of a large vessel is half as great as in the inside of it. The γ activity, in particular, amounts to an energy production in the inside of the solution of large volume

$$\frac{28 \text{ kw } 2200}{3520} \frac{1}{3200 \text{ liters}} = 5.5 \times 10^{4} \text{ erg/cm}^{3}\text{/sec.}$$

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Assuming an average γ energy of 1 Mev = 1.6×10^{-6} erg and an energy absorption coefficient of 0.03 cm^{-1}, gives an nv for the γ radiation

$$nv = \frac{5.5 \times 10^{4} erg/cm^3/sec}{1.6 \times 10^{-6} erg \times .03\ cm^{-1}} = 1.1 \times 10^{12} cm^{-2} sec^{-1} = 3.5 \times 10^{19} cm^{-2} yr^{-1}.$$

The nv at the tank wall is half as great.

The nv at the surface of a tube with a radius below a few inches is proportional to the radius of the tube. The number of γ produced per cm length of the tube is $\pi r^2\ 5.5 \times 10^4/1.6 \times 10^{-6} = 3.4 \times 10^{10}\ \pi r^2/cm^3/sec$. All these γ will have to cross the tube, i.e., go through a surface of $2\ \pi r$. Hence the number of γ crossing unit surface of the tube is $3.4 \times 10^{10}\ \pi r^2/2\ \pi r$ sec, but there is an obliquity correction of two. Hence the nv at the surface of a rather narrow tube is

$$3.3 \times 10^{10}\ r/cm^3/sec = 10^{18}\ r/cm^3/yr.$$

This already contains the factor 1/2. One should remember that this calculation is valid only if the liquid is at rest in the tank, i.e., if one does not replace the partially decayed liquid with a fresh one at the tank wall. Under this assumption it contains the β and γ radiation for all time to come, or rather for the number of years for the lifetimes of which the table contains radio-elements. The eternal radiation would not be much higher.

It is possible to reduce the irradiation of tank walls by numerous methods. One of them provides that the wall is covered with an inert

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material such as sand and the waste liquid fills only the interstices which may amount to 35%. This reduces the activity not only by the factor 0.35, but also by the change in bulk density 1.8. Thus the total reduction is by about a factor of five. One can further reduce the activity by filling the bottom of the tank with an inert liquid of high density, which will then act as a shield. Finally, one may hold up the waste solutions in a suitable radiation-resistant tank for a couple of months. This would give an additional factor 1/2.

D. Consideration of Problems Caused by Heat Evolution and Radiation

The heat evolution introduces problems in the storage of the fuel in the course of the process and for the disposal of the wastes. Only the last one is serious and will be discussed in Section 4 below. The heat evolution of the stored fuel is adequately taken care of by the natural circulation of the water in which it is kept. The heat liberated in the course of the process is eventually carried off by the ventilating air, by heat losses to the earth, and by the canal overflow.

The radioactivity presents, of course, many and much more serious problems. They arise 1) in the storage of the fuel, 2) in the course of the process, 3) in the disposal of the gaseous wastes, 4) in the disposal of the liquid wastes, 5) in the product, 6) in the recovery of process chemicals, and 7) during and as a result of failures and maintenance.

1) The radioactivity of the fresh fuel is so intense that it

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has to be transported and be stored in a 4 m deep canal. We have given some consideration to replacing part of the water shielding by lead but decided against it. The adequacy of about 340 cm water shield is shown in detail in Appendix III.

2) The principal shielding of the process equipment by the usual concrete walls. Enough experience has accumulated in this connection to make further discussion at this point unnecessary.

3) The disposal of the waste gases causes three types of hazards from radiation: the radiations from the noble gases which escape from the dissolver, the radiation of other substances escaping from the dissolver particularly iodine, and the radiation of particles entrained in the disposable and ventilating gases.

In order to cope with all these problems, it appears desirable to build a stack of about 60 m high even if no such stack should be required for the reactor. It should be noted that for this plant the stack can be economically made much higher than 60 m. The diameter of the stack required to carry the gas volume which is discharged is about 5 cm, so the diameter is set by engineering strength considerations rather than off-gas volume restrictions. Thus it would be easy, for instance, to erect a 5" diameter pipe 200 m in height for use as a stack. In our cost calculations we have not taken advantage of this saving - instead we have assumed a deluxe design stack 60 m high one half the cost of which we have assigned to the chemical plant. All

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the ventilating air and the outlets from condensers would be released through this stack. According to information received from the Health Physics Division, particularly R. F. Myers, the wind velocity at 60 meters is usually more than 1 m/sec--only about 4% of the time it is below 1/2 m/sec. The released gas will stay in the immediate neighborhood of the stack for 1/2 hr about 1% of the time; it will stay there 4 hours at the very most.[1] Particles which may be entrained with the stack gas will remain with the gas so long as their size is below 0.5 μ and this can be counted upon if the gas is filtered through CWS filters. These filters reliably remove all particulates above 0.3 μ size at an average gas velocity of 20 to 50 cm/sec. They have to be exchanged and disposed of and this is one of the problems which we have not investigated in sufficient detail.

The stack gases will probably come down to ground level at a distance of about 1 mile in the daytime and at a distance of about 6 miles at night. They will be diluted, by that time, at least by a factor of 100, usually by a factor of 10^5 in daytime.

In analyzing the problem in detail let us consider the noble gases first. It should be noted that the daughters of the radioactive noble gases are themselves not radioactive. The Xe^{133} constitutes the

[1] Graphs of the wind condition parameters are given in Appendix IV.

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bulk of these. We assume that the Xe^{133} will be swept away with a velocity of 1/2 m/sec. Its total effect will then be as if it stayed at the top of the stack $1/2\ \pi \times 60/(1/2) = 190$ sec. In view of the information above, this is a pessimistic assumption. The total γ energy production of this is, according to our table,

$$\frac{190}{8.6 \times 10^4} \times 28 \text{ kw} \times \frac{3.3}{1320 + 2200} = 0.055 \text{ watts.}$$

The energy flux at 60 m distance will be

$$\frac{55 \times 10^4}{12.6 \times 36 \times 10^6 \text{cm}} = 1.2 \times 10^{-3} \ \frac{\text{erg}}{\text{cm}^2 \text{ sec}}$$

With a mass absorption coefficient of 2.3×10^{-2} cm^2/gm or 1.75×10^{-5} cm^{-1} this corresponds to an energy production of $1.2 \times 10^{-3} \times 1.75 \times 10^{-5}$ erg/cm^3 sec $= 2.15 \times 10^{-8}$ erg/cm^3 sec (the attenuation of the γ rays in air is neglected.) This is about 10% of swimming water activity and hence quite tolerable. It should be mentioned, though, that in the "non-occurring" cases when the stack gases remain 4 hours at the top of the stack, the activity at the bottom will be about 8 times swimming water activity. Hence, the Xe^{133} activity constitutes an element which tends to make a larger purification plant more expensive by making it increasingly necessary to eliminate the noble gases from the off-gases by absorption or otherwise.

The noble gases' radioactivity presents some problems also to the surroundings. It seems, offhand, that if the activity is tolerable

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at the bottom of the stack, it will be tolerable also at greater distances. According to the usual rules, the volume which the stack gases would occupy at a distance of 1 mile would be more than $(100\ m)^3 = 10^{12}$ cm^3. This is far from enough dilution for a full day's discharge; in fact, it is only enough for a little more than a minute's discharge. It is conceivable even that the dissolver must not operate under particularly adverse atmospheric conditions.

Naturally, the noble gases are not resorbed by organisms and do not enter the life cycle.

We propose to have the radioiodine liberated in the dissolution absorbed in a silver nitrate bed while the entrained particles will be removed by a CWS filter.

4) The activity of the liquid wastes is, of course, the most serious and the most abiding of all radiation problems. It will be discussed in detail along with the methods we expect to use to cope with it.

5) The purification plant will discharge an average of 4.4 kg fissionable material per day as uranyl nitrate. This material will have to be kept in relatively small bottles or else sufficiently diluted to avoid criticality. If acceptable, we would prefer the latter method and leave the handling of this very dangerous material to the fabrication unit which must be set up anyway to handle it. The number of disintegrations of the above amount of U^{233} is

[redacted]

[redacted]

$$\frac{4400 \times .69 \times 6 \times 10^{23}}{233 \times 1.6 \times 10^{5} \times 3.16 \times 10^{7}} = 1.55 \times 10^{12}/\text{sec},$$

and the number of γ rays emitted is about 1000 times less. The γ energy is only about 20 ev per disintegration, i.e., $1.55 \times 10^{12} \times 20 \times 1.6 \times 10^{-12} = 50$ erg/sec. If the uranium is dissolved in 1000 ℓ $= 10^6$ cm^3 of water, which would make it safe as far as criticality is concerned, the γ energy would be 5×10^{-5} erg/cm^3 sec, i.e., as far as γ activity is concerned five times less than swimming water activity. The β activity also hardly exceeds swimming water activity. The real danger comes from the product entering the organism either through the mouth or the lungs and this danger must be rigorously guarded against. There are, however, well-established procedures for preventing such accidents.

The value of one average day's production at the price used here is $105,000; the value of one cm^3 of the liquid product specified above is only about 10¢.

6) The recovery of process chemicals, in particular of the Amsco solvent and the TBP, is a hot operation as planned at present. There is a good possibility, however, as will be discussed in the next section, of making this a cold operation.

7) The underwater type of maintenance was chosen to decrease the difficulty of maintaining and replacing equipment, and the rest of this section will be devoted to the discussion of the advantages and problems of this system.

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E. Maintenance Philosophy

It is the opinion of the group that a relatively inexpensive hot equipment maintenance scheme can be devised that will incorporate the advantages of the Hanford-Savannah River remote and the ARCO contact philosophies of plant maintenance.

One such scheme was proposed and partially developed by the Kellex Corporation for their underwater plant (cf. KLX-52, "Summary Report, Job 11 Research and Development"; KLX-54, "Job 11 Completion Report"). All hot equipment was to be continuously submerged in a basin of water and maintained by direct techniques through the water shielding cover. Preliminary testing of this maintenance method indicated that it was feasible. Unsolved problems, at the termination of the project, were the prevention of water inleakage to minimize solution dilution during equipment exchange, hot pipe connectors, thermal insulation, and the disposal of a large volume of highly contaminated basin water from a major breakdown. The ease of maintaining underwater equipment indicated such a scheme would be a desirable technique to incorporate in a high activity level radiochemical plant.

Our proposed scheme of operating the Hope plant dry and flooding in order to maintain by direct methods appears to circumvent the cost of the remote scheme, the exposure hazard of the contact scheme, and the disadvantages of the continuously flooded plant. In the following discussion, maintenance work means replacement of a defective

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equipment unit.

By operating dry the following advantages may be realized:

1) Vessel height and internal pressure limitations imposed on the Kellex underwater plant to minimize basin contamination via outleakage are removed.

2) Thermal insulation is necessary only where it would be used in a conventional radiochemical plant. Foam glass is an excellent insulator for this application.

3) Process solution leaks may be readily detected and returned to the process. Thus, small but significant leakage of fissionable material can be tolerated without requiring plant shutdown for maintenance.

4) Gross contamination of the shielding water is avoided by a mild decontamination of interior (and exterior surfaces if necessary) surfaces prior to flooding a cell for maintenance. Maintenance operations are conducted in such a way that inleakage of basin water occurs only during the short time required to disconnect and plug pipe and tubing connections. Thus it may be permissible to discard the shielding water without treatment for fission product removal. Some work has been done to investigate means of preventing inleakage of any shielding water into the equipment when exchanging piping or equipment units. Hydraulic crimping or freezing of trapped

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water in the line are attractive possibilities for positively sealing lines before disconnecting. The results of experiments using tools developed for implementing this maintenance philosophy are reported in detail in Appendix V. In general the preferred method is to disconnect the piping and immediately cap the open ends as this appears to be a rapid method that does not damage the equipment. Equipment requiring replacement is removed from the cell by motor crane into the decontamination pit where intensive decontamination prior to disposal or direct maintenance may be done at leisure. The repaired unit is then stored until needed.

By flooding to maintain the plant by direct means through the water shield cover, the following advantages may be realized:

1) Unlimited maintenance working time is achieved without a complete, time-consuming, and occasionally destructive decontamination of the equipment.

2) Maintenance work may be done directly with long-handled tools, thus eliminating expensive remotely controlled maintenance cranes, precision fabrication of all cell equipment, and an extensive inventory of prefabricated pipe jumpers.

3) Ordinary piping techniques, using bolted flanges for large pipes and Parker tube fittings for small lines, may be used.

Possible disadvantages of this type of maintenance are:

1) Plant down-time for replacement of a defective equipment

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item may be somewhat longer than in a remotely maintained plant because of the necessity for cleaning both the interior and the exterior of process vessels. However, the 20% down-time allowance should be adequate if intelligent attention is given to all details of equipment design, installation, preventive maintenance, and operation.

2) Equipment height limitations because of the minimum water cover of three feet over the empty, unrinsed equipment and the workable depth of about 20 feet for long-handled tools may restrict the design. However, most connectors on deep tanks can be considerably elevated above the cell floor so this limit may not be serious.

3) Shielding water contamination is an ever-present hazard.

All things considered, it appears that the proposed scheme for operating dry and flooding to maintain the plant offers significant advantages over previous schemes.

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IV. Review of the Work Done

A. Process Chemistry

The proposed process utilizes solvent extraction to recover the enriched uranium and to separate it from the fission products and the fuel element structural material. The process consists of continuous nitric acid dissolution of the fuel element, solvent extraction of the uranium using dilute tributyl phosphate in Amsco hydrocarbon diluent for the extractant, and stripping to produce an aqueous product solution 1.0 molar in uranyl nitrate. The dilute TBP flowsheet was chosen because of the wide range of concentration of acceptable feeds and the high decontamination demonstrated in one cycle. One cycle of solvent extraction will probably be sufficient for adequate decontamination, although plant cost estimates have been made for both one and two extraction cycles to satisfy the "let's not have to be careful in operating" school of thought. All chemical features of the flowsheet have been tested on a preliminary basis in the laboratory and appear to be at least 80% probable at this time. An abbreviated flowsheet for the Hope process is given on the next two pages. The complete flowsheet is is included in Appendix VI.

1. Dissolving and Feed Preparation

The fuel elements selected are similar to the latest type proposed for the MTR. These elements have 1/8" thick side plates with 19 fuel

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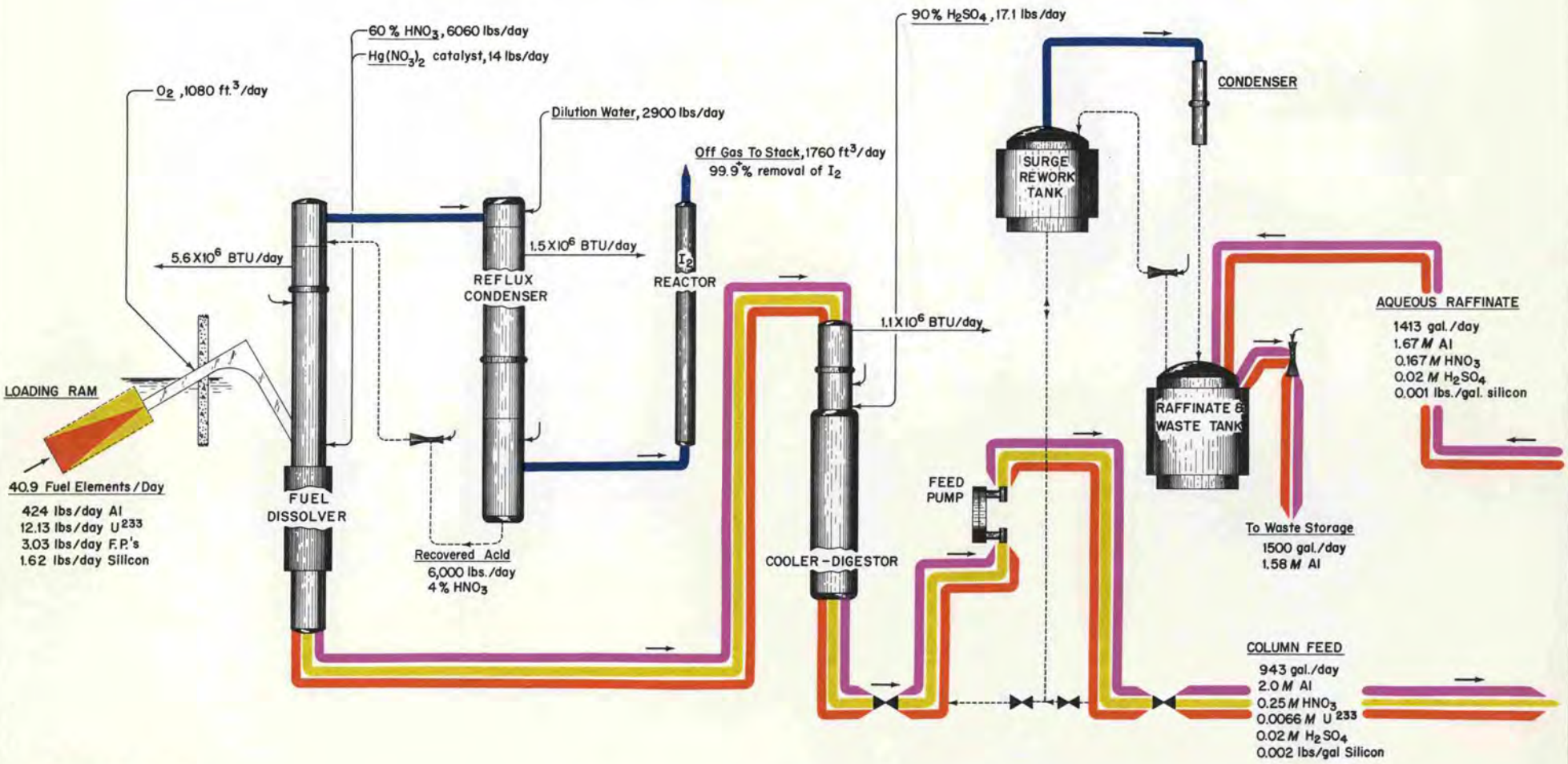
FEED PREPARATION



HOPE PROJECT CHEMICAL FLOWSHEET

CHEMICAL TECHNOLOGY DIVISION-12-53-JSF Dwg No 22212

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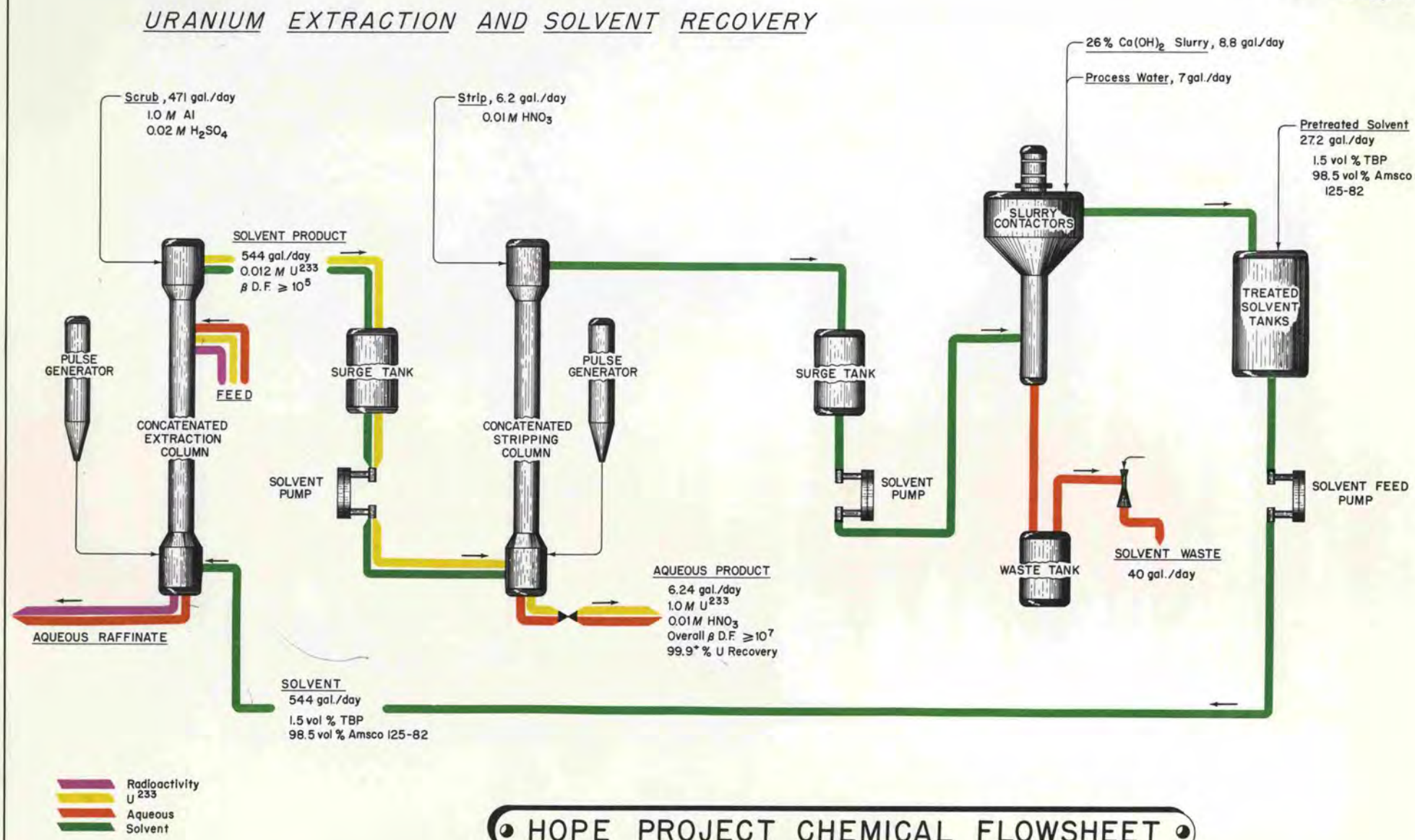

HOPE PROJECT CHEMICAL FLOWSHEET

CHEMICAL TECHNOLOGY DIVISION-12-53-JEF-Dwg No 22213

plates, each 50 mils in thickness. The assembly and fittings will be removed at the reactor and the 3" square by 26" long center section will be processed in the chemical plant. The approximate composition of each fuel element as given previously is as follows:

Aluminum	~ 4700 gms.
U^{233} + higher isotopes	~ 134.4 gms. (168 gms. of fissionable material initially)
Fission Products	~ 33.6 gms.
Silicon (from brazing alloy)	~ 10 gms.
Plutonium	insignificant amounts

The fuel elements will be dissolved continuously in nitric acid with mercuric nitrate catalyst to produce a solution 2.0 + 0.2M $Al(NO_3)_3$ and from -0.2 to 1.0M HNO_3. Oxygen is added to the dissolver to recover nitric oxide as nitric acid in the downdraft reflux condenser. Off gases are passed through a heated tower packed with silver nitrate-coated Berl saddles for radioactive iodine removal before filtration and discharge to the atmosphere.

The dissolving and acid recovery reactions are given in ORNL-1208 as follows:

$$Al + 3.75\ HNO_3 \xrightarrow{2\%\ Hg^{++}/Wt.\ of\ Al} Al(NO_3)_3 + 0.225\ NO\uparrow + 0.15\ N_2O\uparrow + 0.1125\ N_2\uparrow + 1.875\ H_2O$$

$$U + 4\ HNO_3 \longrightarrow UO_2(NO_3)_2 + 2NO\uparrow + H_2O$$

$$NO + 0.75\ O_2 + 0.5\ H_2O + \longrightarrow HNO_3 \text{ (net reaction)}$$

[redacted]

Silicon in the brazing alloy remains principally as a finely divided precipitate of elemental silicon and is discharged with the extraction column raffinate.

The rate of dissolution of aluminum in 2.0M $Al(NO_3)_3$; 0.25M HNO_3 solution is not reported. However, the mercury catalyzed dissolution of aluminum metal (reported in ORNL-1208) in neutral aluminum nitrate solution to prepare di-basic aluminum nitrate indicated a rapid initial reaction rate with an overall rate of only 0.2mm/hr due to the extremely high final Al concentrations (4.6M). Assuming penetration occurs only on one side of the fuel plate, complete penetration of the fuel plate should occur in 6-1/2 hours at the low overall penetration rate. With the 2.0M Al, 0.25M HNO_3 dissolver product solution contemplated, the violent circulation caused by the gas and heat evolution and the conservative assumption of penetrations from only one side, it appears probable that the fuel plates would be dissolved in 4 hours or less. Unless the improbable case of end-on-end stacking of 5 fuel elements occurs in the 12" ID dissolver, sufficient fuel elements can always be immersed in liquid to give reaction times not to exceed 4 hours. Of course, the thick side plates will not be dissolved as rapidly as the fuel plates. However, dissolution of the fuel plates will collapse the element and decrease the probability of stacking. The constant volume heel of aluminum and pieces of alloy presents no particular chemical or criticality problem.

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It should be noted here that there have been several different designs of dissolver under consideration. Each design is particularly suited to a given dissolving cycle. If it should turn out that the reaction time is markedly different from 4 hours, the dissolver which is shown in our present sketches would be redesigned.

Nitric oxide is recovered from the dissolver off-gases to reduce the net acid consumption and the volume of dissolver off-gas.

The recovery of nitric oxide as nitric acid involves simultaneous oxidation and absorption reactions. Oxygen, in 10% excess, is fed to the dissolver to oxidize the NO and to purge the slug chute. The recovery unit, consisting of the precondenser, the oxidation chamber and the downdraft condenser, was designed by analogy with the Idaho Chemical Plant Unit as reported in ORNL CF 51-10-20. Plant experience indicated essentially complete recovery of nitric oxide at comparable metal dissolution rates with considerable air inleakage and the normal flow of oxygen. It is believed that the assumption of 95% recovery of the nitric oxide is valid in the proposed sealed dissolver. It is possible that air could be substituted for oxygen with equal success.

Radioiodine-131 is removed from the dissolver off-gas to prevent discharge of hazardous amounts of iodine into the atmosphere. The reaction of iodine vapor with silver nitrate is essentially quantitative at elevated temperatures. The exact reaction mechanism is not known.

The iodine removal tower was designed by analogy with the Redox Plant Silver Reactor as reported in HW-18700. Iodine is removed by

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heated silver nitrate–coated 1/4" Berl saddle ceramic packing. At an inlet gas temperature of 475°F, iodine removal efficiencies of the order of 99.9% or better are expected. A minimum service life of one to two years is expected for the removal unit. The unit is designed for easy replacement.

Operation of a trickle type continuous dissolver for "J" slugs[1] showed that the effluent solution contained a very fine suspension of undissolved metal particles. It was observed that this suspension would completely dissolve to give a clear solution if kept in contact with the hot dissolver effluent for a few minutes. Holdup volume and trays and screens to retain the larger particles are provided in the dissolver to complete the dissolution of the fines.

Preliminary studies of the dilute TBP Flowsheet reported in ORNL 53-7-183 showed that the addition of 0.02M H_2SO_4 to the feed and scrub solution increased the previously limiting zirconium decontamination by a factor of >57 over-runs made without sulfate ion. Concentrated sulfuric acid is added continuously to the feed and is "reacted" for one hour at room temperature before extraction. An alternate suggestion would be to add concentrated sulfuric acid directly to the dissolver feed solution. Complete investigation to define the best conditions for treatment with sulfate ion or other additives is required.

2. Extraction and Stripping

The extraction and stripping flowsheet was developed by A. T. Gresky and is reported in "Tentative Chemical Flowsheet for the

1 Private communication from E. O. Nurmi and D. L. Foster, Unit Operations Section, Chemical Technology Division.

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Decontamination and Recovery of U^{235} from Irradiated Aluminum Alloys Using a 1.5% TBP System," ORNL 53-7-183.

High decontamination factors were obtained for zirconium, rare earths, ruthenium and niobium with an overall uranium loss of ~0.01%. The use of Amsco 125-82 diluent, stabilized with respect to HNO_3, permitted much higher decontamination factors than are usually observed in the TBP system, particularly with respect to the colloidal fission products. Amsco 125-82 has the approximate composition in volume %:

Paraffins and napthenes	99.4%
Aromatics	0
Olefines	0.6%

The olefines and other constituents which are susceptible to oxidation or nitration may be removed by reactions with $CrOCl_2$ (chromyl chloride). About 60% of the material which is reactive to $CrOCl_2$ can be removed by sulfonation with concentrated H_2SO_4.

Table 3 summarizes the laboratory flowsheet development.

Uranium losses were about 0.015% for extraction and 0.001% for stripping. The decontamination factors through extraction were as follows: Gross β 8.0×10^5; T.R.E. $\beta > 6 \times 10^6$; Ru β 8×10^4; Zr $\beta > 1 \times 10^5$; Nb $\beta > 3 \times 10^5$.

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Table 3

SUMMARY OF LABORATORY RESULTS FOR THE ENRICHED URANIUM 1.5% TBP FLOWSHEET

	FEED	SCRUB	SOLVENT	SOLVENT EXTRACT	AQUEOUS RAFFINATE	STRIP	AQUEOUS PRODUCT	SOLVENT WASTE
Volume Ratio	1.0	0.5	1.5(2)	1.5	1.5	0.25(3)	0.25	1.5
$Al(NO_3)_3$ M	2.0	1.0	- -	- -	1.67	-	-	- -
H_2SO_4, M	0.02(1)	0.02(1)	- -	- -	0.02	-	-	- -
HNO_3, M	-0.2 to 1.0	-	- -	0.008	0 to 0.67	0.01	0.06	< 0.002
U, gms/ℓ	4.0	-	- -	2.67	$<10^{-4}$	-	16.0	$< 10^{-4}$
Gross β, cts/m/ml	1.2×10^8	-	- -	100	7.3×10^7	-	~ 370(4)	80

NOTES:

(1) The addition of 0.02M H_2SO_4 to the aqueous feed and scrub effected a Zr D.F. increase by a factor of > 57 over other runs which had no sulfate ion.

(2) The volume ratio of solvent is adjusted to maintain about 40% saturation of the TBP with respect to uranium. Solvent is preconditioned Amsco 125-82 diluent with 1.5 vol. percent Tributyl Phosphate.

(3) The extremely high U distribution coefficient in the stripping system in favor of the aqueous (A/O = 200) indicates uranium could be recovered as a 0.5 to 1.0M solution by using a strip volume ratio of 0.03 to 0.015. Such a system might serve as a substitute for the proposed ion exchange concentration and isolation facility. An additional D.F. of 20 to 40 would result from such a scheme giving overall D.F.'s of about 5×10^7 through extraction and stripping. Because of the extremely large stage height for such an operation, development of special contactor units would be required.

(4) Activity of product uranium was 30 cts/m/mg of U (i.e., ~30% of natural background).

(5) Four extraction stages, 6 scrub stages and 3 stripping stages were used. Two additional extraction stages would permit wider variations (of the order of ±10%) in salting strength and uranium concentration without affecting overall efficiency.

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Stripping increased the gross β D.F. to 1.3×10^6 and the Ru D.F. to 1.3×10^5. By concentrating in the stripping column to 1.0M U, an overall gross β D.F. of 5×10^7 should be realized in one cycle. This method of concentration is preferred over ion exchange as the decontamination factors are comparable and the equipment for concentrating via stripping is less complex. Demonstration of decontamination at full process activity level would be required to prove definitely the process as a one-cycle operation. This is particularly true with respect to iodine which has been one of the D.F. limiting fission products in TBP systems.

3. Solvent Recovery

Previous processes using TBP have used the high aromatic content diluent, Amsco 123-15 and an alkaline wash procedure for recovery. High residual iodine, zirconium, ruthenium, and niobium activity levels remained in the solvent due to irreversible reactions with aromatic, nitro-aromatic, or other organic compounds in the diluent.

Laboratory work has indicated low aromatic content Amsco 125-82, pretreated before use to remove unsaturated compounds, gives much lower residual activity levels in the solvent. Evidence of this can be seen in the results of the laboratory testing of the 1.5% TBP flowsheet. Further work has been done to develop a more efficient method of solvent recovery. The favored process is the $Ca(OH)_2$ Recovery System reported in ORNL 53-6-110 by P. A. Haas, as it has lower waste volumes and better decontamination potential than the carbonate wash system.

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The solvent is contacted with $Ca(OH)_2$ slurry in two mixer-settler type contactor units with a one-hour contact time in each unit with an average retention time of 1-1/2 hours per unit. The solvent is then centrifuged to remove any particulate matter and fed back to the process. Decontamination tests on long-stored Purex solvent reported in ORNL 53-6-110 did not give as good results as laboratory tests on fresh solvent due to irreversible reactions with fission products during the storage period. Excellent removal of emulsion-forming compounds was noted.

It is believed that the combination of pretreated Amsco 125-82 diluent with an immediate treatment of the used solvent with $Ca(OH)_2$ slurry will prevent buildup of fission products in the solvent to the levels observed in Purex processing. It is possible that additional development may also indicate that the solvent recovery operation would not have to be a hot operation as presently contemplated.

Solvent will be reclaimed by water washing and then contacting with $Ca(OH)_2$ slurry in two series mixer-settler units with a contact time of one hour per mixer. The solvent will not be centrifuged as the small amount of solids carryover should not affect the process. Overall recovery of 98 to 99% of the solvent should be realized. Slurry is drained from each mixer once a day.

4. Second Extraction Cycle

If the decontamination factors predicted on the basis of the

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laboratory work are realized, a second extraction cycle should not be necessary. If the predicted decontamination factors are not realized in one cycle, it is possible that a second solvent extraction cycle would not be very effective as the limiting fission products could carry through the second cycle also. In that case, resin and silica gel columns would be favored over a second extraction cycle, although the procedure for regenerating resin and gel columns appears for uranium purification to be laborious and time-consuming at present. To permit comparison with existing recovery processes, allowance has been made in the cost study for a second extraction cycle which is 1/5 the volume scale of the first cycle and has corresponding columns, tanks, pumps, and solvent treatment units. The second extraction cycle is unshielded.

5. Chemical Treatment of Wastes

No specific treatment of the plant wastes, other than sampling for U loss determination, is planned. Evaporation will be done in the storage pits using fission product heating. Solvent recovery system wastes will be sent to the same tank system.

Raffinate batches or solvent recovery wastes containing recoverable amounts of uranium may be recycled to the process as first extraction cycle feed.

B. Waste Disposal

1. The General Problem

The permanent storage of radioactive liquid process waste to date

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has been a very costly operation and constitutes a significant fraction of the cost of processing spent reactor fuel. The present method of storing these wastes, in underground steel and concrete tanks, is considered a semi-permanent system and although already costly, the eventual storage of these liquid wastes, say for 100 years, may be even more costly.

A review of present liquid waste storage costs follows:

Site	Materials and Type of Construction	Capacity Per Tank	Approximate Cost[1] Per Gallon
Idaho Chemical Processing Plant	Stainless steel tank[2] enclosed in concrete tank	330,000 gals	$1.40
Hanford	Concrete tank[3] with[3] mild steel liner	750,000 gals	.45
Hanford (Proposed Purex facilities)	Jacketed mild steel tank (presumably within concrete tank)[4]	750,000 gals	.60 (estimate)

It is conceivable, however, that these costs do represent the cost of permanently storing these wastes if the following were done. As proposed at Hanford[5], allow the liquid wastes to boil[6] slowly to dryness and permit the last increment of water to escape the tank just before the rate of heat evolution falls to the rate of heat absorption by the ground.

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1 These costs are given in detail in Appendix VII.

2 Based on unreported ICPP costs.

3 HW-22888, Purex Process Evaluation, R. E. Tomlinson (12/10/51).

4 HW-25273, Storage of Radioactive Aqueous Wastes from Purex Processes, A. H. Platt and J. T. Krieg (8/1/52).

5 HW-25274, Preliminary Specifications for Storage Facilities for Purex, Salt Wastes, F. W. Woodfield and R. E. Tomlinson.

6 This assumes the availability of sufficient fission product heat.

[redacted]

This tank then, presumably almost free of corrosion, is essentially a permanent sealed container of non-flowing solid fission products.[1]

Although these costs may represent permanent storage costs they are still prohibitively high and could conceivably make waste storage the controlling cost item. It was, therefore, necessary to investigate modified versions of the present metal and concrete systems of storage and also to propose unique methods that could conceivably work. One proposal stands out among the rest and entails a new concept in waste storage methods.

2. A New Concept of Waste Storage

a. The General Design

A system for permanent storage of high activity liquid wastes must be capable of

1) isolating the wastes

2) concentrating and drying to solids the wastes

3) removing the fission product heat evolved in the waste

4) biologically shielding the wastes.

A unique, low cost system fulfilling these requirements is proposed as follows:

A large open pit is dug and completely lined with a ductile,

1 If radioactive material must be permanently isolated from animal and vegetable life, it appears essential that the material must ultimately be stored as an insoluble, unleachable solid or as a solid isolated from solvents and leaching substances, inasmuch as the lifetime of containers made of metal, plastic, asphalt, etc., is at present an unknown factor.

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impermeable material, such as tar, an asphalt, or a plastic. The pit is then filled with sand and/or crushed stone. Prior to complete filling of the pit with sand, a piping system is placed at some predetermined level below the top of the bed with which the wastes can be controllably distributed within the bed. The pit and sand bed are sized so that when filled to the design capacity with wastes[1] there remains above the liquid level of the wastes a height of packed bed which is adequate both for shielding requirements and for prevention of entrainment of particulates in the vapors from the hot wastes. This pit is shown schematically on the following page, Figure 4.

The operation of this system is as follows:

Liquid wastes, capable of providing sufficient heat to self-concentrate to dryness[2], are delivered to the pit via the piping system. The wastes in time boil very gently[3], the vapors rising through the voids of the packed section above the liquid level. Rain or artificial spray of water on the

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1 The wastes occupy the void space in the bed.

2 Based on calculations of the activity of the wastes of this study; low activity wastes would necessitate some artificial means for concentrating and drying the wastes.

3 Gentle boiling is due to the fact that the pit is so designed as to provide a very large horizontal surface area, and thus extremely slow boilup rate.

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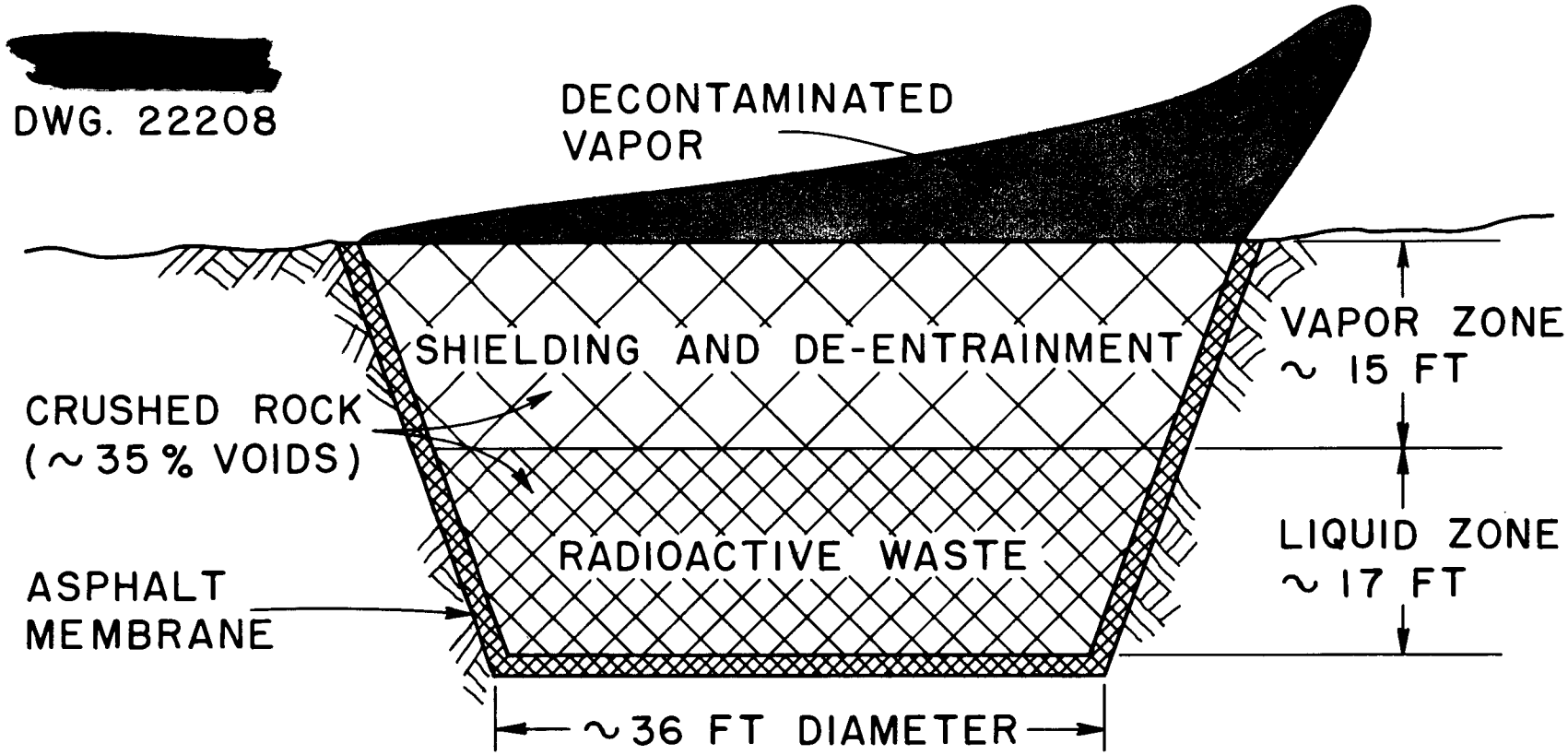
FIGURE 4

HOPE PROJECT WASTE STORAGE PIT

ESTIMATED WASTE STORAGE COST = $ 0.14 / GALLON



CAPACITY : $\sim 5 \times 10^6$ GALLONS

MAXIMUM HEAT LOAD : 1×10^7 BTU/HR

BOILUP RATE : 0.1 LBS/HR/FT^2

VAPOR VELOCITY IN VOIDS : 0.002 FT/SEC

[redacted]

surface of the sand bed will both replace the evaporated water and also trickle down through the deentrainment section of the bed, acting as a scrubbing solution. After the addition of wastes is discontinued, the rate of heat evolution will subside. The wastes are then allowed slowly to concentrate, by decreasing the amount of reflux water[1], with the last increment of water being allowed to boil away when the heat losses to the ground equal the heat evolved. The top of the bed is then permanently sealed with asphalt and presumably the original impermeable membrane will remain intact sufficiently long to prevent permanently washout of gross quantities of activity by subsurface water. Temperature probes and liquid level instrumentation would be included in the design to facilitate the operation of this system.

b. The Specific Design

A preliminary design and cost estimate for a system storing 5×10^6 gallons of liquid wastes indicates an approximate cost per gallon stored of $0.14. The bases for the design of this system are as follows:

Capacity required - - - - - - - - - - - - - - - - - 5×10^6 gallons

Maximum calculated heat load - - - - - - - - - - - 10×10^6 Btu/hr

1 This can be done by either reducing the amount of artificial spray or, in the case of rain, by partially or totally covering the pit with asphalt or a supported tarpaulin covering.

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Decontamination factor, liquid to vapor, required - - - - 10^{12}

Assumed maximum boilup rate required to get desired - - - 0.1 lb/hr-ft^2
decontamination factor

Assumed height of bed above liquid level required - - - - 15 ft
for shielding and deentrainment

Void space in sand and/or crushed stone - - - - - - - - - - 35%

The sand pit dimensions that resulted are: diameter, 360 ft; depth of liquid containing bed, 17 ft; total bed height, 32 ft. The boilup rate of 0.1 lb/hr-ft^2 results in a superficial vapor velocity through the deentraining section of 0.0008 ft/sec.

c. Validation of Assumptions

A number of assumptions, inherent or explicit, have been made which may raise considerable doubt as to the feasibility of this system. The following discussion is presented to somewhat validate these assumptions.

Assumption 1 - An impermeable, acid and radiation-resistant, high-temperature, pliable, inexpensive material is required; this is the greatest unknown of this design. The best material to date appears to be asphalt or an asphalt base material. Plastics, rubbers, and soil cements, or any combination of these, are other possibilities. Multiple membranes with layers of gypsum[1], soil cement or aggregate between them may

1 Gypsum is mentioned inasmuch as the sulfate of Sr is quite insoluble in water; this might be one means for long-term isolation of this long-lived hazardous fission product.

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may provide some insurance against outleakage and against radiation damage to the outer membranes. The radiation damage, acid resistance, and temperature characteristics of asphalt are presently being investigated at ORNL.

Assumption 2 - A decontamination factor 10^{12} from liquid to vapor can be obtained at a boilup rate of 0.1 lb/hr-ft^2 and a superficial vapor velocity of .0008 ft/sec through the deentrainment section. The basis for this assumption is the result of a D.F. vs. boilup rate study[1] performed on a large-scale evaporator. These data indicate a D.F., pot to condensate, of over 10^7 at a boilup rate of 40 lbs/hr-ft^2 and a vapor velocity of 3.0 ft/sec. This boilup rate is 400 times and the vapor velocity 4000 times the assumed rates of this study and could conceivably account for the difference in D.F. of 10^5.

Assumption 3 - No fission products volatilize from the boiling wastes. Iodine appears to be the only fission product that might volatilize. To avoid this hazard, a storage tank is provided to temporarily hold up for 60 days the hot wastes so that the iodine can decay in a closed, cooled system.

1 ORNL 51-11-113, Evaluation of Full-Scale Savannah River Project Evaporator, W. B. Watkins.

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Assumption 4 - A deluge of rain can be tolerated. Since a 15 ft deentrainment section is provided, a ten-inch rise in the liquid level in the pit would be of little concern. For this specific design, evaporation more than keeps pace with the average annual rainfall for the U.S., 36 inches, as long as fresh wastes are being introduced to the pit. In areas where the rainfall is excessive or when the energy production of the fission products has subsided, it will be necessary either to partially or totally cover the sand bed surface with an asphalt layer or supported tarpaulin in such a way as to permit the remainder of the water in the wastes to escape as vapor from the surface of the bed.

The above proposal, a sand-filled pit, appears to be a most promising low cost method of storing wastes. Experiments are presently underway at ORNL to determine the feasibility of the design.

3. Other Investigations

Four other investigations were made, three of which were based on modified versions of the metal-concrete tank system and the fourth based on a modified version of the pliable, impermeable membrane system. These systems were not subjected to severe scrutiny since preliminary cost and feasibility studies indicate these systems to be very unattractive when compared with the sand pit proposal. They are presented, though, for comparison with present methods and costs of storage.

[redacted]

[redacted]

Design Proposal

a. (See Figure 5) A stainless steel jacketed tank placed in an open pit and shielded with a concrete shadow shield. Cooling by evaporation-condensation-reflux of boiling wastes. The feasibility of this system is excellent, but the cost $0.76/gallon is prohibitively high. Corrosion may also be a serious problem.

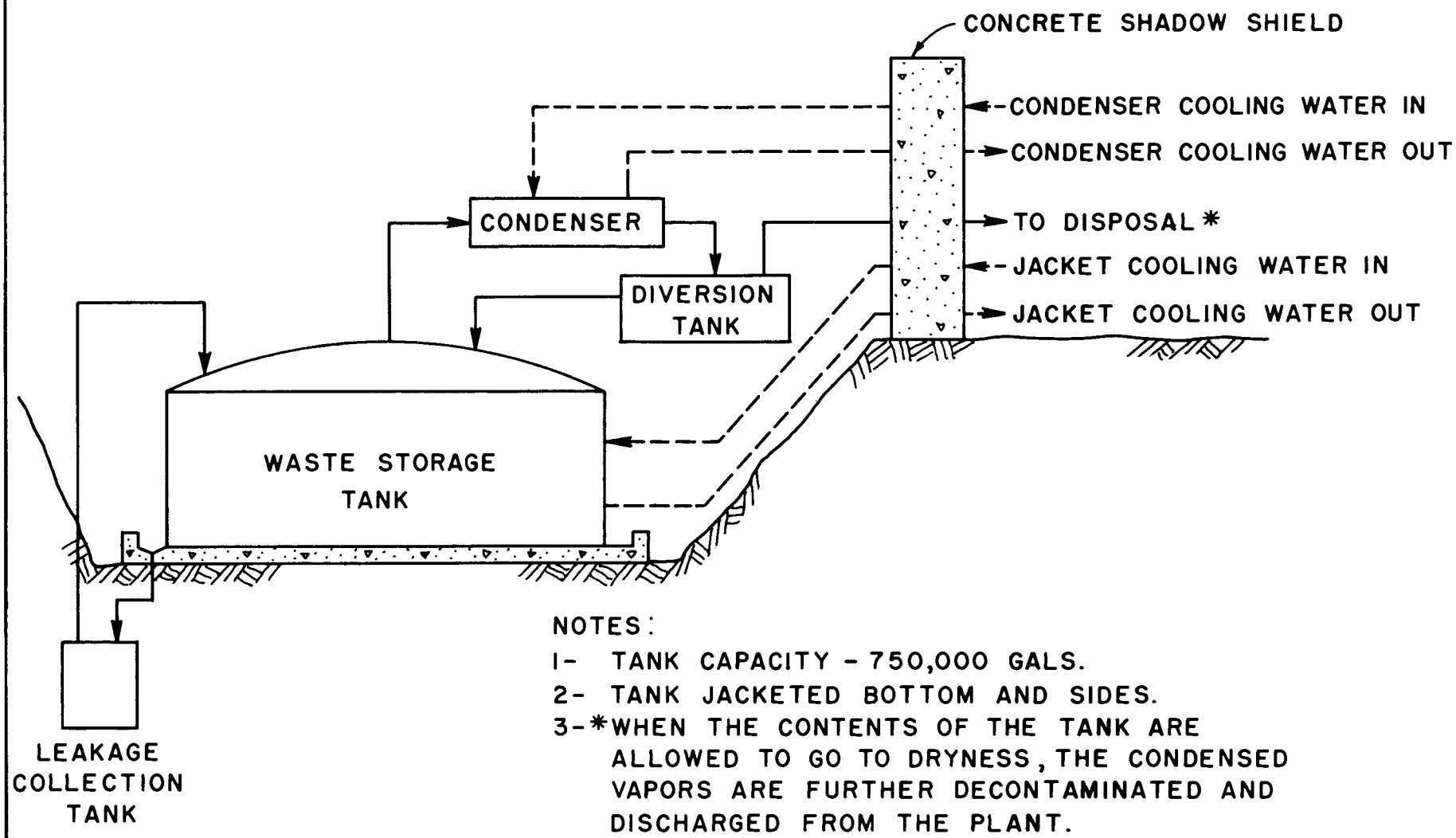
b. (See Figure 5) As above, only tank is mild steel for alkaline waste service. Feasibility is good but dependent on corrosion of tank. Cost is high, $0.54/gallon.

c. (See Figure 6) A concrete tank placed in an open pit and shielded with a concrete shadow shield. Cooling by evaporation-condensation-reflux of boiling wastes. The feasibility is good[1] if the tank is lined with acid-resistant brick, the concrete is of the very dense non-porous type, and sufficient high-temperature steel is incorporated into the structure. For acidic wastes, this method appears to be less expensive than the stainless steel tank system. The cost is $0.57/gallon.

d. (See Figure 7) An asphalt-lined canal with mild steel cover, shadow shielded. Cooling by submersed stainless steel cooling coils. The feasibility of this system is dependent on finding an asphalt or other material which is ductile, acid and radiation-resistant, and can withstand high temperature (250°F). A dis-

1 Based on the experience of Mr. A. C. Martinsen of the Catalytic Construction Company.

[redacted]



NOTES:

1- TANK CAPACITY - 750,000 GALS.

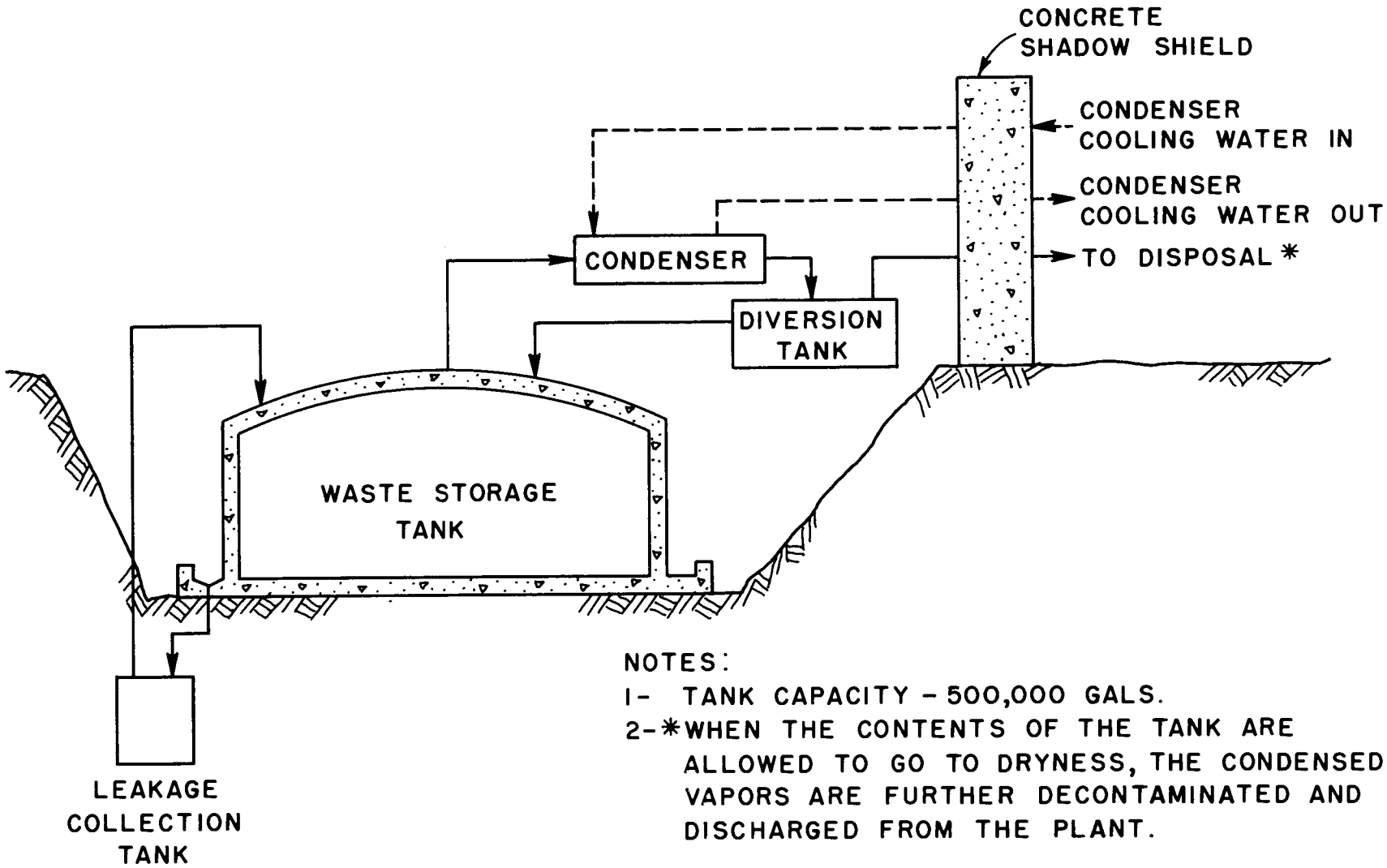
2- TANK JACKETED BOTTOM AND SIDES.

3- *WHEN THE CONTENTS OF THE TANK ARE ALLOWED TO GO TO DRYNESS, THE CONDENSED VAPORS ARE FURTHER DECONTAMINATED AND DISCHARGED FROM THE PLANT.

FIGURE 5

STAINLESS STEEL OR MILD STEEL TANK WASTE STORAGE SYSTEM

DWG. 22603



NOTES:

1- TANK CAPACITY - 500,000 GALS.

2- * WHEN THE CONTENTS OF THE TANK ARE ALLOWED TO GO TO DRYNESS, THE CONDENSED VAPORS ARE FURTHER DECONTAMINATED AND DISCHARGED FROM THE PLANT.

FIGURE 6

CONCRETE TANK WASTE STORAGE SYSTEM

DWG. 22604

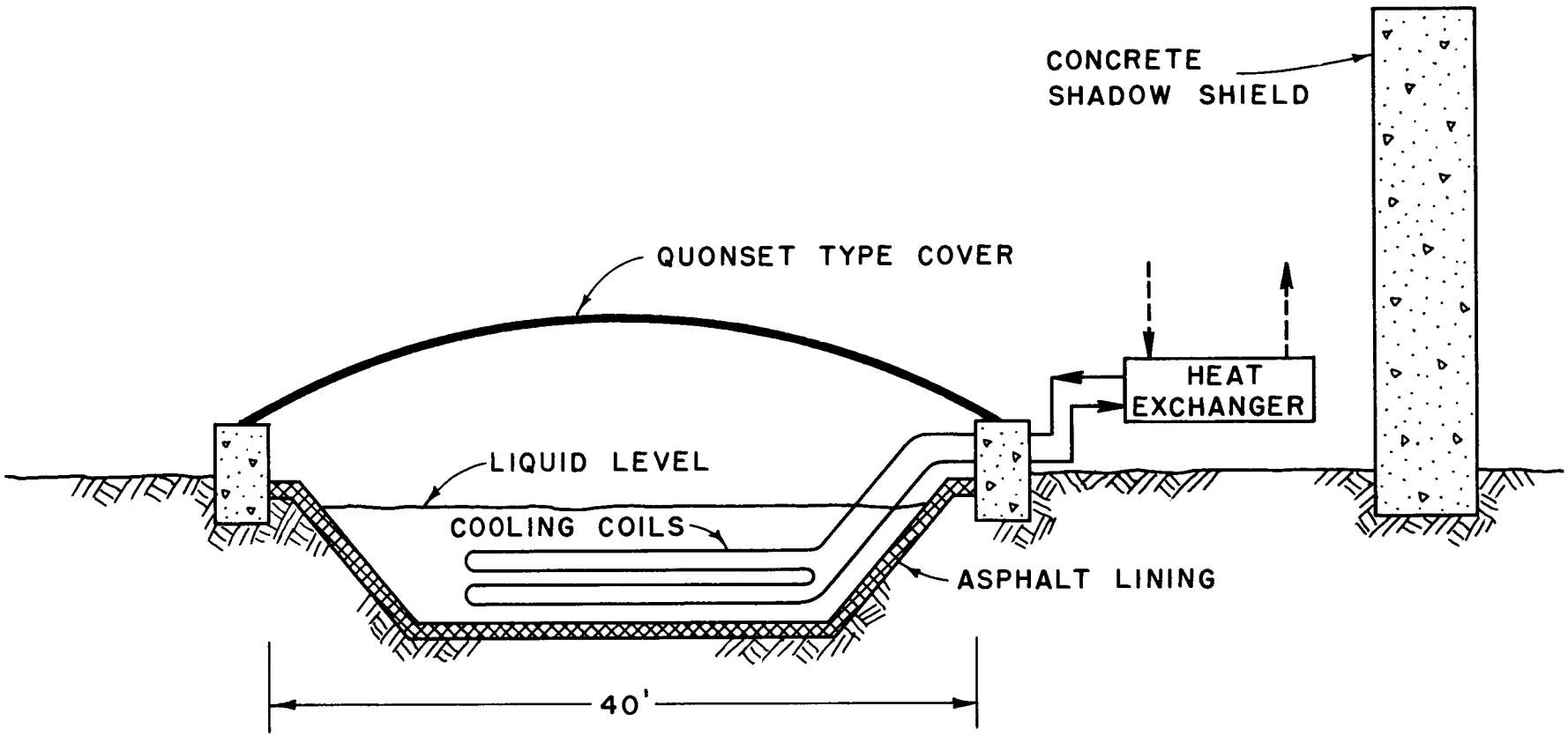


FIGURE 7

ASPHALT LINED CANAL WASTE STORAGE SYSTEM

DWG. 22605

advantage of the system as illustrated is the inability of the wastes to self-concentrate to dryness by evaporation. To accomplish this a more costly vapor-tight cover would be required, possibly made of stainless steel, and condenser and ion exchange equipment for removal and discard of the vapor would have to be included. This would increase the cost considerably, $0.29/gallon, but would still render this the second least costly system of all that were investiagated.

The following table summarizes the costs of the various systems investigated and compares with the cost of the tank type system the cost of their present counterpart.

System	Estimated Cost $/gallon	Present Cost $/gallon
Stainless steel (acid)	0.76 [1]	$1.40
Mild steel (alkaline)	0.54 [1]	.60
Concrete (acid)	0.57	
Asphalt lined canal (acid)	0.29	
Asphalt membrane sand pit	0.14	

1 The differences between Estimated Cost and Present Cost of the first two systems are principally due to elimination of the concrete tank in which the metal tanks sit, by placing the metal tanks above ground. It should be pointed out that the volume of neutralized aluminum bearing waste is approximately four times greater than the acid volumes.

[redacted]

V. Description of Recommended Plant

Detailed, final design of the process equipment has not been done because of the limited scope of the project. In general, studies of the equipment have been carried to such a point that rough dimensions, layouts, and promising schemes of operation could be selected for cost evaluation. The following description will indicate the unique details of the major equipment items.

A. Fuel Element Storage[1]

The fuel elements are stored in the cooling basin, which is a continuation of the reactor service canal, after the end fittings have been removed at the reactor. The elements are held vertically in a "honeycomb" grid structure consisting of 22"-long sections of 5-inch steel pipe welded together side by side to provide individual storage pockets for a total of 1200 fuel elements. Fuel elements are loaded into and removed from the storage grid by long-handled tongs through the 10-foot deep water shield. The storage grid is subdivided into small units, each holding 80 fuel elements, that can be moved by a monorail hoist to the reactor portion of the canal for loading, to the storage area for cooling, and to the fuel element charging machine area for unloading. The illustration of the process building does not show all details of the fuel charging machine, but the following description will clarify its function.

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1 For design details of the storage facility the reader is referred to Appendix VIII.

[redacted]

[redacted]

B. Fuel Element Charging Machine[1]

Fuel elements are loaded into 40 individual pockets on the periphery of a rotating, vertical axis 4-foot diameter cylinder. The pocketed cylinder serves as a fuel element reservoir so only occasional operator attention is required to reload the drum. The cylinder is rotated intermittently on the desired time cycle to load the fuel elements one by one into the opening in the pusher mechanism that elevates them from the canal through the cell wall shield concrete into the dissolver. The pusher mechanism consists of a straight, inclined 3-1/4-inch square section stainless steel duct that contains the fuel element, a motor-driven long shaft screw mounted on the underside of the duct, and a free-running nut on the screw with a finger that projects into the duct through a slot and pushes the element up the duct as the screw is turned. The motor drive for the screw is reversible so the pusher finger is returned to the charging opening. When the fuel element reaches the upper travel limit of the pusher, it enters an enlarged section of the duct arranged so the fuel element is turned at an angle of approximately 90° and falls under the force of gravity into the dissolver. The elevations of the canal surface and the dissolver are adjusted so the upper portion of the pusher duct is above the canal water surface, thereby maintaining an air-tight dissolver. Oxygen, required for nitric oxide recovery, is introduced into the duct near the water level and serves as a purge gas to prevent canal water contamination by radioactive spray from the dissolver. A light-weight hinged

1 For design details of the charging facility the reader is referred to Appendix VIII.

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[redacted]

flap door, similar to a swing check valve that would pass a fuel element and then close, is provided in the slug chute near the dissolver to further block dissolver spray.

The fuel element charging machine is provided with indicating and remote control devices so all phases of the operation can be supervised and controlled from the operating gallery.

C. **Dissolver**

The dissolver is a vertical, 12-inch inside diameter, 18-foot tall heat-treated type 309 stainless steel vessel. The fuel elements enter the dissolver 12 feet above the bottom and are supported in the solution by a grating 2.5 feet above the bottom of the dissolver. A two-inch diameter core pipe is mounted on the centerline of the dissolver in the fuel element space to prevent stacking of more than six fuel elements side by side and to furnish some slight neutron poisoning. A 7-foot long jacket is provided for heating or cooling the reaction zone. The 2.5-foot high section below the support grating serves as a digestor with 20 minutes holdup time to permit dissolution of the fine metal particles in the effluent solution. It is fitted with six baffles or trays to prevent undissolved particles falling directly into the bottom outlet. Silicon from the brazing alloy is carried out of the dissolver as a dilute slurry.

Immediately above the fuel element inlet is located a 12-inch diameter, 2-foot long precondenser section with approximately 30 square feet of transfer area. This removes approximately 80% of the water vapor so

[redacted]

[redacted]

a reaction holdup time of approximately 15 seconds is provided for nitric-oxide oxidation in the 2-foot high baffled space above the precondenser. Reflux and makeup water is continuously sprayed into the oxidation chamber from the reflux condenser sump jet and flows down through the precondenser into the dissolver. Vapor velocities are 3.0 feet per second above the liquid surface and 0.13 feet per second in the oxidation chamber above the precondenser.

D. **Reflux Condenser**

The reflux condenser is a vertical, falling film condenser-absorption tower that is 10 inches inside diameter, 17 feet 8 inches tall, with 14-foot long tubes and has 211 square feet of transfer area. Makeup water enters the top of the condenser as a spray with the incoming vapor and is collected with the condensate and recovered nitric acid in a sump in the bottom of the condenser shell. The reflux liquid is continuously jetted to the dissolver. The reflux condenser is mounted beside the dissolver to minimize the heat room requirements.

E. **Silver Reactor**

The silver reactor consists of a vertical 2-inch pipe preheater section, 8 feet long, to preheat approximately 1.7 feet^3/minute of gas from 70°F to 475°F and a 4-inch iodine removal tower packed with an 8-foot depth of 1/4-inch ceramic Berl saddles, coated with silver nitrate to remove the radioiodine. The preheater is heated electrically with tubular "Calrod" heaters clamped onto the exterior of the preheater

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pipe. The preheater and packed tower are mounted side by side in a liquid-tight, 7-inch diameter by 9-foot high enclosure filled with granular insulation to minimize heat losses and damage of the silver nitrate coating by local condensation of moisture.

F. Feed Cooler - Zirconium Digestor

The upper portion of this vessel consists of vertical 5-inch diameter by 2 feet 6-inch tall feed cooler with 4.1 square feet of transfer area to cool the nearly boiling feed solution to 68°F. Cooled feed passes through a mixing nozzle where it is mixed with concentrated sulfuric acid to produce a solution 0.02M in sulfuric acid. The mixed feed enters the 12-inch diameter by 6 foot 6-inch high digestor section that is provided with half-moon baffles to prevent channeling to the bottom outlet. Feed is held up for one hour in the digestor.

G. Hot Feed Pumps

The hot feed pumps are remote head, Lapp Insulator Company Pulsafeeder Pumps, size CPS-3, with double ball stellite suction and discharge check valves and remote auto-pneumatic stroke adjustment. All other hot pumps are similar. A large number of pumps are required because of the limited head room for an underwater maintained plant.

H. Extraction Column

The extraction column is a 5-inch diameter pulse column with a combined extraction and scrubbing height of 24 feet fitted with 23% free

[redacted]

area pulse plates on 2-inch centers. The column has a 12-inch diameter by 3-foot long top section and an 18-inch diameter by 3-foot long bottom section to give a holdup time of 30 minutes for the respective phases for disengagement. The assumed stage height is 2 feet. Six extraction and six scrubbing stages are provided. The column will operate at 590 gal/hr/sq ft in the extraction section and 300 gal/hr/sq ft in the scrubbing section, and is concatenated into four sections to reduce the over-all height. The column interface is controlled by regulating the flow of aqueous phase from the column with a control valve.

I. Stripping Column

Due to the extremely high organic-to-aqueous flow ratio required to produce a concentrated uranium product, the stage height becomes excessive with a conventional pulse column. Limited development work at ORNL (per private communication from A. C. Jealous) has indicated that this drawback can be overcome by modifying concatenated pulse columns so diffusion of the uranium up the column is minimized. As this is not a fully developed item, the stripping column is shown as a six-section concatenated pulse column for cost estimating purposes.

J. Piping and Valves

All piping, in sizes one inch and smaller, will be stainless steel tubing with Parker Appliance Company Triple-Lok flared tube fittings for connectors.

All piping in sizes greater than one inch will be stainless steel

[redacted]

pipe with ring joint flanges. The flanges will be connected to the pipe with flexible metal bellows and will be gasketed with inert, soft metal (such as gold) ring gaskets to permit easy assembly and disassembly of the piping under water.

Pipe nozzles on vessels and connections on lines will be arranged for maximum accessibility for underwater maintenance.

Process valves will be air-actuated, bellows-sealed, bellows-operated valves with stellite trim.

K. Samplers

Sampling is accomplished with the conventional air jet type of recirculating unit. The sample unit incorporates the recently developed improvements (i.e., hypodermic needle sample tips, piping changes to eliminate liquid passage through the sample jet, and chain conveyor bottle handling), being considered for the ORNL Thorex Pilot Plant. Details of the design of this sampler are given in Appendix IX.

L. Instruments

Conventional instrumentation is used throughout the plant. Liquid level and density are determined by the usual "bubble tube" method. As this plant will be a fairly small, continuous flow plant, it is desirable from the standpoint of minimizing operating personnel to design for completely automatic, coupled operation of the fuel element charging, dissolving, feed preparation, extraction, and solvent recovery

[redacted]

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portions of the plant. Allowance has been made in the cost estimate for such a scheme.

[redacted]

[redacted]

VI. Details of Cost Estimate

Cost estimates were obtained by: (1) direct estimation of required material and labor, (2) application of a factor to some basic cost, and (3) application of a unit cost to a calculated number of units. To illustrate, the first method was used to obtain basic equipment costs[1]; the second method was used in determining piping costs; and the third method was employed in obtaining building costs.

Sources of and justification for the factors and unit costs are embodied in the detailed estimate. Estimates are based on an Engineering News Record construction cost index of 600.

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1 Wherever possible in estimating equipment costs the Idaho Chemical Processing Plant equipment costs were used.

[redacted]

[redacted]

Annual Cost Summary
for a One-Extraction Cycle Process

Cost	$	$ per Gram Uranium
Fixed Investment[1]	417,900	.208
Operating Cost	1,274,600	.635
Working Capital[2]	2,200	.001
SF Inventory[3]	167,000	.083
Total	1,861,700	0.927

1 Fixed investment charged at 16%; F. I. = 0.16 x $2,616,200

2 Working capital charged at 2%; working capital = .02 x $127,700

3 Inventory charged at 4.5%; SF inventory = 0.045 x $3,700,000

[redacted]

[redacted]

Fixed Investment Summary
for a One-Extraction Cycle Process

Manufacturing Facilities	$1,329,900
Site Development	99,800
Subtotal	$1,429,700
Engineering and design, 15% subtotal	214,500
Construction (supervision, insurance, taxes, overtime, etc.), 20% subtotal	285,900
Contractor's fee or profit, 2% subtotal	28,600
Contingency, 25% subtotal	357,500
Preoperation and startup	300,000
Total	$2,616,200

[redacted]

[redacted]

Working Capital for a One-Extraction Cycle Process

Material in process (plant fillup of HNO_3, Amsco and TBP)	$ 200
HNO_3 - negligible	
Amsco - 3100 lbs x $.05/lb = $160	
TBP - 66 lbs x $.64/lb = 40	
Cash and miscellaneous items	
(30 days operating cost), $1,274,600/12	106,300
Total	$106,500

SF Inventory

Storage for 35 days required.

$24/g x 4400 g/day x 35 days	$3,700,000

[redacted]

[redacted]

Manufacturing Facilities Summary for a One-Extraction Cycle Process

Processing basin	$ 269,000
Fuel storage canal	92,000
Operating-laboratory-office building	267,000
Warehouse	11,300
Process equipment, piping, and instrumentation	400,600
Special equipment	205,000
Vessel and cell off-gas system and stack	85,000
Total	$1,329,900

Site Preparation

7.5% of manufacturing facilities cost [1]	$99,800

1 Based on ICPP costs.

[redacted]

[redacted]

Operating Costs for a One-Extraction Cycle Process

Raw materials	$ 143,400
Labor, 64 men x $6500/man-year	416,000
Overhead, 75% of labor	312,000
Operating supplies, 10% of labor[1]	42,000
Electricity, 100 kw[2] x $\frac{8,760 \text{ hours}}{\text{year}}$ x $\frac{\$.005}{\text{kwh}}$[1]	4,400
Steam, 10,000 lbs/day[2] x $\frac{365 \text{ days}}{\text{year}}$ x $\frac{\$0.75}{1000 \text{ lbs}}$[1]	2,700
Water, 2,000 gal/day[2] x $\frac{365 \text{ days}}{\text{year}}$ x $\frac{\$0.20}{1000 \text{ gal}}$[3]	200
Compressed air, 25 CFM[2] x 525,600 $\frac{\text{min}}{\text{year}}$ x $\frac{\$0.10}{1000 \text{ ft}^3}$[3]	1,300
Maintenance material, 10% of fixed investment[1]	261,600
Analytical lab material, $\frac{\$.70}{\text{analysis}}$ x $\frac{24,000 \text{ anal.}}{\text{year}}$[4]	17,000
Waste storage, 1,450 gal/day x $\frac{365 \text{ days}}{\text{year}}$ x $\frac{\$0.14}{\text{gal}}$[5]	74,000
Total	$1,274,600

1 C. H. Chilton, "Cost Estimating Simplified," Chem. Eng., 58, p. 108 (June 1951)

2 The service requirements are conservative estimates. The water requirement represents only process water; cooling water is circulated in a closed system. Steam is principally for building heat; little process heating is done.

3 E. C. Dybdal, "Engineering and Economic Evaluation of Projects," Chem. Eng. Prog., 46, p. 57 (1950)

4 Based on ORNL pilot plant laboratory costs.

5 Based on cost for storage in sand-filled pit (see page 71).

[redacted]

[redacted]

Raw Materials
for a One-Extraction Cycle Process

	lb/yr (100%)	$/lb (100%)	$/yr
60% HNO_3	1,060,000	.05	53,000
ANN	430,300	.15	64,600
$Hg(NO_3)_2$ 1/2 H_2O	4,120	4.30	17,700
Amsco	49,600	.05	2,500
TBP	940	.64	640
O_2	28,300	.16	4,600
H_2SO_4	6,850	.024	160
$Ca(OH)_2$	9,880	.02	200
Total			143,400

Basis: 5% loss per day of Amsco and TBP based on throughput,

95% recovery of NO and NO_2 from dissolver off-gas.

[redacted]

[redacted]

Manufacturing Facilities, Details for a One-Extraction Cycle Process

1. Processing basin

Concrete; 975 yd^3 at $150/$yd^3$ in place [1]	$146,000
Stainless steel linings; (floors and walls); 4370 ft^2 at $13/$ft^2$ [2]	57,000
Excavation and backfill; 5400 yd^3 at $3/yd3	16,000
Miscellaneous; (painting, lighting, cat walks, handling equipment)	50,000
Total	$269,000

2. Fuel storage canal [3]

Canal structure; (concrete, steel, earthwork, etc.)	47,000
Equipment; (crane, platform, charger, element racks, etc.)	45,000
Total	$ 92,000

3. Operating-laboratory-office building

Office area; 800 ft^2 at $30/$ft^2$ (furnished) [4]	$ 24,000
Laboratory area; 1700 ft^2 at $60/$ft^2$ (furnished) [4]	102,000
Operating area; 4700 ft^2 at $30/$ft^2$ [4]	141,000
Total	$267,000

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1 High cost of concrete used to compensate for more costly rolling roof slabs and piping through concrete.

2 Based on Idaho Chemical Processing Plant costs.

3 A detailed estimate was made but not included here for the sake of brevity.

4 P. Jandrisevits, "Preliminary Estimating by Selective Unit Costs," Ind. Eng. Chem., 43;2299 (1950). (Note: These unit costs include electric power panels, main service piping headers, heat, ventilation, compressed air facilities, and lockers).

[redacted]

[redacted]

4. Warehouse

Dimensions: 25' x 50' x 15'h

Volume: 18,750 ft^3 at \$.60/ft^3 \$11,300

5. Process equipment, piping, and instrumentation

Major equipment in basin:

Item	Description	Delivered Cost
dissolver	12" Φ x 18'; jacketed; pre-condenser and digestor sections	\$ 6,000
reflux condenser	210 ft^2 heat transfer area	8,000
iodine reactor	3" Φ x 9' pipe packed with $AgNO_3$-coated Berl saddles and 2" Φ x 8' pipe preheater mounted in 7" Φ x 9' sheath, insulated with SiO_2	1,600
cooler-digestor	cooler (4 ft^2 heat transfer area mounted on 12" Φ x 8' baffled digestor)	1,800
surge-rework tank	600 gal., jacketed	4,000
condenser	30 ft^2 heat transfer area	2,000
knockout drum		400
2 raffinate tanks	600 gal. each, jacketed	8,000
cooler	3/4" Φ x 5' pipe jacketed	200
extraction column	5" Φ, 12 ft, extract., 12 ft scrub	6,000
inter-column surge tank	125 gal.	1,200
stripping column	4" Φ, 30 ft	5,000
solvent surge tank	125 gal	1,200
3 mixer settler contactors		9,000
solvent waste tank	125 gal.	1,200
2 solvent hold tanks	220 gal, each	3,400
Subtotal		\$59,000

[redacted]

[redacted]

Major equipment outside basin:

Item	Description	Delivered Cost
scrub makeup tank	1200 gal., agitated	$ 6,000
scrub feed tank	600 gal.	3,000
strip makeup tank	25 gal., agitated	550
strip feed tank	25 gal.	400
slurry makeup tank	40 gal., agitated	800
slurry feed tank	40 gal., agitated	800
diluent makeup tank	75 gal., agitated	1,000
catalyst makeup tank	70 gal., agitated	1,000
catalyst feed tank	70 gal.	800
HNO_3 storage tank	1200 gal.	20,000
air heat exchanger		4,200
surge tank, cooling water		1,000
8 pulsafeeders		36,000
10 pumps (6 Milton Roy, 4 centrifugals)		7,500
2 pulse generators		10,000
Subtotal		$ 93,100
Delivered equipment total		$152,100
Installation, 0.14 x D.E.[1]		21,300
Installed equipment total		$173,400

1 The value 0.14 is the installation factor based on ICPP costs.

[redacted]

[redacted]

Piping, fittings, and valves, 1.0 x I.E.[1]	$173,400
10 samplers at $1500[2] each	15,000
Instrumentation, $16,740 x 1.387 x 1.67[3]	38,800
Total, equipment, piping, and instrumentation	$400,600

6. Special equipment and facilities

Storage and dilution cell	$100,000
Laboratory equipment (mass spectrometer, radiation instruments, miscellaneous shielding, water distillation unit, etc.)	75,000
Shop equipment	25,000
Handling equipment, (fork lift truck, etc.)	5,000
Total	$205,000

---

1. The piping factor 1.0 assumed is considerably higher than the value 0.6 of Langs for fluid processing plants; it is considerably less than the probable factor of 1.5 for the ICPP. The factor was chosen as a median since a remotely operated plant requires piping through concrete, parallel piping, remotely operated valves, etc., which make for piping costs higher than normally encountered in fluid processing plants; the factor was chosen less than the ICPP factor inasmuch as less parallel piping through concrete is required and also since the size of equipment is larger than that of the ICPP.

2. These samplers are identical to those in the Thorex Pilot Plant.

3. The value 1.387 is a piping and panel factor and 1.67 is an installation labor factor. Both factors are based on ICPP costs.

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Additional Costs for a Two-Extraction Cycle Process

(2nd Cycle cold operation)

Manufacturing facilities: (for 2nd extraction cycle and 2nd solvent recovery system)

1. Building: Approximately 250 ft^2 of additional area in operating building at $30/$ft^2$ — $ 7,500

2. Equipment, piping, and instrumentation

Item	Description	Cost
feed surge tank	5" Φ x 6' tall	$ 400
extraction column	1-1/2" Φ, 12 ft extract. 12 ft scrub	4,500
inter-column surge tank	15 gal.	150
stripping column	1" Φ x 30 ft	3,700
solvent surge tank	15 gal.	150
3 mixer-settler contactors		4,200
2 solvent hold tanks	50 gal. each	1,400
scrub head tank	125 gal.	1,200
strip feed tank	25 gal.	400
butt makeup tank	600 gal.	3,000
butt feed tank	300 gal.	2,100
8 pulsafeeders		24,000
pumps (3 Milton Roy, 1 centrifugal)		2,700
2 pulse generators		6,000
Delivered equipment total		$ 53,900

[redacted]

[redacted]

Item	Cost	
Installation, 0.14 x D.E.	$ 7,600	
Installed equipment total	$ 61,500	
Piping, fittings, and valves, 1.0 x I.E.	$ 61,500	
Instrumentation, $6,750 x 1.387 x 1.67	$ 15,700	
Total, equipment, piping, and instrumentation	$138,700	$138,700
Total, manufacturing facilities		$146,200
A. Additional fixed investment		
Manufacturing facilities, subtotal		$146,200
Engineering and design, 15% subtotal		21,900
Construction, 20% subtotal		29,300
Contractor's fee or profit, 2% subtotal		2,900
Contingency, 25% subtotal		36,600
Total		$236,900
B. Additional operating cost		
Raw materials - 43,640 lb ANN		$ 6,600
Other operating expenses change negligibly		
Total		$ 6,600

[redacted]

[redacted]

Annual Cost Summary
for a Two-Extraction Cycle Process

Cost	$	$ per gram uranium
Fixed investment[1]	$ 456,500	.227
Operating cost[2]	1,304,900	.650
Working capital[3]	2,200	.001
SF inventory	167,000	.083
Total	$1,930,600	$0.961

1 0.16 x ($2,616,200 + 236,900) = $456,500

2 $1,274,600 + 30,300 = $1,304,900

3 Change is negligible

[redacted]

[redacted]

VII. Recommendations for Development Programs

Parts of the following recommendations have their origin in observations which were made in our Project Hope work; some have an earlier origin. We shall distinguish between two broad classes of desirable development work: (1) those developments which must be completed before the present Project Hope design can really be labeled as feasible; and (2) specific developments which are not necessary for Project Hope but which would be very desirable in the design of later chemical purification plants.

A. Developments for Project Hope

Altogether there seem to be only three major pieces of development work which must be carried out before Project Hope can be considered as a completely rationalized and workable design of a chemical plant. So that there can be no doubt in the reader's mind as to what is necessary, let us consider these one at a time.

1. Maintenance Principles

We believe that the underwater maintenance principle is a completely workable one and we are convinced that it is cheaper than any maintenance principle presently in use. It is necessary, however, to demonstrate that underwater maintenance is possible under all of the conditions which one is likely to encounter in routine and extraordinary operation of the plant. While this can only come from experience, there are two factors which lead us to believe that even now we could operate such a plant successfully.

In the first place, the MTR experience has shown us that it is simpler than we had expected to work through as much as eighteen feet of water with long handled tools. In the second place, in order to obtain some

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feeling for the problems to be overcome we developed some very rudimentary maintenance tools in connection with our Project Hope work. The details of these maintenance experiments are given in Appendix V, but it is clear that the problems to be overcome are not difficult.

2. Solvent Treatment

It is wholly desirable to make the solvent recovery a cold process. We do not know as yet whether or not this is possible but experiments to determine the feasibility of cold solvent recovery have been initiated. The results of these are at present very encouraging. They are reported in detail in Appendix IV.

3. Properties of Tars and Asphalts

In order to make the waste disposal system operate, it is necessary to have a better understanding of the properties of tars and asphalts in the presence of moderately high radiation fields. We have found that there is very little experience with comparable pits even in the storage of nonradioactive materials, so in order to obtain information we have had to initiate test programs in cooperation with industry. A report of the status of these programs is given in Appendix XII.

It appears altogether likely that we can find a satisfactory material in the asphalt-tar group which will permit us to build a low-cost waste storage facility along the general lines sketched in the report.

B. Development of Equipment

It is the job of the design engineer to plan on the basis of existing equipment. Usually he has neither the time nor the desire to see new equipment invented or developed if he can carry out his job with existing

[redacted]

means and methods. However, the development of new equipment which may facilitate later designs is one of the important functions of major laboratories and, with this in mind, we mention a few types of equipment which may be useful for the design of later purification plants. None of these are necessary for the design described in the following chapters, although a few might be usefully applied.

1. Artificial Fission Products

We mean by this a mixture of chemicals of the same composition as fission products with only enough activity to permit their tracing and identification. Although in the past it has been the practice to use radioisotopes in testing chemical plants before beginning high level hot runs, to our knowledge, it has not been common practice to attempt to actually mock-up the products which will be presented at low level, paying particular regard to the chemical form of each of the constituent fission products. It seems reasonable to expect that such a development would be very useful in predicting the high level performance of a given installation.

2. Criticality Detector

This has been mentioned in more detail in Section III. Its purpose is to detect the accumulation of large amounts of fissionable material which may have plated out in vessels, etc. The plant design described in the following sections does not necessitate the use of such a detector. It may be, however, a very useful all-around instrument.

We envisage that it would consist of a slow neutron source; the neutrons from which would induce fission in a vessel containing sizeable amounts of

[redacted]

fissionable material. The fast neutrons from the fission would be detected with a suitable second instrument, such as a U^{238} fission chamber. The principal difficulty in developing the instrument is the low intensity of the secondary neutrons and the high gamma background.

According to Wattenberg's figures, a 30 curie Sb-Be source, with a Be layer about 5 cm thick, emits 10^9 neutrons per second. It should be possible to direct these neutrons to a certain extent and to have about 1% of them enter the tank in question. If there is real danger of criticality, the number of fast neutrons emitted will be at least close to the number of slow neutrons entering the tank; i.e., about 10^7 per second. If a fast neutron crosses a fission chamber with 10 foils, the sensitive depth will be 20 times the average distance from which the fission products enter the gas. This distance is $1/2 \times 3 \times 10^{-4}$ cm, so that the sensitive depth will be 3×10^{-3} cm. Since the fast fission cross section is about .7b, the efficiency of the fast neutron counter will be of the order

$$\frac{18.6 \times 3 \times 10^{-3} \times .6 \times .7}{235} = 10^{-4}.$$

Hence, in order to obtain 1 ct/sec, the fission chamber must subtend at least an angle of 10^{-3} with the tank. This appears difficult, particularly if it should prove so difficult to "bias" against gamma rays that one would have to shield the detector with a lead or bismuth shield. It is not necessarily impossible.

3. Soft Gaskets

The preceding remarks emphasize the importance of soft gaskets; i.e., gaskets which deform under the pressure applied and assume the form of

[redacted]

the hard surface against which they are pressed. The calculation in Section III C shows that it is in the realm of possibility to find a sufficiently radiation-resistant plastic for this purpose. It is to be remembered, though, that the beta radiation which was omitted in the aforementioned calculation may aggravate the situation. This will depend on the access of the radioactive liquid to the gasket which will depend on the seal design.

The extreme in soft gaskets is the liquid one. We were disappointed to find that no liquid has a surface tension which would be orders of magnitude higher than that of water. If the liquid is not chemically resistant to the process liquid it may be possible to separate it by a gas layer.

4. Air Valve

The simplest safe valve still appears, at least to some of us, to be an air valve. Such a valve is possible only if the pipe can have an inverted U. If the gas pressure is increased by means of the valve indicated, which is in the cold area, the flow of liquid is blocked from one stem of the U to the other. The upward tube could be of very small diameter and the valve on it a simple greased stopcock. Both could be replaced if they should gradually become radioactive. This is not expected to happen, however, in foreseeable time. Such valves have been used many times in industry but not in radioactive chemical processing.

5. Rotor in, Stator out

This is a stirrer which does not introduce gas bubbles and thus avoids the entrainment problem. It is similar to the canned rotor pump

[redacted]

which is presently in use. It avoids the use of a lubricating gland through which the stem of the stirrer would enter the tank. It consists of a stator outside the tank and a "squirrel cage" rotor inside. A propeller is attached to the latter to stir the liquid and, possibly, to lift the rotor from its seat.

No calculations were made to gauge the effectiveness of this equipment which surely could be developed in some form.

6. Instruments in General

The instruments used in purification plants, and also those contemplated in the present study, are standard instruments designed and fabricated for controlling chemical processes which do not involve radioactive substances or inaccessible areas. (The sampler is the single obvious exception hereto.) It seems a priori unlikely that the same instruments are best suited also for the control of processes with high radioactivity. This does not mean, of course, that they cannot be used.

The ordinary level and density recorder is an example of the above point. The instrument used for this purpose bubbles gas through the liquid; the pressure necessary to maintain a low but steady gas flow is a measure of the weight of the liquid column of unit cross section above the point where the gas is introduced. The idea of the equipment is ingenious and is well developed. Its only obvious disadvantage, from the point of view of handling radioactive substances, is the need for a constant gas stream with the unavoidable entrainment of activity.

One might conceive instead of a density and level recorder based on the principle of the float. Many such recorders have been developed and

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are in use; however, they are not used in radioactive chemical processing as we think they should be in the interest of economy. One possible variety consists of an elastic strip of metal fastened at one end to the tank wall and containing on the other end a piece of soft iron and an empty sealed container. The hydrostatic force acting on the container is proportional to the density of the liquid which surrounds the container and so will be the position of the soft iron above the point where it stands in the tank when it is empty. For a level recorder the empty container has the shape of a vertical pipe; the force acting upon it is proportional to the density of the liquid and the height of the submerged part. The advantage of such an arrangement is that the position of the soft iron can be gauged from outside the tank by measuring the self-induction of a coil nearby if the material of the tank is non-magnetic. Ordinary stainless steel is, of course, non-magnetic.

We hold no brief that the most satisfactory level and density recorder would be based on the foregoing principle though we are sure that the principle could be implemented and a fast-acting, reliable, accurate, and relatively inexpensive recorder developed. We do feel certain, however, that the field of instrument development for the specific purposes of purification plants will prove to be a rewarding one. It would be surprising if the radioactivity of the process solutions and the properties of the fissionable substances could not be used to a greater extent for process control than is done at present. A beginning along this line may have already been made at the MTR where the content of fissionable materials in fuel elements has been measured by the fissionability property itself. We are informed that the accuracy of this method is higher than of the usual chemical methods.

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7. Design Principles

There are, in addition, two or three principles which appear to have final utilization if they can be applied to radioactive processing. The first of these will be to take advantage of gravity flow in order to eliminate difficulties with pumps. This could be done, for example, by carrying out the dissolution operation in a cell high above ground and by having each succeeding cell slightly below the level of its predecessor. The principle of the air valve which was described above could be used successfully in such an arrangement. The main disadvantage of the gravity flow plant is the requirement for operating the cells to permit flooding them individually. Another design principle which ultimately could become very important has to do with concentrating the wastes. If the asphalt-tar retention basis should prove feasible, as we think it will, it may already constitute a step towards the more economical solution of the waste problem. However, we have not exhausted this problem either in the direction of disposal or utilization. Although the utilization idea has already received a great deal of attention without much success in trying to use the fission product as a prominent source of concentrated heat, we believe that the problem should remain on the active list because one can never feel completely satisfied with any other solution of the waste problem.

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Appendix I

The Design of the Building

It was clearly impossible in the short time available for us to engineer a design for the building which would house the Hope plant. Nevertheless, in order to make our cost estimates as realistic as possible we felt that it was essential that the conceptual design of the building be carried sufficiently far so that we could root out the important operational pitfalls which are inevitably present in every new scheme. Accordingly such a design was made. The tentative floor plan of the building and an artists' drawing of the entire Hope plant are appended so that the reader can ascertain for himself the validity of the detailed building cost estimates.

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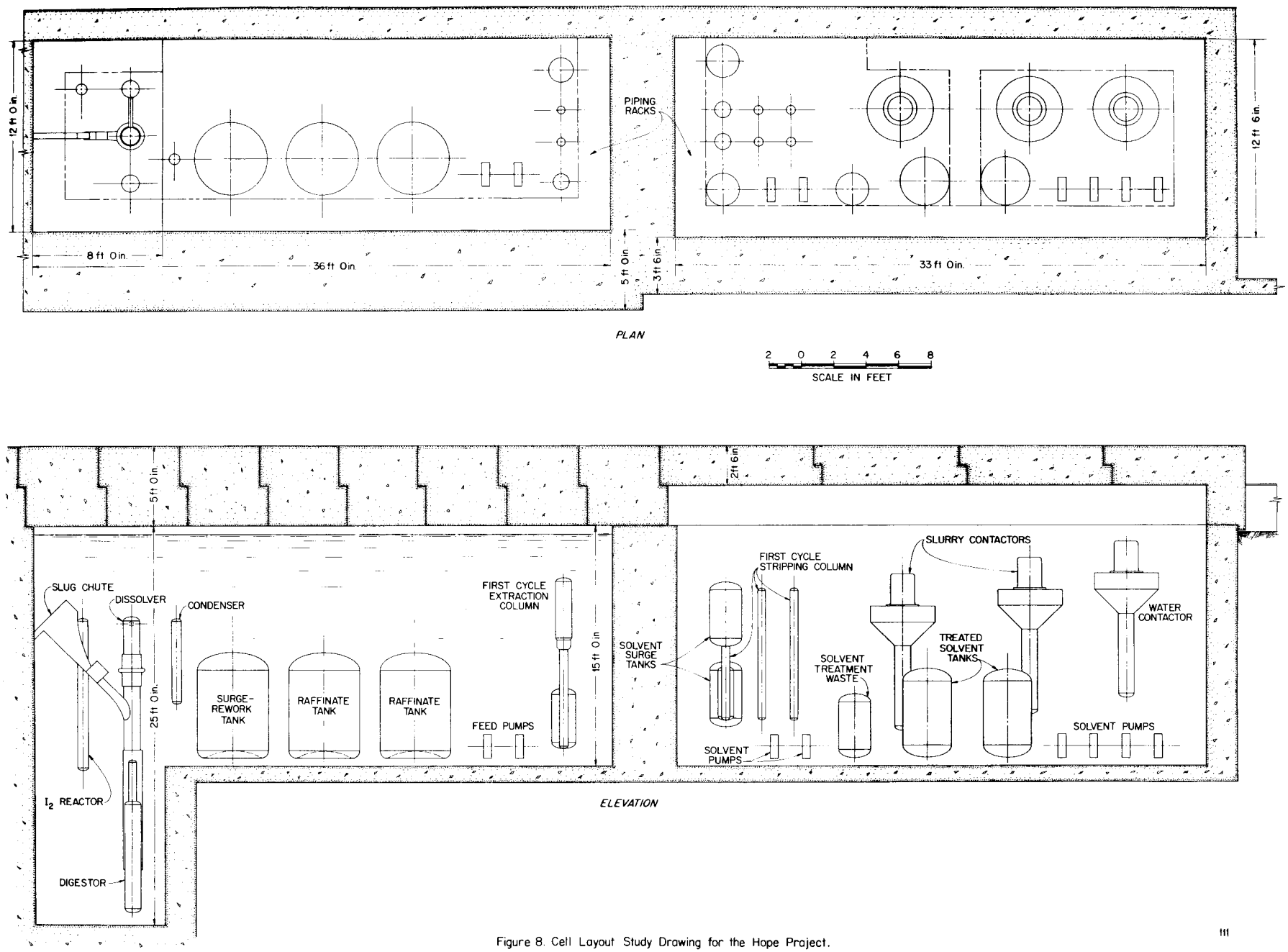

Figure 8. Cell Layout Study Drawing for the Hope Project.

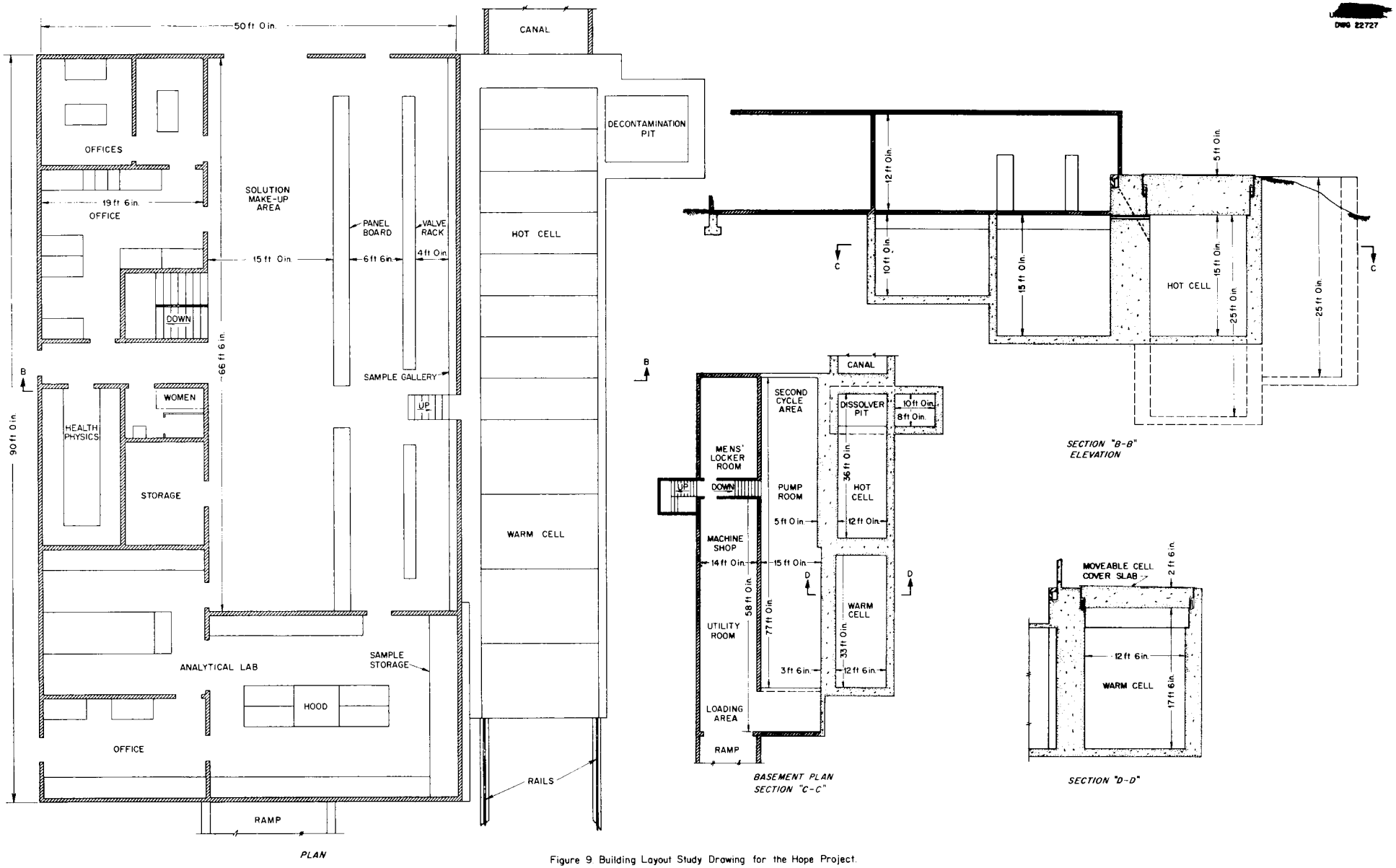

Figure 9. Building Layout Study Drawing for the Hope Project.

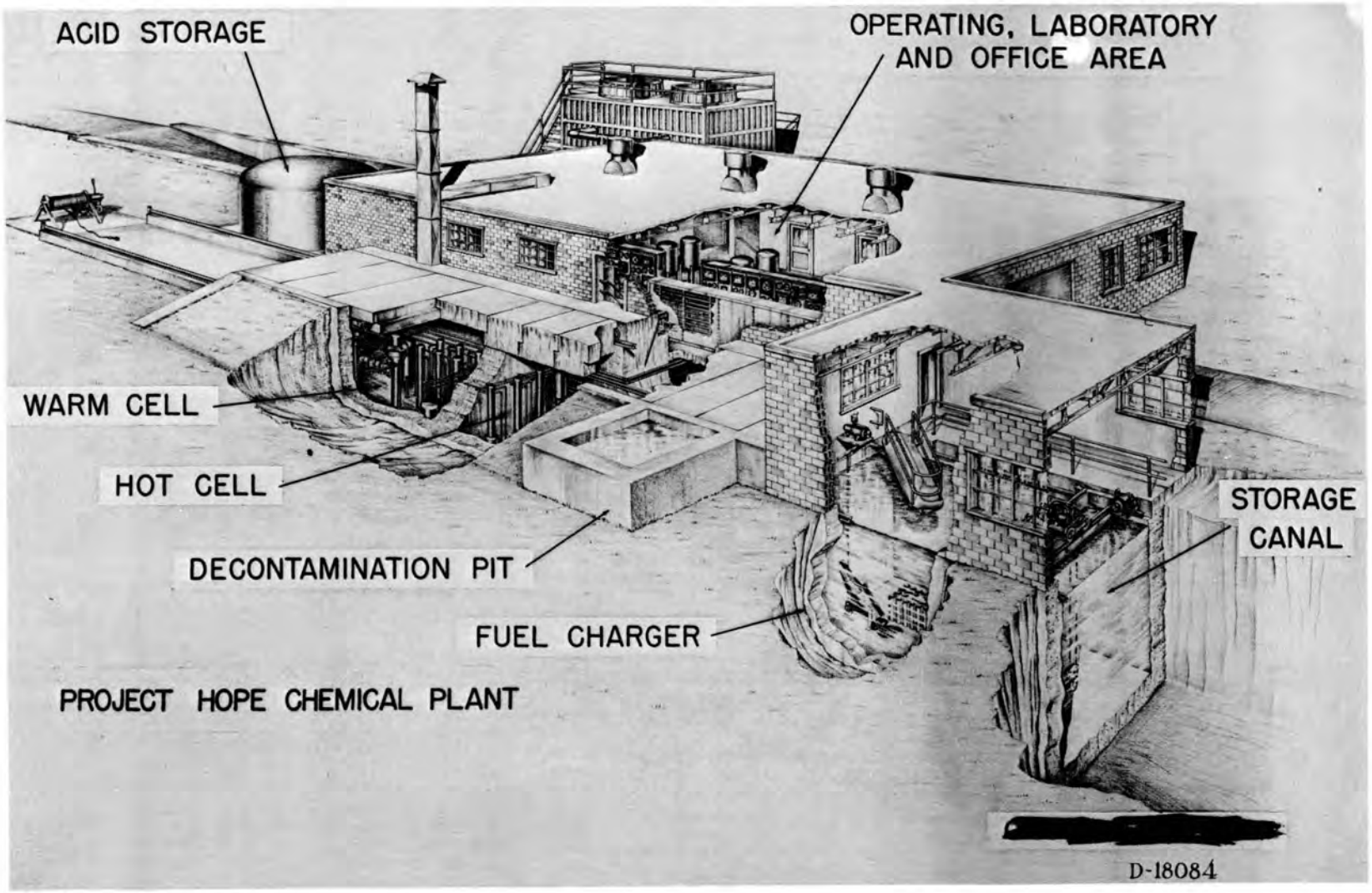
ACID STORAGE
OPERATING, LABORATORY AND OFFICE AREA
WARM CELL
HOT CELL
DECONTAMINATION PIT
FUEL CHARGER
STORAGE CANAL
PROJECT HOPE CHEMICAL PLANT
[redacted]
D-18084

FIGURE 10

[redacted]

Appendix II

Alkaline Chemistry

An alternate to the acid dissolution of the MTR type fuel elements makes use of caustic for this step. This procedure is, of course, possible here because of the amphoteric character of aluminum. The advantages of the method are primarily related to the initial removal of the bulk of the aluminum from the uranium prior to the extraction and the usual economic advantage of storing alkaline aluminum wastes compared to storing acid aluminum wastes. The disadvantages of the alkaline dissolution procedure stem largely from difficulties of filtration or centrifugation of the uranium particles left from the aluminum dissolution, enhanced by the problems of fission heat in a small bulk of precipitate.

A caustic dissolution procedure would consist of dissolving the Al-U fuel element in about 5M NaOH such that an initial Al : NaOH mole ratio of 1 : 1.5 is established. The solution is further adjusted to .02f $Ba(NO_3)_2$ which serves to precipitate the carbonate in the alkali. The residue containing the uranium and most of the fission products is either filtered or centrifuged from the $NaAlO_2$ solution. This residue is then dissolved in HNO_3 and introduced as the feed in the extraction cycle.

Since the Al-U atom ratio in the fuel element is several hundred, a dissolution procedure which removes the bulk of the aluminum prior to the extraction is inherently attractive. The extraction stage is thus simplified so as to relax the rigidity of the chemical control and pre-

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sents reasonable promise for adequate decontamination in a one-cycle solvent extraction. Because of the smaller volumes, the scale of the equipment and the related operating and maintenance problems are appreciably reduced.

Since $NaAlO_2$ solutions may be stored at about 5M concentration compared to about 2.5M $Al(NO_3)_3$ a two-fold saving in volume may be realized for the caustic waste. Perhaps even more advantageous cost-wise, should it prove desirable to store the waste solutions in steel containers, is that a heavier mild steel rather than the thinner stainless steel can be used for the tank construction. The use of the heavier mild steel tank would eliminate massive and costly concrete supporting structures for the stainless steel tanks.

In view of the hazardous character of the 25-year Sr^{90}, lowering the concentration of strontium in waste solutions appears very desirable. Precipitating an insoluble strontium salt or carrying strontium with another precipitate might offer a simple means of accomplishing this - possibly using multiple precipitations. It is, of course, necessary to precipitate the strontium but not the aluminum. Sulfate suggests itself for acid solutions; however, both in 2.5M and 0.5M $Al(NO_3)_3$ solutions the solubility of $SrSO_4$ is quite high and the carrying of Sr by $BaSO_4$ is quite poor. On the other hand, the solubility of $SrCO_3$ appears quite low in concentrated $NaAlO_2$ solutions, presenting a distinct possibility of achieving a low Sr

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concentration in the alkaline waste solution.

A potentiality of the alkaline system, as yet unexplored, is the possibility of completely discarding the aluminum waste following additional cleanup (i.e., decontamination). The waste material that would remain to be stored could be confined to smaller volumes, the magnitude of which would be governed largely by the fission product heat evolution. Should a market develop for either mixed or separated fission products, it would appear that chemical separations and packaging could be simplified by this prior removal of the inert bulk.

Perhaps the major obstacle to an alkaline system is the uniform antipathy to filtration and centrifugation procedures, although in this particular case the character of the precipitate is unusually favorable. The special problems peculiar to proposed equipment for such remote operation and the equipment maintenance suggest the desirability of avoiding filtration and centrifugation. Furthermore, the large quantity of fission heat, about 5 kw/day, associated with the precipitate presents as yet untested problems in its handling. A probable requirement is that the precipitate be left continually covered with liquids.

A filtration scheme in which the solution is passed upward through the filtering surface has been considered. The character of the precipitate is apparently such that a cake need not be built up to provide the filtering medium. A large fraction of the precipitate would settle away from the filtering surface, simplifying the problem of keeping the

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precipitate constantly covered with solution.

The dissolution of the precipitate following filtration might perhaps best be accomplished directly in the filter unit. The precipitate is readily soluble in 6M HNO_3 at 85°C. A useful scheme to accomplish this in minimal volumes might handle the dissolution somewhat as a Soxhlet extractor, boiling and recycling the HNO_3.

The caustic dissolution of aluminum has been studied in connection with the jacket removal of uranium slugs, the dissolution of J slugs (J slugs are a U-Al mixture containing 7-1/2% U) and for the MTR RaLa process (RaLa process refers to the production of Ba^{140} which is used as a source of La^{140}). In the latter case Blanco and co-workers (cf. ORNL-1148, "Laboratory Development of the MTR-RaLa Process for the Production of Ba^{140}") have done much to demonstrate the feasibility of the alkaline dissolution of the MTR fuel elements. Although the motivation for the RaLa process is the separation of Ba^{140}, the dissolution of the MTR element and the subsequent filtration, or centrifugation steps, are analogous to the present problem. For this reason this portion of the RaLa process is briefly outlined.

In the RaLa process the dissolution is carried out with a mixture of NaOH and $NaNO_3$. Blanco recommends mole ratios of 1:0.5:1 for Al:$NaNO_3$:NaOH such that the final aluminum concentration is 5M. The principal net reaction involves the oxidation of Al by NO_3^- to yield NH_3. In the

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absence of NO_3^- the reaction results in the evolution of H_2. The purpose of the NO_3^- is to supplant the hydrogen producing reaction which is a potential explosive hazard. The uranium is oxidized to yield the orange $Na_2U_2O_7$ (private communication from R. D. Ellison). The x-ray pattern confirmed this compound and showed the absence of UO_2, UO_3, or U_3O_8. The precipitate formed here is quite fine and is difficultly filterable. However, Blanco reports that with the use of filter aid on a 10 micron average porosity sintered steel filter adequate filtration was achieved with uranium losses averaging about 0.3% (cf. ORNL-1148). He further reports that 97.5% of the β^- activity remained with the precipitate and that less than 1% of Ba, Sr, Rare Earths, and I went into solution. Centrifugation was shown to be a satisfactory substitute for filtration yielding approximately the same uranium losses. The dissolution rate of the fuel element is quite rapid and some control must be exercised in the rate of addition of the solution. The $Na_2U_2O_7$ dissolved rapidly and completely in 6M HNO_3 where the HNO_3 : U ratio is initially 8 : 1. The $NaAlO_2$ solutions were stable with respect to the formation of precipitates over wide ranges of concentration of NaOH, $NaNO_3$, and $NaAlO_2$. An aluminum concentration of 5M is readily achieved.

Many conclusions drawn from the MTR-RaLa process are pertinent here. However, a significant improvement in the filterability of the solid matter is achieved by omitting the $NaNO_3$ in the dissolution and utilizing

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the reaction producing H_2. When a sample of J slug material is dissolved in 1-5M NaOH, the uranium is not attacked by the medium. A granular (gritty to the touch) gray black precipitate results which is readily filterable. The uranium in the residue appears to consist largely of the compounds UAl_3 and UAl_4. X-ray patterns served to identify these compounds (with UAl_4 in excess) as well as some $Al_2O_3 \cdot 3H_2O$ (private communication from R. D. Ellison). The patterns for $Na_2U_2O_7$, the uranium oxides, and uranium metal were absent. The Al/U mol ratio was found by chemical analysis to be about 7 and the ratio appeared to be reduced by alkaline washing. However only about 35% of the weight of the precipitate was accounted for by the uranium and aluminum. The uranium losses, using 5M NaOH at mole ratios of Al : NaOH of 1 : 1.5 and containing .02f $Ba(NO_3)_2$ averaged about .05%. The addition of $Ba(NO_3)_2$ serves to reduce the carbonate concentration of the alkali and has been observed to improve the filterability of $Na_2U_2O_7$. Although carbonate complexes UO_2^{++} strongly enhanced losses were not observed even in the presence of an added excess of 0.1f Na_2CO_3. However, because the problem had been incompletely explored, $Ba(NO_3)_2$ was used since it appeared that some advantage might accrue.

The rate of dissolution of the fuel element was appreciably slower in the straight caustic dissolution compared to the NaOH-$NaNO_3$ dissolution; however, quantitative data were not obtained. Since the surface-to-mass ratio of the MTR element is relatively large, this factor was not considered serious.

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The distribution of gross fission products at the tracer level was noted in an experiment two days after J-slug material had been briefly irradiated. As measured with a NaI scintillation counter, the precipitate contained 98% of the gamma activity. The distribution of ruthenium, strontium, and iodine between the precipitate and filtrate were followed. Both ruthenium and strontium were found principally with the precipitate. However, about 90% of the iodine appeared in the filtrate. This latter result is in variance with data reported by Blanco who found 1% of the iodine in the filtrate. Possibly his higher concentration of fission products may have precipitated insoluble iodates.

I. Design of a Filter

The success of the caustic dissolution process depends to a large degree on the design and development of a satisfactory filter or centrifuge, handling of H_2 gas, and determination of dissolving rates.

As mentioned previously development work has been done on a filter for separation of insoluble uranium from the slurry leaving the dissolver in the MTR-RaLa process (cf. ORNL-1148, "Laboratory Development of the MTR-RaLa Process for the Production of Ba^{140}"). However, since the precipitate in this case was sodium diuranate, which is more difficult to filter than the insoluble uranium residue in the straight caustic dissolution process, the latter process appears to be more favorable from a filtration standpoint.

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Recent laboratory work on the straight caustic dissolution process indicates that a porous stainless steel filter element with an average pore opening of five microns will efficiently separate the uranium residue from the slurry without the necessity of a cake build-up.

A filter unit design which should give satisfactory service is shown in Figure 11. The nine-inch, porous, stainless steel disc is available commercially. The tangental inlet will give desirable flow conditions during filtration and subsequent acid dissolution. The solution is passed upward through the filter to obtain a good filtration rate and decrease the possibility of pulling the residue dry. A baffle in the unit is provided to decrease entrainment in the offgas during acid dissolution. Parker fittings on the three nozzles of the unit would make replacement of the unit possible.

At the completion of the filtration the residue would be washed with dilute caustic. All the wash possible would be removed through the outlet line by vacuum. Nitric acid and enough live steam to bring the temperature to 85°C would be introduced. After the vigorous dissolution of the residue, the solution would be pulled off through the outlet line. The unit would then be filled with wash water, which would be pulled to a still, from which the condensate would be continuously returned to the filter until the concentration of uranium in the filter is negligible.

Porous tantalum is available commercially as an alternate in the event corrosion of the porous stainless steel is excessive.

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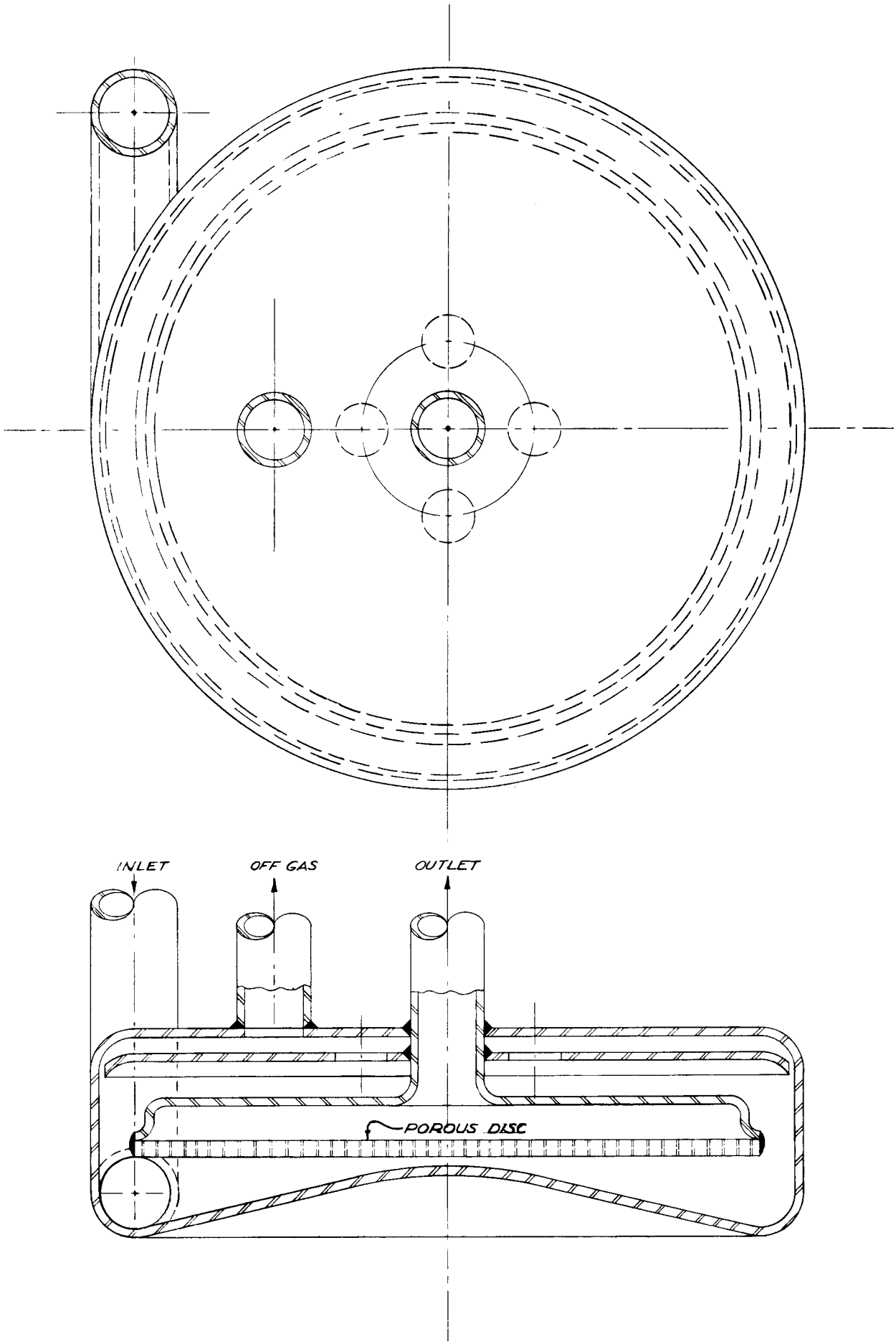

Figure 11. Filter.

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Appendix III

Fuel Element Storage - Shielding Calculations

We shall compute the maximum dose received through "d" centimenters of water:

1) Consider a cylindrical array of fuel elements 60 cm high and 235 cm in radius. This arrangement has the same volume as one month's supply (1200 elements) stored on 12 cm centers.

2) Assume we need to shield for the 2.5 Mev γ's whose concentration is determined by the La^{140} yield (.057 curies La^{140} curie of fission products).

3) The concentration of total fission products is .52 curies/cc.

4) Finally check that La is the troublemaker by computing the dose to be expected from 100% of the γ's all having the average energy (600 kev). Replace volume source by plane source of strength $N\lambda$ where

N = number of 2.5 Mev γ's/cc - sec and

λ = mean free path of 2.5 Mev γ's in H_2O.

$$N = .52\frac{\text{curies}}{\text{cc}} \times .057\frac{\text{cu La}}{\text{curie}} \times 3.7 \times 10^{10}\frac{\gamma\text{'s}}{\text{sec/curie}} \times \frac{1}{.042}\text{cm} = 2.6 \times 10^{10}\frac{\gamma\text{'s}}{\text{cm/sec}}$$

$$\Phi_P(d) = \frac{Nd\,\Sigma_\gamma}{2}\left\{e^{-\Sigma_\gamma d}\left[\frac{1}{(\Sigma_\gamma d)^2} - \frac{2}{(\Sigma_\gamma d)^3}\right] - e^{-\Sigma_\gamma (R^2+d^2)^{\frac{1}{2}}}\left[\frac{1}{\left[\Sigma_\gamma (R^2+d^2)^{\frac{1}{2}}\right]^2} - \frac{2}{\left[\Sigma_\gamma (R^2+d^2)^{\frac{1}{2}}\right]^3}\right]\right\}$$

For $.057\frac{\text{cu La}}{\text{cc}}$

$$\Phi_P(300\text{cm}) = 2.92 \times 10^3\ \gamma\text{'s/cm/sec}$$

$$\Phi_P(400\text{cm}) = 3.24 \times 10^1\ \gamma\text{'s/cm}^2\text{/sec}$$

[redacted]

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For 0.52 curies/cc of .600Mev γ's

$$\Phi_P(300\text{cm}) = 10^{-2}\ \gamma\text{'s/cm}^2\text{/sec}$$

$$\Phi_P(400\text{cm}) = 10^{-6}\ \gamma\text{'s/cm}^2\text{/sec}$$ so shielding against the La γ's is indeed the entire problem.

Now

$$\text{Dose}\ \frac{R}{\text{sec}} = \frac{\Phi(\text{cm}^{-2}\text{-sec}^{-1})\ E(\text{Mev})\ \tau(\text{cm}^{-1})}{6.79 \times 10^4\ \text{Mev cc}^{-1}\text{-R}^{-1}}$$

Therefore

$$D\ \frac{\text{mr}}{\text{wk}} = \frac{\Phi E \tau \times 10^3\text{mr-R}^{-1} \times 3.6 \times 10^3\text{sec-hr}^{-1} \times 40\text{hr-wk}^{-1}}{6.77 \times 10^4}$$

For $E = 2.5$, $\tau = 1.77 \times 10^{-5}$

$$D\ \frac{\text{mr}}{\text{wk}} = \frac{\Phi(2.5 \times 1.77 \times 10^{-5} \times 3.6 \times 10^6 \times 40)}{6.77 \times 10^4} = .094\Phi$$

Thus: D(300cm) = 275 mr/wk

D(400cm) = 3 mr/wk

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Appendix IV

I. Observations on Iodine Chemistry

The solvent extraction process for the recovery of enriched uranium poses a difficult problem of solvent recovery. As mentioned in Section IV, since a rather short cooling time (30 days) is proposed for the fuel elements before processing, high iodine, zirconium, ruthenium and niobium activities may be expected to accumulate in the solvent during extraction. Whether or not solvent recovery can be a cold operation will depend on attaining either low initial extraction of these contaminants or high decontamination factors in the proposed solvent recovery method [$Ca(OH)_2$ slurry treatment].

At present little appears to be known concerning the chemical states or species of these elements in aluminum nitrate solution and even less appears to be known about their chemical state in the organic solvent. It is felt that a study of the chemical state of these expected contaminants under the conditions of the proposed process is the first step in devising an efficient method for solvent recovery. Experiments have been carried out to eludicate the chemistry particularly of iodine.

A. Reaction of Iodine with Amsco and Paraffinic Hydrocarbon Solvents.

Iodine solutions (initially 2×10^{-4} M I_2) were prepared by dissolution of iodine in "Amsco" and "paraffinic" hydrocarbon solvents. "Amsco" refers to Amsco 123-15, a diluent of ca. 11% aromatic content. "Paraffinic" refers to Amsco 125-82, which contains less than 1% aromatic content. The reaction rates of iodine with these solvents were measured spectrophotometrically by

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following the decrease with time of the absorption band for iodine which, in these solvents, occurs in the region 400 to 600 mμ with an absorption maximum near 520 mμ. Except during the spectrophotometric measurements, the solutions were exposed to the fluorescent lighting of the room. All measurements were made at room temperature, ca. 25° C.

Iodine was found to react fairly rapidly with both solvents under exposure to fluorescent light and the I_2 concentration decreased approximately exponentially with time. For Amsco a "half time", $t_{1/2}$ = 10 hrs, was found for the reaction and for the paraffinic solvent, $t_{1/2}$ = 38 hrs.

Placing the solutions in the dark very markedly lowered the reaction rate in both solvents, indicating the iodination reaction is photochemical and thus probably also catalyzed by nuclear radiation.

Iodine fixed in the organic solvents principally as organic iodination products was difficult to extract. Poor extraction of the iodine (not more than 60% in any case) took place when separate portions of the organic solvents were shaken with equal volumes of aqueous solutions containing NaOH, Na_2SO_3-NaOH, Na_2SO_3-HNO_3, Hg(II)-HNO_3, and KI-NaOH. Iodine distributions were measured radiometrically using iodine-131 tracer in these experiments.

B. Effect of Hg(II) on the Extractability of Iodine by Hydrocarbon Solvents

In view of the high solubility of HgI_2 in organic solvents and the high stability constant for HgI_2 [1], considerable extraction of iodine as an Hg(II) iodide complex might be expected. The extent of such extraction has been

1 L. G. Sillen, Acta Chem. Scand. 3, 539-53 (1949).

[redacted]

[redacted]

measured with Amsco and "paraffinic" solvents for several conditions of Hg(II) and iodide concentration in aluminum nitrate solution. To facilitate analyses I^{131} and Hg^{203} tracers were used in the experiments.

Typical results are shown in Figure 12. In the absence of Hg(II) air oxidation of iodide occurs and large amounts of iodine extract into the organic phase presumably as I_2. With increasing amounts of Hg(II) decreasing extractability of iodide was found but the amount extracted was still appreciable. Experiments with Hg^{203} tracer indicated that the iodide which does extract is associated with the Hg(II).

Hg(II) iodide complexes were rapidly and fairly completely removed from Amsco solvent by extraction with equal volumes of NaOH or NaOH $(NH_4)_2S$ solution. Thus, it appears that iodide bound to Hg(II) as an iodide complex will be easily extracted from the organic phase. However, the possibility that such extractable Hg(II) iodide complexes may slowly react irreversibly with the solvent under irradiation and high temperatures should be borne in mind.

C. Volatilization of Iodine from $Al(NO_3)_3$ Solutions.

The difficulties anticipated in removing iodine from organic solvents suggest that reasonably complete volatilization of the iodine is desirable before contacting the dissolver solution with the solvent. Several experi-

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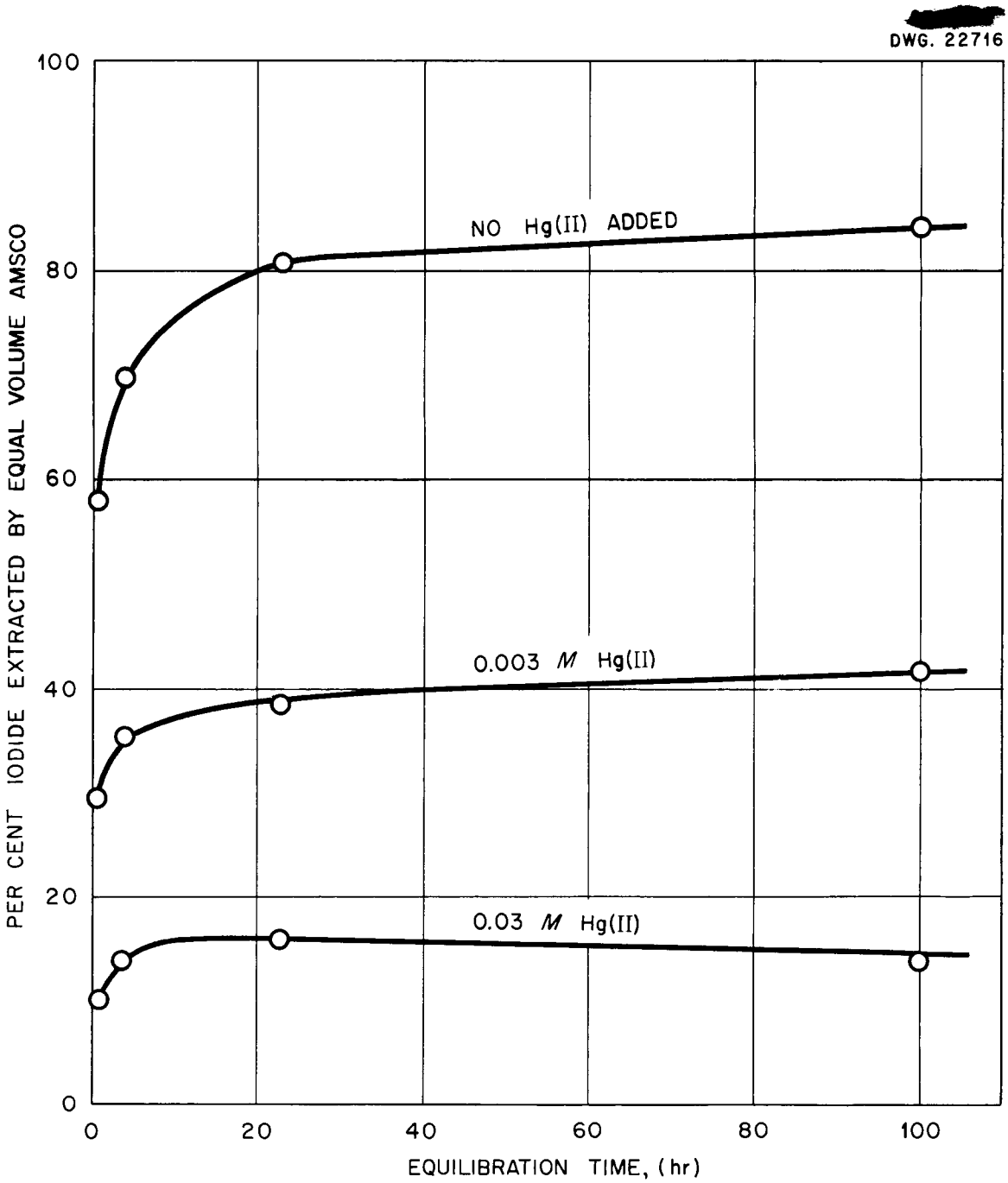

Figure 12. Extraction of Iodine from Aluminum Nitrate Solutions, 25°C. 2.2 *M* $Al(NO_3)_3$, 0.5 *M* HNO_3, 10^{-5} *M* KI; Shaken With Equal Volume Amsco 123-15.

[redacted]

ments were carried out to elucidate the factors influencing iodine volatility.

1. Absence of Hg(II). The volatility of iodine from solutions containing 2.2 M $Al(NO_3)_3$ and 0.5 M HNO_3 was measured for iodide concentrations [1] of 10^{-6}, 10^{-5} and 10^{-4} M by a series of "open beaker" experiments. Iodide concentration was adjusted by adding appropriate amounts of 0.1 M KI solution containing tracer I^{131} to facilitate iodine analysis. After addition of the iodide, the solutions were warmed to 95-100° C in a water bath and loss of iodine was determined radiometrically at approximately 5 minute intervals. Fairly complete and rapid volatilization to the iodine was observed for all solutions. About 15 minutes heating was required to volatilize 99% of the iodine at 10^{-4} and 10^{-5} M initial iodide concentrations and about 30 minutes was required to volatilize 98% of the iodine at an initial concentration of 10^{-6} M.

2. Presence of Hg(II). A similar series of experiments was made for solutions containing 2.2 M $Al(NO_3)_3$, 0.5 M HNO_3 and .01 M $Hg(NO_3)_2$. Initial iodide concentrations were adjusted to 10^{-6} and 10^{-5} M. No significant loss of iodine was observed for these solutions even after heating at 95-100° C for about 60 minutes.

Attempts to increase the volatility of iodine by the addition of strong oxidizing agents $K_2Cr_2O_7$ and $Ce(NO_3)_4$ did not give significantly increased

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1 The concentration of fission product iodine-131 in dissolver solutions after 30-day cooling of the fuel element is estimated to be 10^{-5} - 10^{-6}M.

[redacted]

[redacted]

evolution rates.

The results of the tests indicate that iodide in the absence of Hg(II) is fairly quantitatively and rapidly oxidized (by air or nitrate) to iodine which can be volatilized from solution. When excess Hg(II) is present, however, the oxidation and subsequent evolution of iodine is greatly retarded.

One cannot conclude from these experiments that little or no iodine will be volatilized during the dissolution of aluminum fuel elements, since during dissolution probably a large fraction of Hg(II) catalyst amalgamates with the aluminum. However, even small amounts of Hg(II) remaining in solution are expected to retard and perhaps prevent complete volatilization of iodine. Apparently the well-known strong complexing of Hg(II) with iodide ions[1] either prevents the oxidation of iodide due to a large shift in the oxidation potential of iodide or lowers the rate of oxidation by removing iodide ion as such from solution due to the formation of HgI^+ and HgI_2, etc. The stability constants for HgI^+ and HgI_2 at 25° C for solutions of ionic strength (μ = 0.5 M) are reported [2] as follows: $(HgI^+)/(Hg^{++})(I^-) = 7.4 \times 10^{12}$ and $(HgI_2)/(Hg^{++})(I^-)^2 = 6.6 \times 10^{23}$. Using these constants the equilibrium concentration of I^- in solutions containing 10^{-6} M total iodide and 0.01 M Hg(II) is estimated to be approximately 10^{-14} M.

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1 L. G. Sillen - op. cit.

2 I. Qvarfort and L. G. Sillen, Acta Chem. Scand. 3, 505-19 (1949).

[redacted]

[redacted]

3. Displacement of Iodide by Bromide. The foregoing results suggest that methods for removing iodine by volatilization must necessarily involve destruction of the strong Hg(II) iodide complexes.[1] By adding excess bromide ion (as HBr) iodide was found to be displaced sufficiently by bromide to permit its removal by volatilization. The results of several experiments are summarized in Table 4.

Table 4

Volatility of Iodine in $Al(NO_3)_3$ Solutions
2.2 M $Al(NO_3)_3$, 0.5 M HNO_3, 0.01 M $Hg(NO_3)_2$

(I^-)	(Br^-)	$(Br)/[Hg(II)]$	% Iodine Volatilized(1)
10^{-4}	0	0	0 ± 5
10^{-4}	.02	2	95 ± 2
10^{-5}	.02	2	95 ± 2
10^{-4}	.05(2)	5	98 ± 2

(1) After 20 minutes heating at 95° C with gentle air stirring.
(2) Some oxidation of excess bromide to bromide appeared to occur in this experiment.

Table 4 shows that although Hg(II) bromide complexes are considerably less stable than the Hg(II) iodide complexes; i.e., $HgBr^+/(Hg^{++})(Br^-) = 1.12 \times 10^9$ and $HgBr_2/(Hg^{++})(Br^-)^2 = 2.1 \times 10^{17}$ at 25° C ($\mu = 0.5$) [2], the addition of amounts of Br^- to the extent of 2 or more moles of Br^- per mole of Hg(II) sufficiently displaces iodide from Hg(II) to enable fairly rapid and complete removal of iodide by volatilization methods. Addition of HBr might best be

1 The possibility of iodide being strongly bound to fission products such as ruthenium should also be considered. The addition of bromide would also be expected to have a large effect on the extractability of Ru(III) or (IV) by hydrocarbons.

2 P. O. Bethge, I. Jonevall - Westoo and L. G. Sillen, Acta Chem. Scand. 2, 828-38 (1948).

[redacted]

[redacted]

made in the cooler-digester tank which could be fitted with an air bubbler to purge the liberated iodine.

From a corrosion standpoint, Br^- concentration as such in solution should be rather low if about 2 moles of Br^- are added per mole of Hg(II). Using the stability constant above for $HgBr_2$, the uncomplexed Br^- ion concentration in solution should be ca. 10^{-7} M. Corrosion damage to stainless steel due to the addition of HBr is thus not expected to be large.

II. Possible Application of Anion Exchange.

A. Anion Exchange of U(VI) from $Al(NO_3)_3$ Solution.

The adsorbability of U(VI) from $Al(NO_3)_3$ solutions was measured using the strong base quaternary amine resin, Dowex-1. The distribution coefficients, D = amount of U(VI) per kg resin/amount of U(VI) per liter solution, were determined after equilibrating for two hours. Under the conditions of experiments, i.e., 1.5×10^{-2} M U(VI), loading of resin was of the order of 10 - 50% of the capacity of the resin. The distribution coefficients found at several $Al(NO_3)_3$ concentrations are summarized in Table 5.

[redacted]

[redacted]

Table 5

Adsorbability of U(VI) from $Al(NO_3)_3$ Solutions by a Strong Base Anion Exchange Resin, T = 25°C; all solutions 0.5 M in HNO_3 (24 hr. shaking).

M $Al(NO_3)_3$	D*
0.5	7
1.0	20
1.5	60
2.0	275
2.5	660

*Based on air dried weight of NO_3^- -form Dowex-1.

The strong adsorbability of U(VI) from $Al(NO_3)_3$ solutions ($D \cong 660$ at M $Al(NO_3)_3$ = 2.5) suggests that anion exchange methods might be considered for the removal of U(VI) from $Al(NO_3)_3$ solutions. For the resin used in these experiments the rates of equilibration were found to be quite slow however, for example with 2.5 M $Al(NO_3)_3$, 0.5 M HNO_3 $D \cong 120$ after 30 minutes as compared to D = 660 near equilibrium. More rapid equilibrium rates were observed as aluminum nitrate concentration decreased.

B. Recovery of Hg(II) from Waste Solutions. Since rather large amounts of $Hg(NO_3)_2$ catalyst are used in dissolving aluminum fuel elements, methods for recovering mercury from waste solutions warrant consideration. Possibly anion exchange methods might be used taking advantage of the facts that negatively charged Hg(II) halide complexes readily form on the addition of small amounts of halide and that complexes of this type have been found to adsorb very strongly on strong base anion exchangers. For example, the distribution coefficient of Hg(II) in 1 M HCl was found to be ca. 10^5 with Dowex-1 resin [1].

1 Chem. Div. Quarterly Prog. Report, ORNL-1260, Dec. 31, 1951.

[redacted]

[redacted]

Appendix V

I. Status of Maintenance Experiments 11-30-53

In order to establish the operability of the maintenance philosophy which must inevitably have a profound effect upon the plant design, we have embarked on a program to develop satisfactory tools to be used in routine and emergency maintenance shutdowns. It is important to distinguish between these two categories of maintenance situations. As an example of the distinction, let us consider the replacement of a processing vessel which is necessitated by a leak.

A. Routine Maintenance

The process line is emptied of solutions which are transferred to a temporary storage vessel. The vessel to be replaced is rinsed several times alternately with steam and HNO_3. If the leak is large enough to have released significant quantities of U^{233} and fission products, the outside of the vessels is steam cleaned and the liquid is collected. The processing basin is then filled with water and the vessel to be replaced is emptied.

The locking units on the Parker fittings leading to the vessel are spun off one by one and blind plugs spun on the two open pipes. This is accomplished by using a remotely driven torque wrench developed for this purpose. Any leakage is into the vessels so the basin water will not become contaminated. This technique has not been demonstrated experimentally to date.

[redacted]

[redacted]

B. Emergency Maintenance.

A situation in which for some reason it might be impossible to empty the leaking tank might be characterized as an emergency maintenance situation. In this case it is required to be able to seal off the tank without dumping fission products into the basin. We have tested experimentally the following procedure which works very well.

A hydraulically operated vise which transmits 15,000 p.s.i. to the jaws (see Figure 13) is used to crimp the tube over approximately a 1 square inch area. Such a crimp leaks if it is sawed in half. However, if one merely cuts the crimped pipe with an underwater welding torch, the two ends of the cut are sealed against leaks. Thus one can remove the leaky tank. It is necessary, however, to replace not only the tank but also the entrance and exit lines back to the dividing valves.

C. Alternate Procedure.

An alternate maintenance procedure which can be used in general has been tested experimentally. The tank and its leads are filled with water. A hollow C-shaped clamp (see Figure 14) is placed around the lead and a refrigerant circulated through the clamp. A positive seal ice cake ~5 in. long is formed in about 15 minutes. It is necessary to insulate the clamp from the basin water but this has been shown to be altogether practical experimentally.

[redacted]

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PHOTO 11808

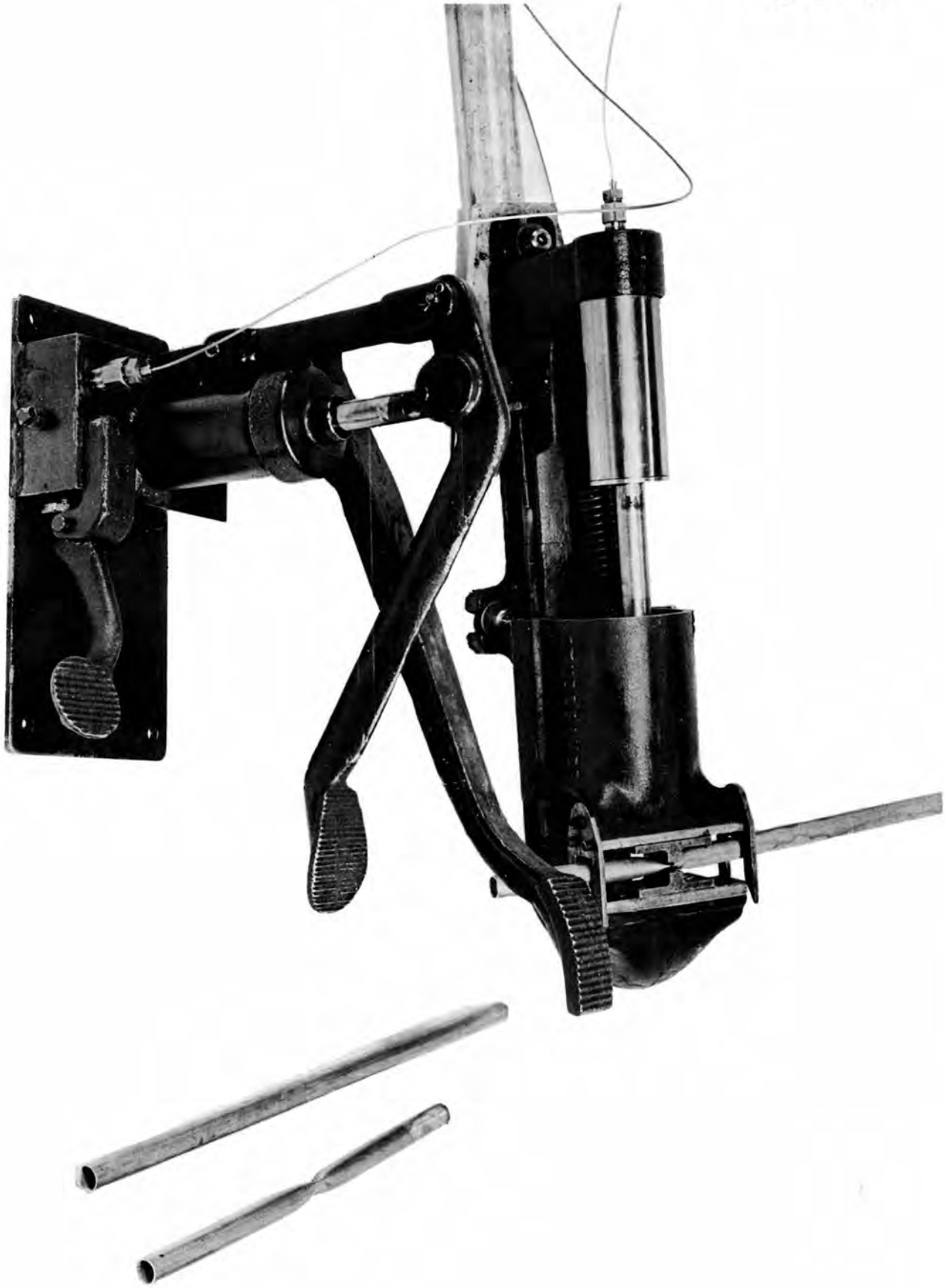

FIGURE 13

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FIGURE 14

[redacted]

Appendix VI

Complete Flowsheet for the HOPE Process

Although it was necessary in Section IV in discussing the Hope process to introduce the flowsheet for the sake of clarity no exhaustive details were given. It is felt that the reader who is familiar with existing extraction processes might wish to compare the Hope proposition in detail with other separation schemes. Accordingly we are including in the following four figures the full details of the flowsheet as we now believe it should be. All quantities are given in engineering units.

[redacted]

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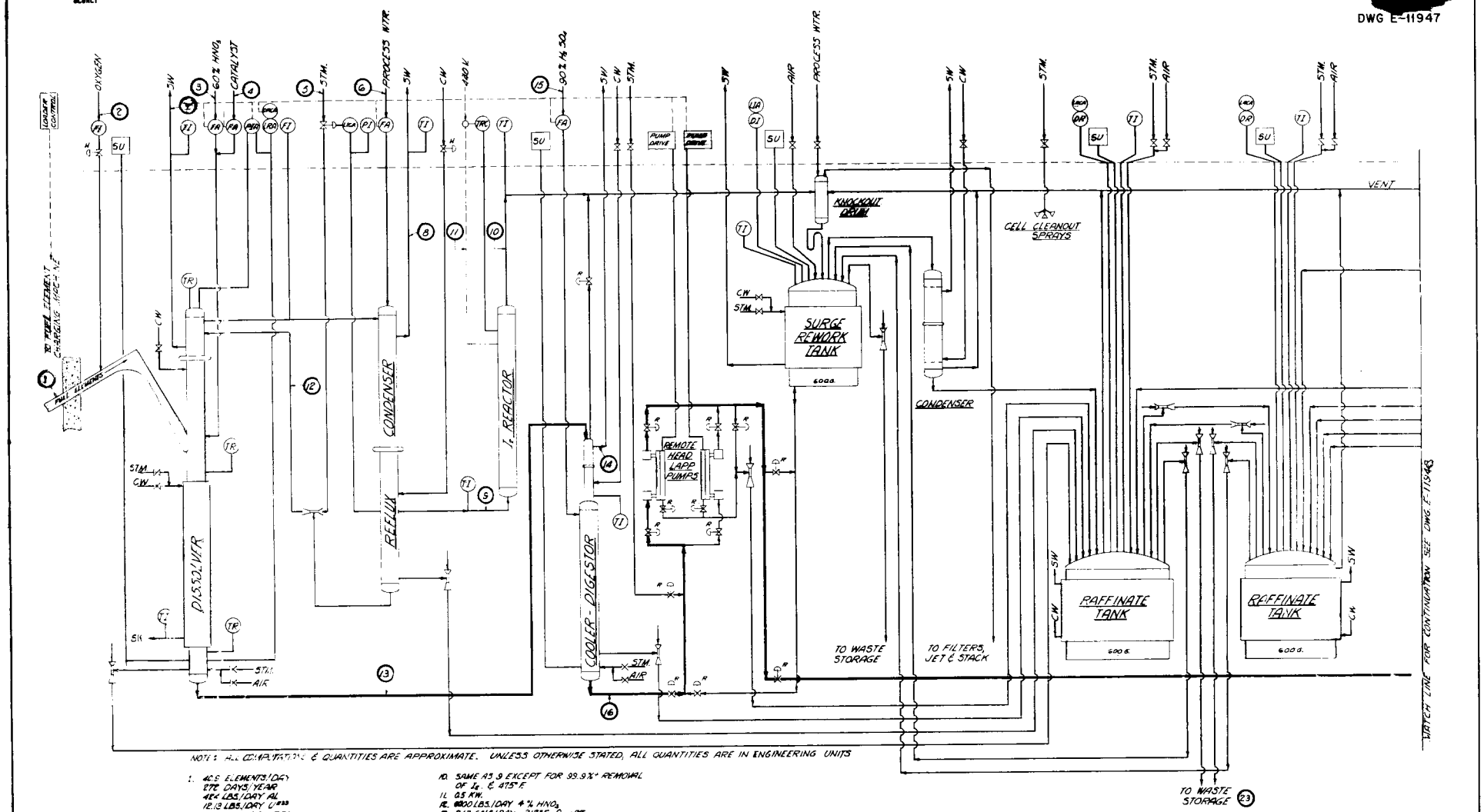

NOTE: ALL COMPUTATIONS & QUANTITIES ARE APPROXIMATE. UNLESS OTHERWISE STATED, ALL QUANTITIES ARE IN ENGINEERING UNITS

1. 40.5 ELEMENTS/DAY
   272 DAYS/YEAR
   416 LBS./DAY AL
   12.13 LBS./DAY U^{235}
   3.03 LBS./DAY F.P.'s
   1.68 LBS./DAY SILICON
2. 1080 FT³/DAY, S.T.P. OF O_2
3. 16060 LBS./DAY 60% HNO_3
4. 14.04 LBS./DAY $Hg(NO_3)_2$
5. 360 LBS. DAY OF STEAM
6. 12900 LBS./DAY OF PROCESS WTR.
7. 3.6 X 10⁶ BTU/DAY OUT
8. 1.3 X 10⁶ BTU/DAY OUT
9. 1880 FT³/DAY @ 77°F.
   0.41 MOLES/DAY O_2
   1.77 MOLES/DAY N_2
   0.00 MOLES/DAY $NO + NO_2$
   0.15 MOLES/DAY N_2O
   2.35 MOLES/DAY H_2O
10. SAME AS 9 EXCEPT FOR 99.9%+ REMOVAL OF I_2 & 47.5°F
11. 0.5 KW.
12. 6800 LBS./DAY 4% HNO_3
13. 943 GALS/DAY, 21.6°F, ρ = 1.27
    2.0 M AL
    0.25 M HNO_3
    0.00663 M U^{235}
    0.0017 LBS./GAL. OF Si
14. 1.1 X 10⁶ BTU/DAY OUT
15. 171 LBS./DAY 90% H_2SO_4
16. 943 GALS/DAY, 90°F
    2.0 M AL
    0.25 M HNO_3
    0.00663 M U^{235}
    0.02 M H_2SO_4
    0.0017 LBS./GAL. OF SILICON
23. 1800 GAL./DAY
    1.80 M AL
    CONCENTRATE IN WASTE TANK

NOTE:
FOR NOTES & SYMBOLS SEE SHEET 4 DWG. #E-11949

SECRET

Figure 15. Dissolving, Feed Preparation and Raffinate Sampling.

MATERIALS LIST					
ITEM NO.	DWG. NO.	QTY.	NAME	SIZE	MATERIAL

REVISIONS					
SYM.	REVISIONS	DATE	APPR.	APPR.	

DRAWING NO. E-11947

DRAWN	DATE	CHECKED	DATE	APPROVED	DATE

THIS DRAWING CLASSIFIED
NO.
FOR
SCALE NONE

REFERENCE DRAWINGS | DWG. NO.

DISSOLVING, FEED PREPARATION RAFFINATE SAMPLING

OAK RIDGE NATIONAL LABORATORY
OPERATED BY
CARBIDE AND CARBON CHEMICALS COMPANY
A DIVISION OF UNION CARBIDE AND CARBON CORPORATION
OAK RIDGE, TENNESSEE

SUBMITTED | ACCEPTED | APPROVED

SHEET 1 OF 4

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SECRET

MATERIALS LIST					
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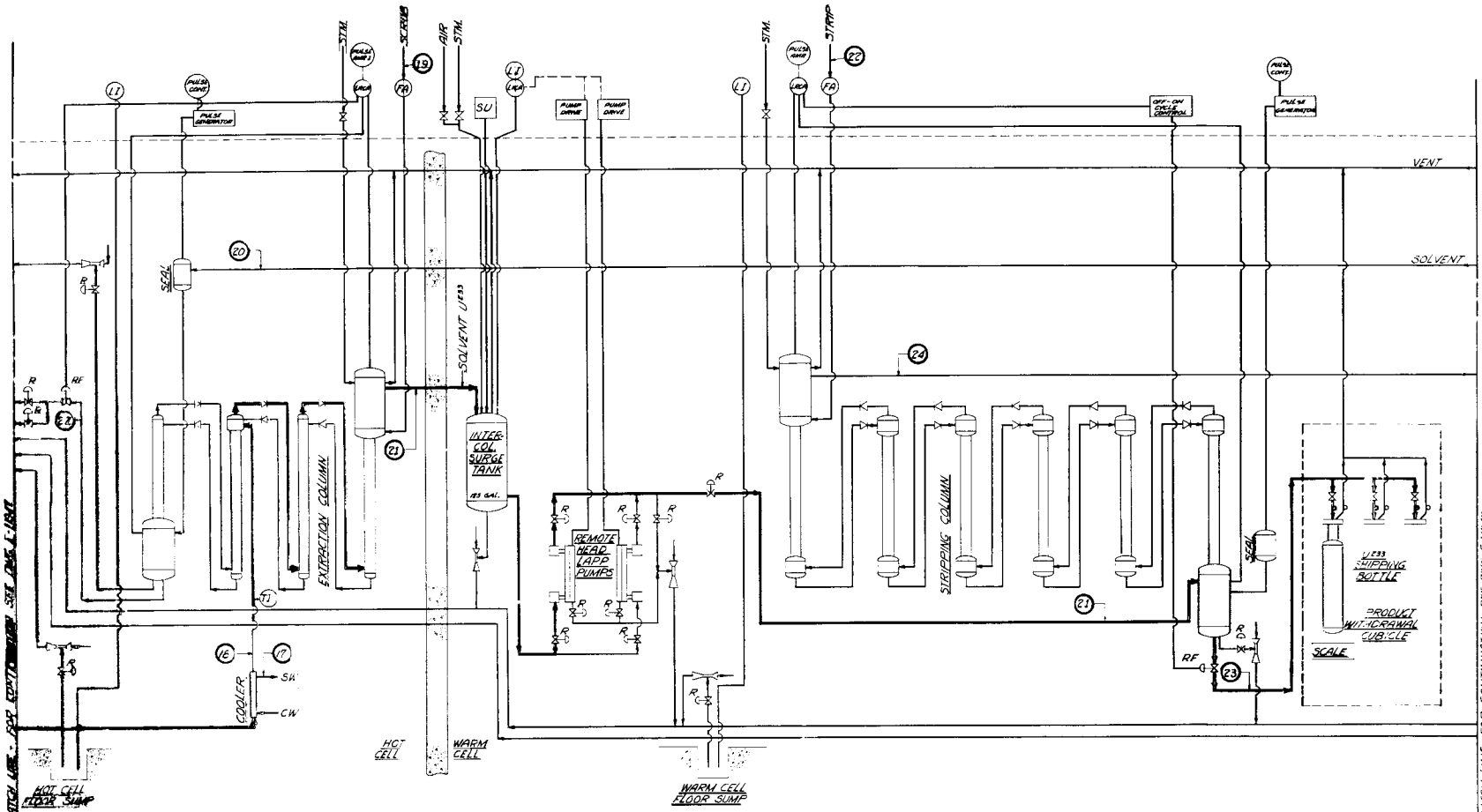
DWG E-11948



17. 1.1 X 10⁶ BTU/DAY OUT
18. 943 GAL./DAY, 68°F, FEED
19. 471 GAL./DAY, SCRUB
    1.0 M $Al(NO_3)_3$
    0.02 M H_2SO_4
20. 544 GAL./DAY, SOLVENT
    1.5% (VOL.) TBP
    98.5% (VOL.) AMSCO 125-82
21. 544 GAL./DAY, SOLVENT URANIUM
    0.0115 M U^{233}
    B.D.F. > 10⁶
22. 1413 GAL./DAY AQUEOUS RAFFINATE
    1.67 M Al
    0.167 M HNO_3
    0.02 M H_2SO_4
    0.0011 LBS./GAL. SILICON
22. 6.2 GAL./DAY, STRIP
    0.01 M HNO_3
23. 6.24 GAL./DAY, AQUEOUS PRODUCT
    1.0 M U^{233}
    .01 M HNO_3
    B.D.F. = 10⁶

OVERALL B.D.F. > 10⁶
URANIUM RECOVERY = 99.9%
24. 544 GAL./DAY SOLVENT RAFFINATE

NOTE:
FOR NOTES & SYMBOLS SEE SHEET 3 DWG. #E-11949

SECRET

REFERENCE DRAWINGS	DWG. NO.

EXTRACTION, STRIPPING AND PRODUCT REMOVAL

OAK RIDGE NATIONAL LABORATORY
OPERATED BY
CARBIDE AND CARBON CHEMICALS COMPANY
A DIVISION OF UNION CARBIDE AND CARBON CORPORATION
OAK RIDGE, TENNESSEE

SUBMITTED | ACCEPTED | APPROVED

SHEET 2 OF 140 | REV. 8

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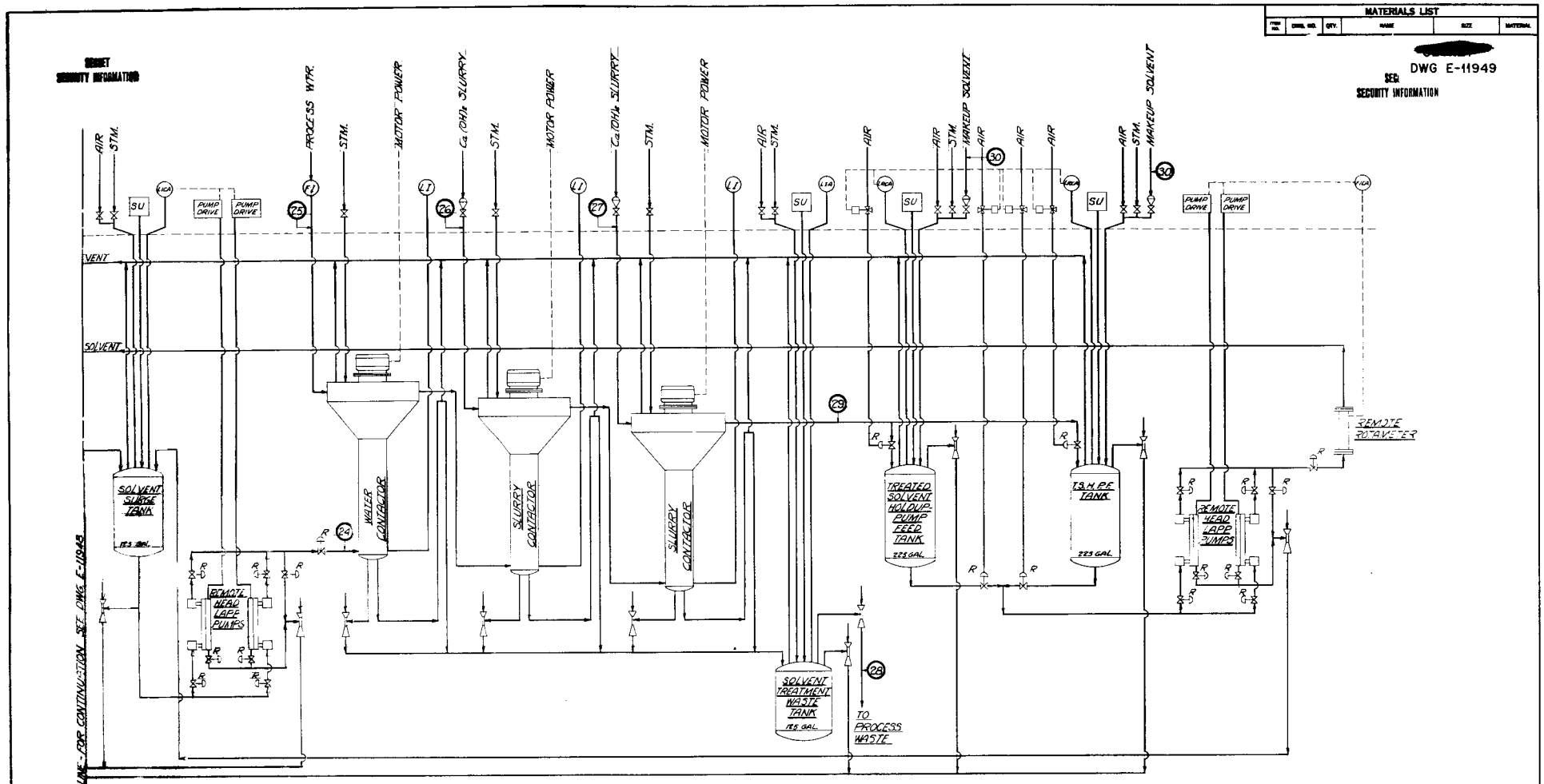
Figure 16. Extraction, Stripping and Product Removal.



24. 544 GAL./DAY, SOLVENT RAFFINATE
25. 6.9 GAL./DAY, PROCESS WATER
26. 4.4 GAL./DAY OF 26% $Ca(OH)_2$ SLURRY
27. 4.4 GAL./DAY OF 26% $Ca(OH)_2$ SLURRY
28. ~43 GAL./DAY OF MIXED WASTE CONSISTING OF $Ca(OH)_2$, $Ca(NO_3)_2$ AND SOLVENT DECOMPOSITION PRODUCTS 5% SOLVENT LOSS ASSUMED

29. 516.8 GAL./DAY TREATED SOLVENT 15% (VOL.) TBP 98.5% (VOL.) AMSCO 125-82
30. TOTAL 27.2 GAL./DAY MAKEUP STREAM 15% (VOL.) TBP 98.5% (VOL.) AMSCO 125-82

NOTES:
1. CONTINUOUSLY FLOWING STREAMS ARE INDICATED BY ---------- (XX) WHERE THE NUMBER XX IDENTIFIES THE COMPOSITION & QUANITY OF THE STREAM.
2. UESS PATH IN PROCESS IS INDICATED BY ——

SYMBOLS

INSTRUMENTS
- L = LEVEL
- D = DENSITY
- T = TEMPERATURE
- P = PRESSURE
- F = FLOW
- R = RECORDER
- I = INDICATOR
- C = CONTROLLER
- A = ALARM
- SU = SAMPLE UNIT

PIPING
- R = REMOTE OFF-ON VALVE
- RF = REMOTE FLOW CONTROL VALVE
- = FUNNEL TO PROCESS TANK
- M = HAND OPERATED CONTROL VALVE
- = INSTRUMENT OPERATED CONTROL VALVE
- = HAND VALVE

Figure 17. Solvent Recovery.

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REFERENCE DRAWINGS | DWG. NO.

SOLVENT RECOVERY

OAK RIDGE NATIONAL LABORATORY
OPERATED BY
CARBIDE AND CARBON CHEMICALS COMPANY
A DIVISION OF UNION CARBIDE AND CARBON CORPORATION
OAK RIDGE, TENNESSEE

SHEET 141 9

NO.	REVISIONS	DATE	APPR.	APPR.
DRAWING NO. E-11949				

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DESIGNED | DATE | SUBMITTED | DATE | APPROVED | DATE

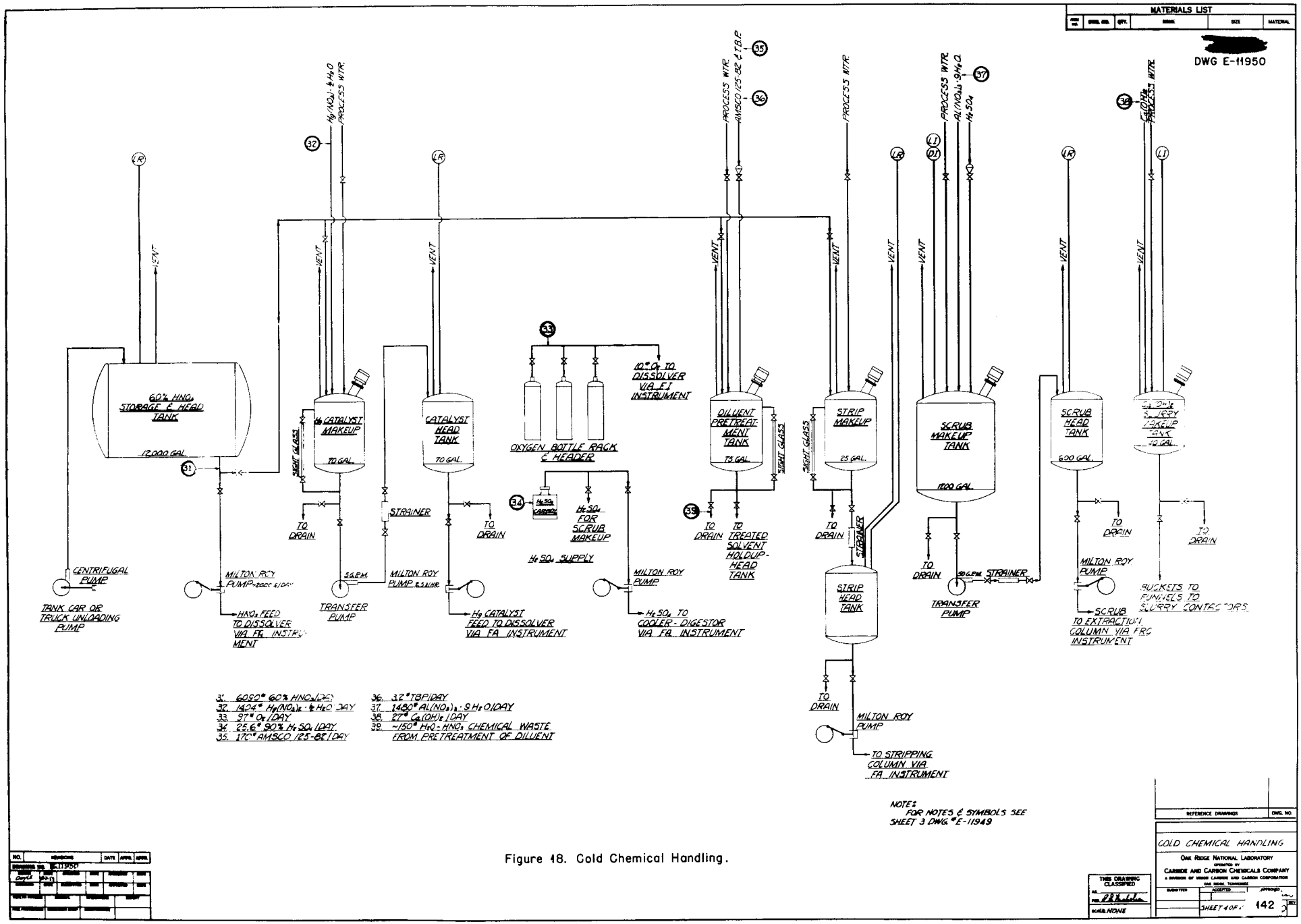

Figure 18. Cold Chemical Handling.

[redacted]

Appendix VII

Calculations Concerning Alternate Waste Disposal Methods

In order to be able to estimate the costs of the various waste storage systems under consideration, it is necessary to compute the total amount of shadow shield required for personnel protection when the storage scheme is an open tank or pit. These calculations are summarized below.

Consider the waste tank to be a source of gammas of strength N_o/cc/sec and let us erect a shadow shield "a" meters high at a distance "b" meters from the source.

The gamma flux received at a point P is

$$\Phi_p = \int \frac{N_o}{4\pi r_1^2} \frac{1}{r_2^2} \frac{d\sigma(\alpha + \beta)}{d\Omega} r_1^2 \sin\alpha \, dr_1 d\alpha \, d\psi$$

Now let us make a change of variables from r_1 to β as

$$r_1 = \frac{s \sin\beta}{\sin(\pi - \alpha - \beta)} = \frac{s \sin\beta}{\sin(\alpha + \beta)}$$

Since the situation is completely symmetrical with respect to the angle ψ we can integrate with respect to ψ to give

$$\int d\psi = \pi - \tan^{-1} \frac{a}{b}$$

[redacted]

Thus the flux received at the point P is given by

$$\Phi_p = \frac{N_o(\pi - \tan^{-1}\frac{a}{b})}{4\pi s} \int_{\beta_1}^{\beta_2} d\beta \int_{\alpha_1}^{\pi-\beta} \frac{d\sigma(\alpha+\beta)}{d\Omega}\, d\alpha$$

Where:

$$\alpha_1 \equiv \tan^{-1}\frac{a}{b}$$

$$\beta_1 \equiv \tan^{-1}\frac{a}{s-b}$$

$$\beta_2 \equiv \pi - \alpha_1$$

The problem now is to find some numerical method for computing this integral. It has been done as follows. Let us first expand the differential Compton cross-section in Legendre polynomials as follows:

$$\frac{d\sigma(\alpha+\beta)}{d\Omega} = \sum_{\ell=0}^{\infty} A_\ell P_\ell\left[\cos(\alpha+\beta)\right]$$

Then the flux becomes

$$\Phi_p = \frac{N_o(\pi - \tan^{-1}\frac{a}{b})}{4\pi s} \sum_{\ell=0}^{\infty} A_\ell \int_{\beta_1}^{\beta_2} d\beta \int_{\alpha_1+\beta}^{\pi} P_\ell(\cos Z)\,dZ$$

[redacted]

[redacted]

This is easily converted into a useful calculational form by inserting the definition of the polynomials to give

$$\Phi_p = \frac{N_o(\ -\tan^{-1}\frac{a}{b})}{4\ s} \sum_{\ell=0}^{\infty} A_{\ell 2}\,\frac{1}{\ell} \sum_{k=0}^{\left[\frac{\ell}{2}\right]} \frac{(-1)^k(2\ell-2k)!}{k!(\ell-k)!(\ell-2k)!} \int_{\beta_1}^{\beta_2} d\beta \int_{\alpha_1+\beta} (\cos Z)^{\ell-2k} dZ$$

Consider now the following series of indefinite integrals which we shall denote by I_1, I_2, I_3...

$$I_1 = \int \cos x\ dx = \sin x$$

$$I_2 = \int \cos^2 x\ dx = \frac{x}{2} + \frac{\sin 2x}{4}$$

$$\vdots$$

$$I_j = \int \cos^j x\ dx = \ldots.$$

Then we can define the quantity J_j

$$J_j = \int_{\beta_1}^{\beta_2} I_j d\beta$$

$$\Phi_p = \frac{N_o(\ -\tan^{-1}\frac{a}{b})}{4\ s} \sum_{\ell=0}^{\infty} A_\ell J_\ell$$

It can be easily shown that for the fission product gammas it is possible to fit the Compton cross section adequately with four Legendre polynomials with the following coefficients:

$A_o = .233$ $\qquad$ $A_2 = .135$

$A_1 = .313$ $\qquad$ $A_3 = .319$

[redacted]

[redacted]

Table 6

$\frac{d\sigma(0)}{d\Omega}$ Fitted with Four Legendre Polynomials

Height of Shadow Shield	Distance Source-Shield	Distance Source-Dose Point	Attenuation of Dose Obtained
5 m	20 m	50 m	1.04×10^{-9}
		100	0.38
		200	0.16
		500	0.06
15	20	50	1.56
		100	0.79
		200	0.38
		500	0.15
5	40	50	1.47
		100	0.24
		200	0.07
		500	0.02
15	40	50	1.71
		100	.70
		200	.27
		500	.10

[redacted]

[redacted]

Table 7

$\frac{d\sigma(0)}{d\Omega}$ Fitted with a Gauman

Height of Shadow Shield	Distance Source-Shield	Distance Source-Dose Point	Attenuation of Dose Obtained
5 m	20 m	50 m	1.68×10^{-9}
		100	0.87
		200	0.44
		500	0.18
20	20	50	7.10
		100	3.80
		200	1.90
		500	0.80
5	40	50	3.56
		100	2.03
		200	1.04
		500	0.42
20	40	50	20.00
		100	12.30
		200	6.30
		500	2.60

[redacted]

[redacted]

Using the results of the above calculations, preliminary estimates of alternate liquid waste storage tank installations have been prepared to obtain an order of magnitude of costs. The costs represented herein do not include any piping, instrumentation, sampling devices or other incidentals necessary for complete operation of a liquid waste tank farm. The estimates cover only the purchase, erection, and earth or concrete shielding for a tank farm consisting of three 500,000 gallon capacity tanks. The tanks considered are as follows: (1) spherical of mild steel construction internally braced to withstand 10 feet of earth covering, (2) cylindrical of mild steel construction internally braced to withstand 10 feet of earth covering, (3) cylindrical of mild steel construction built to withstand internal pressure of liquid only, and (4) cylindrical of stainless steel construction (347) to withstand internal pressure of liquid only.

The total costs of the four systems as outlined above are as follows and are tabulated in detail in Table 8 underground spherical, $528,000; underground cylindrical, $279,000; above ground cylindrical mild steel, $309,000; above ground cylindrical stainless steel, $615,000.

It is quite evident that spherical and stainless steel tanks are the most expensive installations of the four considered. No conclusive evidence, costwise, can be drawn from mild steel cylindrical tanks placed below ground with ten feet of earth covering or above ground with concrete shielding. The estimates of these two vary by approximately 10%, well within the limits of accuracy of the work.

[redacted]

[redacted]

Table 8

	Underground		Above Ground	
	Spherical (Mild Steel)	Circular (Mild Steel)	Circular (Mild Steel)	Circular (Stainless Steel)
Cost of Tanks - Erected (3)	$375,000	$195,000	$195,000	$501,000
Cost of Excavation at $1/cu yd	77,000	33,000	---	---
Cost of Backfill at $.75/cu yd	51,000	19,000	---	---
Cost of Concrete Foundations at $125/cu yd	25,000	32,000	32,000	32,000
Cost of Site Preparation, Guesstimated	---	---	10,000	10,000
Cost of Shielding			72,000	72,000
Total	$528,000	$279,000	$309,000	$615,000

[redacted]

[redacted]

Appendix VIII

I. A Summary of the Shale Tests

In order to be able to estimate how serious an accident would result from a leak in the waste storage pit it is first necessary to know what happens when the waste solution is contacted with various soils and materials which might be used in contact with the pit. A preliminary set of experiments on Conasauga shale, Bentonite and Aquagel has been completed. The results are given below.

Four columns, measuring 1-1/4 inches (3.175 cm) by 24 inches (60.960 cm) long, were prepared from lusteroid tubing. A rubber stopper, into which was inserted a 2 inch length of 4 mm diameter glass tubing, was placed in one end of each column. A double thickness of sheet glass fiber wool was cut to fit the column and placed on top of the stopper and the shale sample was placed in each column on top of the glass wool. A 2 mm diameter glass overflow tube was inserted just below the upper rim of each column so that a constant head might be maintained. The solutions were allowed to pass through the columns at full gravity flow. The pH measurements were made with a Model N-2 Beckman pH electrometer. The results obtained are shown in Table 9.

A series of jar-stirring tests were made using carrier free Cs^{137}, Ru^{106}, and Sr^{90}-Y^{90} as obtained from the Operations Division. The procedures followed were the same in all cases. Into a 1000 ml beaker was placed 500 ml of either tap water or a 2 M solution of $Al(NO_3)_3 \cdot 9H_2O$. To this was added the isotope and the solution was

[redacted]

[redacted]

stirred and sampled. Samples of pulverized Conasauga shale were then added to certain jars and the series of jars were stirred at 3 feet per second peripheral velocity (216 rpm) and sampled after 15, 30, and 60 minutes of stirring. Each sample was centrifuged for 5 minutes and aliquots were placed in aluminum counting dishes, dried under infrared heat lamps, and counted on the second shelf of an end-window GM counter. The counting results were corrected for coincidence loss (0.5% per 1000 counts) and background. The results are shown in Table 10.

A. Uptake of Aluminum^{+++} by Shale

A sectional column consisting of four sections was prepared and each section contained 20 grams of 70 mesh Conasauga shale. A 1 M $Al(NO_3)_3 \cdot 9H_2O$ solution with an initial pH 2.2 was passed through this column and each section sampled at intervals of 50 milliliters throughput. No change in the Al^{+++} concentration was detected in the effluents from any of the sections when volumetric analyses for Al^{+++} were made.

B. Uptake of Mixed Fission Products by Shale

1. MFP in 2 M Aluminum Nitrate Solution

Two liters of a 2 M aluminum nitrate solution pH 1.35, spiked with mixed fission products (initial count 140,000 c/m/ml), were passed through a column containing 75 grams of 70 mesh shale at a flow rate of about 40 milliliters per hour. A maximum removal of 30 per cent of the radioactivity was obtained after about 100 milliliters had passed through the column, but the percentage dropped rapidly and after 2 liters throughput the removal was only 4 per cent.

The column was leached with 2 liters of tap water and a negligible amount (10-15 c/m/ml) of the radioactivity could be detected in the effluent.

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An absorption curve of a sample of shale from the column indicated only the presence of cesium and this was confirmed by a radiochemical analysis of an acid leachate of the shale.

2. MFP in Tap Water

Fifteen liters of solution with an initial count of 100,000 counts per minute per milliliter were passed at a flow rate of 80 ml/hour through a column containing 75 grams of shale. Decay curves were run on the influent and after 15 liters throughput, a removal of 99.5 per cent was being maintained.

C. Sectional Columns with Bentonite and Aquagel

A sectional column, 1-1/4" diameter, capable of being sampled at each section was prepared. The top section contained 20 grams of 70 mesh shale mixed with 1 gram each of $CaSO_4$, $CaCO_2$, and $Ca_3(PO_4)_2$; the middle section contained 20 grams of shale mixed with 1 gram of Bentonite; the third section contained 40 grams of shale. A second column was prepared in the same manner except that 1 gram of Aquagel was used in the middle section in place of Bentonite. A constant head of 20 inches was maintained. A 2 M aluminum nitrate solution in 0.2 M HNO_3 (pH 1.2) with 0.25 grams per liter of added carrier spiked with Sr^{90}-Y^{90} (7500 c/m/ml) was allowed to flow through each column at a rate of about 50 milliliters per hour. About 1500 milliliters of solution was passed through each column and samples were taken after every 100-200 milliliters throughput. Each sample was checked for radioactivity, Al^{+++} concentration, and pH. The results were negative in the case of both columns and there was no detectable change in the

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concentration of radioactivity, concentration of aluminum, or in the pH value. There was no clogging of the columns due to the presence of the Bentonite or Aquagel.

An attempt was made to neutralize 500 milliliters of the solution used in the above column tests. A total of 185 milliliters of a 6 M NaOH solution was required and the volume of the sludge formed equalled about 3/4 the contents of the beaker. One milliliter of the supernatant liquid counted 60 counts per minute per milliliter.

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Table 9

Column	Influent Solution	Head (inches)	Influent pH	Effluent pH	Total Vol. Through (ml)	Average Flow Rate (ml/hr)	Remarks
100 grams 20 mesh Conasauga shale	2 M $Al(NO_3)_3 \cdot 9H_2O$	17.5	1.60	1.45	3000	2400	Constant flow rate. No apparent clogging.
75 grams 60 mesh Conasauga shale	2 M $Al(NO_3)_3 \cdot 9H_2O$	19.0	1.60	1.50	5000	70	Constant flow rate. No apparent clogging.
75 grams 30 mesh Attapulgite clay	2 M $Al(NO_3)_3 \cdot 9H_2O$	17.0	1.60	1.70	6000	500	Constant flow rate. No apparent clogging.
75 grams 60 mesh Conasauga shale	2.75 M $Al(NO_3)_3 \cdot 9H_2O$	19.5	1.35	1.15	1300	20	Constant flow rate. No apparent clogging.

[redacted]

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Table 10

	Initial pH	Final pH	Initial c/min/ml	After 15 min. c/min/ml	After 30 min. c/min/ml	After 60 min. c/min/ml	Per Cent Removal (60 min)
Cesium^{137}							
500 ml 2 M $Al(NO_3)_3 \cdot 9H_2O$	1.80	1.85	2326	2376	2501	2493	nil
500 ml 2 M $Al(NO_3)_3 \cdot 9H_2O$ 5 grams shale	1.85	1.95	2381	1933	2180	2364	nil
500 ml tap water	7.40	8.10	2521	2446	2550	2438	nil
500 ml tap water 5 grams shale	6.70	7.70	2365	24	19	12	99.49
500 ml 2 M $Al(NO_3)_3 \cdot 9H_2O$ 50 grams shale		1.80	2351			710	69.80
500 ml tap water 50 grams shale		5.60	2440			0	100.
Ruthenium^{106}							
500 ml 2 M $Al(NO_3)_3 \cdot 9H_2O$	1.50	1.50	4584	4609	4578	4602	nil
500 ml 2 M $Al(NO_3)_3 \cdot 9H_2O$ 5 grams shale	1.50	1.35	4779	4073	3875	3758	21.36
500 ml tap water	7.40	8.25	4979	3816	3746	3875	22.17
500 ml tap water 5 grams shale	6.75	7.45	4577	676	533	470	89.73
Strontium^{90}							
500 ml 2 M $Al(NO_3)_3 \cdot 9H_2O$	1.50	1.80	9112	9073	8872	9079	nil
500 ml 2 M $Al(NO_3)_3 \cdot 9H_2O$ 5 grams shale	1.50	1.80	9011	8906	8942	9166	nil
500 ml tap water	7.40	8.00	9234	8466	8390	8196	11.24
500 ml tap water 5 grams shale	6.75	7.45	9008	2671	2644	2384	73.53
500 ml 2 M $Al(NO_3)_3 \cdot 9H_2O$ 50 grams shale		1.80	9062			9175	nil
500 ml tap water 50 grams shale		5.40	9120			244	97.32

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Appendix IX

Sampling Procedure

In line with the general philosophy of the Hope Plant, we have considerably altered the sampling technique from that which is currently in use at Arco. Our criterion for sampling is strictly an economic one. Samples will be taken for only two purposes: to insure effective process control, and to guard against gross losses of U^{233}. It is clearly inconsistent with the philosophy of the Hope project to sample merely for SF accountability as is presently done.

In order to get the plant started it will be necessary to take considerably more samples than are required during routine operation, but even during the start-up period sampling does not overtax the analytic staff which is provided. The start-up will require approximately 6200 samples per month; while routine operation will entail slightly less than 2000 samples per month. Since twelve analysts are provided on a three-per-shift basis, this works out to an analytical load of approximately one sample per chemist per hour, which is not excessive.

A detailed summary of the anticipated analytical requirements of the Hope plant both during start-up and regular operation is contained in Figure 19.

In all cases we have tried to overestimate the total sample load so that we feel that we have provided adequate personnel for carrying out the analytical work.

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FIGURE 19. ANALYTICAL REQUIREMENTS OF HOPE PLANT

SAMPLE SOURCE	REQUIRED FOR: PLANT START-UP	S. F. ACCOUNTABILITY	CRITICALITY	PROCESS CONTROL	TYPE OF ANALYSIS REQUIRED: U	HNO_3	SP. GR.	Hg^{++}	% TBP	β	γ	α	RCA	MASS ASSAY	EMISSION SPEC.	SPECIAL ANALYSIS	SAMPLES PER DAY (DUPLICATES)	SAMPLES PER MONTH (DUPLICATES)	TOTAL ANALYSIS PER MONTH INCLUDING START-UP	TOTAL ANALYSIS PER MONTH, ROUTINE OPERATION
DISSOLVER (F.S.)	X			X	X*	X*	X	X(½)*									6 (12)	180(360)	1260	30
COOLER-DIGESTOR (F.S.)	X			X	X*	X*	X			X*	X*					X(1)(½)	4 (8)	120(240)	1320	45
SURGE-REWORK	X	X	X	X	X	X	X										0.3 (0.6)	9(18)	54	27
IAW	X	X		X	X	X*	X										3 (6)	90(180)	540	360
IAP (F.S.)	X			X	X	X*	X*			X	X*		X*				1 (2)	30(60)	360	120
IBW (F.S.)	X			X	X(½)					X*	X*		X*				1 (2)	30(60)	210	30
IBP (F.S.)	X				X*		X*			X*	X*		X*				1 (2)	30(60)	360	0
ALL CYCLE SOLVENT TREATMENT WASTE	X	X			X												0.5 (1)	15(30)	30	30
IAX	X			X			X		X	X	X						1.5 (3)	45(90)	360	180
IIAW	X	X		X	X	X*	X										3 (6)	90(180)	540	360
IIAP (F.S.)	X			X	X	X*	X*			X	*		X*				1 (2)	30(60)	360	120
IIBW (F.S.)	X			X	X(½)					X*	X*		X*				1 (2)	30(60)	210	30
IIBP	X	X			X	X	X			X	X	X	X	X	X		1 (2)	30(60)	540	540
IIAX	X			X			X		X	X(½)	X(½)									
I AND II AS	X			X		X	X										0.4 (0)	12(0)	24	24
I AND II BX	X			X		X											0.1 (0)	3(0)	3	3
II BUTT	X			X		X	X										0.4 (0)	12(0)	24	24
CATALYST	X			X				X									0.25(0)	8(0)	8	8
HNO_3 SUPPLY	X			X		X	X										2/30(0)	2(0)	2	2
																			6205	1933

SYMBOLS AND NOTES

(½) Analysis requested on one-half the samples.
(F.S.) ≡ Flowing stream.
(1) H_2SO_4.

*Analyses for start-up only. The sample schedule for start-up assumes that all the streams are sampled at frequent intervals. Actually the peak sampling load during start-up should be considerably less than the figure given. Elimination of duplicate sampling during routine operation may be desirable.

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Appendix X

Criticality Conditions for U^{233} - H_2O Mixtures

In order to be able to have confidence in our criticality criteria which are based on a one velocity age theory model, we did a series of two group H_2O reflected calculations for mixtures of U^{233} and H_2O. Such calculations have been reliable for similar systems in the past and they serve as check-points against which to apply the simpler theory.

The calculations are rendered conservative by using 20°C as the temperature. The standard two group-two region procedure is used with the following choice of constants:

Core	Reflector
$\tau = 33\ cm^2$	$\tau = 33\ cm^2$
$D_s = 0.142$ cm	$D_s = 0.142$ cm
$D_f = 1.143$ cm	$D_f = 1.143$ cm
	$L^2 = 8.3\ cm^2$

This choice of constants is valid for moderately dilute solutions in which the diffusion properties of the core (except L^2) are controlled by the water. As mentioned in the body of the report, the more concentrated accumulations of U^{233} as UO_2 which might chain react in a fast reaction are not dangerous since the critical mass exceeds 25 kg.

The results of these calculations are summarized in the following table and graph.

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Volume % 233 in Core	U^{233} Concentration (gm/ℓ)	Critical Radius
.047	8.8	oo
.10	18.7	15.7
.30	56.1	8.48
.60	112.2	7.07
1.00	187	6.37
2.00	374	5.88
3.00	561	5.71

[redacted]

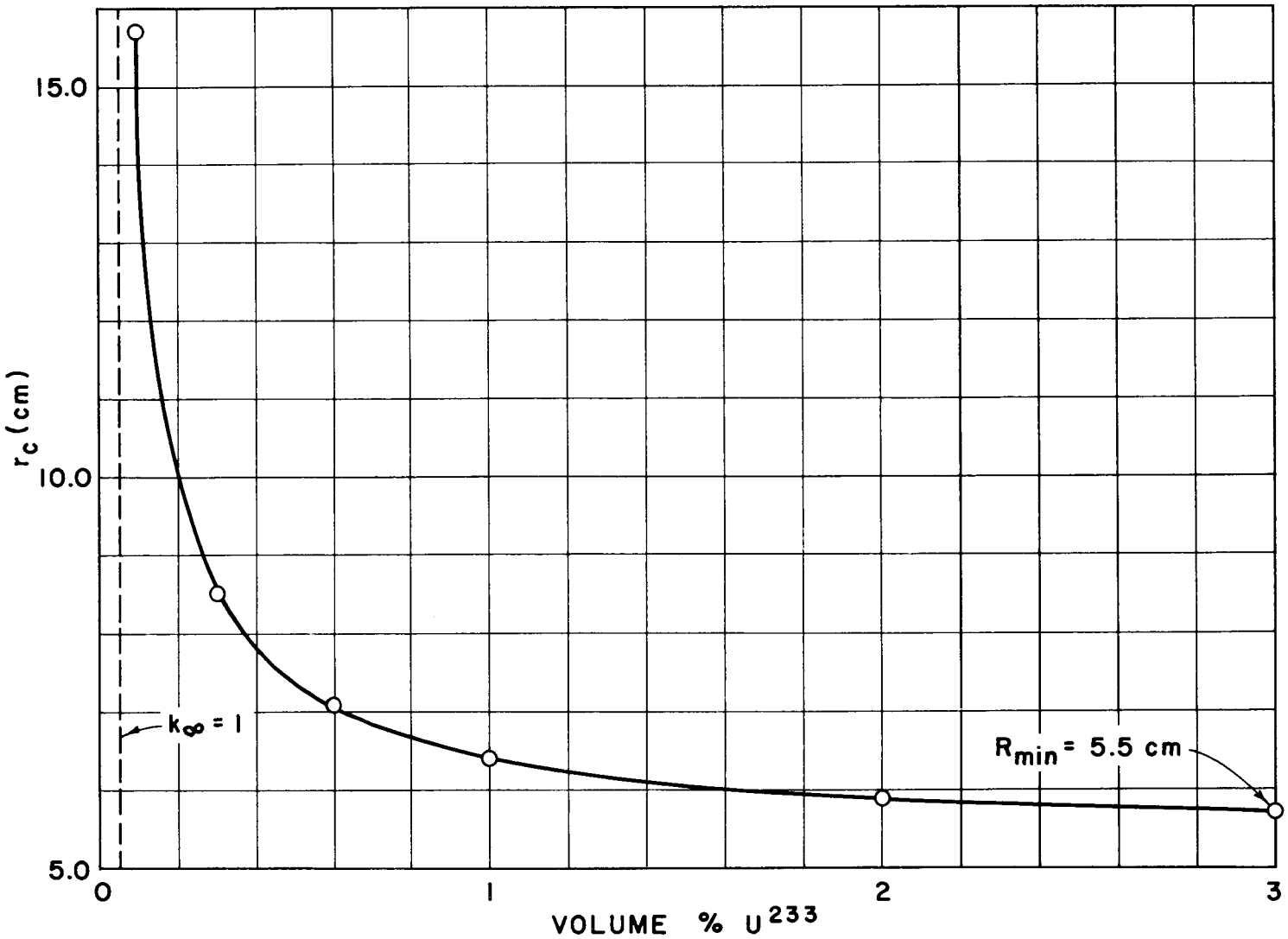

FIGURE 20
CRITICAL RADIUS OF U^{233} - H_2O REFLECTED REACTORS

DWG. 22606

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Appendix XI

Performance Parameters for Stacks

In order to evaluate the hazard which results from discharging radioactive off-gases from tall stacks an extensive study has been made of the parameters which characterize stack performance under a variety of climatic conditions. This work has been conducted cooperatively by the U. S. Weather Bureau and ORNL. Since we found the results very useful we are reproducing them in the two following figures.

[redacted]

FIGURE 21

MAXIMUM GROUND CONCENTRATION PER UNIT EMISSION RATE

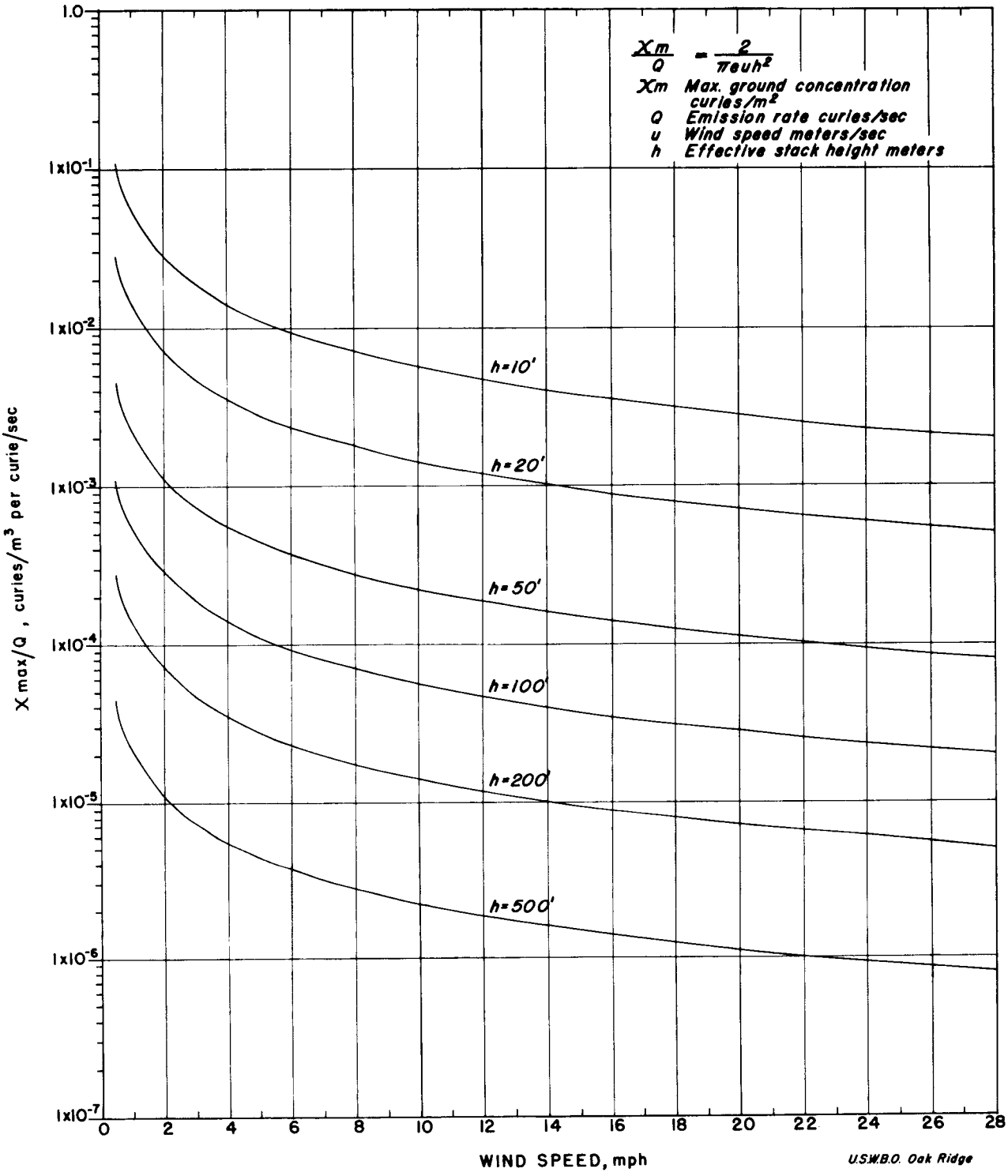
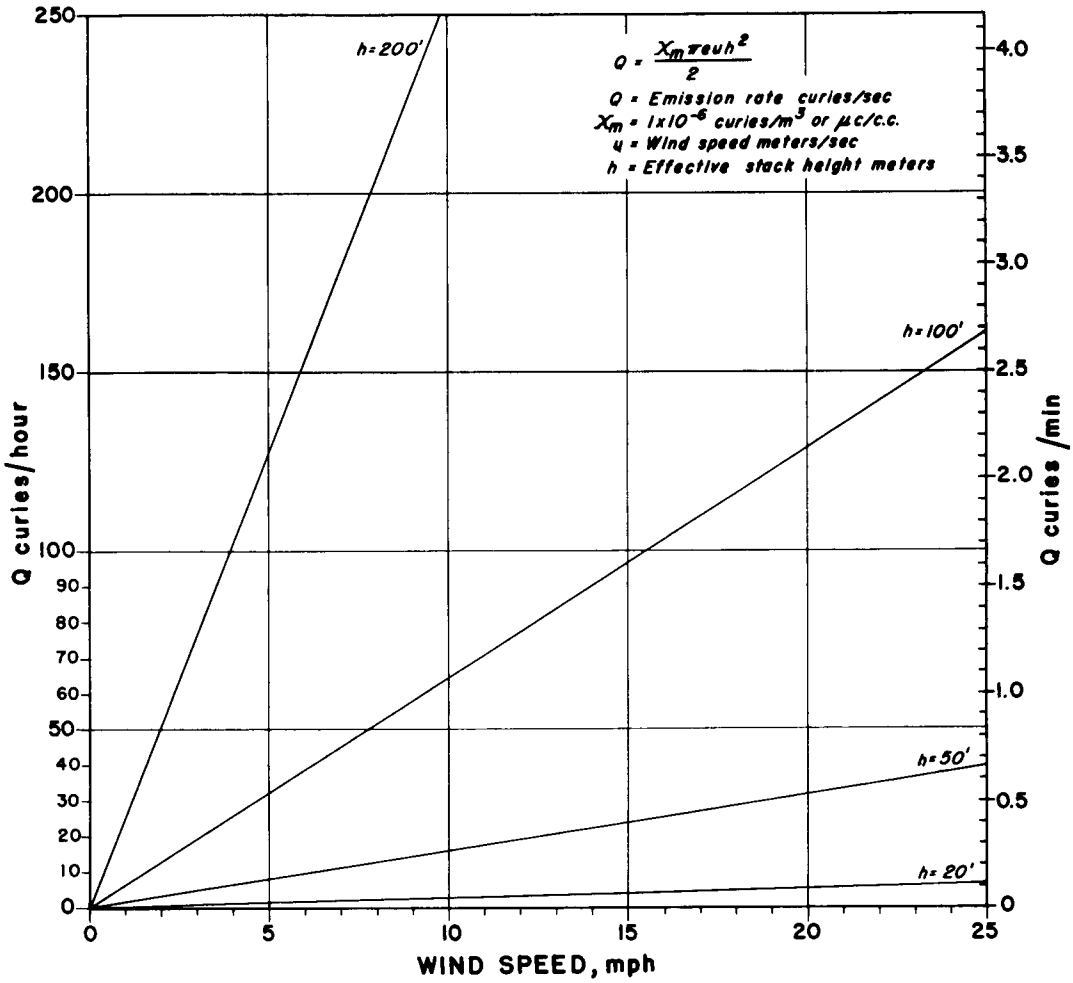
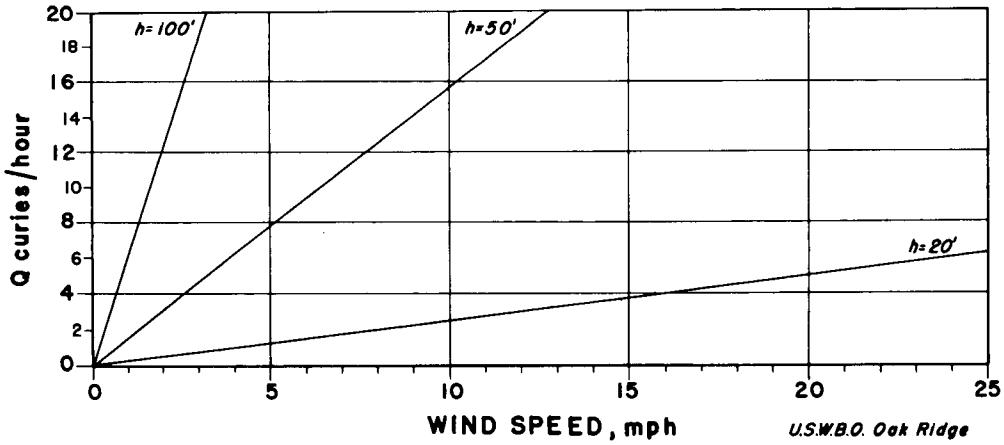


FIGURE 22

PERMISSIBLE EMISSION RATE

FOR MAXIMUM GROUND CONCENTRATION = 10^{-6} μc/cc





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Appendix XII

I. Irradiations of Asphalts and Tars

A. Work Accomplished on the Earthen Basin Program to Date

A literature survey of the use of lined earthen basins as a more economical storage method reveals that, other than for water storage, lined earthen basins have been little used. Apparently there is no valuable user history available from the literature for the storage of chemical solutions in such basins.

Industrial contacts have proven to be more fruitful than a literature search. From contacts with industry, The Bureau of Reclamation and the Asphalt Institute, the following test programs and pending test programs have been developed:

1. Test Programs (in progress)

Bureau of Reclamation - Asphaltic membranes and asphalt cements

Lion Oil Company - Asphalt cements

American Gilsonite Company - Naturally occurring asphalts (Gilsulates)

Johns-Manville Research Center - Asphaltic membranes

Gulf States Asphalt Company - Asphaltic membranes

The Barrett Division (Allied Dye and Chemical Corp.) - Coal tars

The results of gamma exposure (Cobalt-60) of some asphalts and asphaltic membranes have indicated that prefabricated asphaltic membranes and P_2O_5-blown asphalt containing 25% paraffin filler appear to be damaged less (little or no expansion or honeycombing) than the other

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asphalt materials tested. In general, it appears that gamma irradiation produces dehydrogenation of the asphalts with the subsequent occurrence of expansion and honeycombing, Figure 23.

2. Pending Test Programs

Gunite Construction Corp. - Shotcrete

Atlas Mineral Products Company - Acid resistant cements (organic, inorganic mixtures)

Ralph V. Rulon, Inc. - Acid-proof cements (furane, silicate, carbon combinations)

Portland Cement Association - Soil cements

The B. F. Goodrich Company - Rubber linings

Maurice A. Knight - Modified rubber linings (high temp.)

Western Waterproofing Company - Spraycrete (cement, resin) and acid-proof mortars

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FIGURE 23

Asphalt Irradiations

The following asphalts and asphaltic membranes were irradiated in air by a Co^{60} source located 8 ft under water at 72°F. Intensity was 2.0×10^6 R/hr.

Asphalt	Softening Point (R&B) (°F)	Penetration at 77°F	Weight (grams)	Total Irradiation (roentgens)	Outgassing Pressure (psig)	Gas Evolved per gram Asphalt Irradiated (cc)	Rate of Gas Evolution (cc/g/10^6R)	Total Expansion of Asphalt (%)	Remarks
P_2O_5-Blown (Lion Oil Co.)	184	58	525	5×10^8	23.8	3.0	6×10^{-3}	69	(a) Swelled cone-shape and honeycombed throughout.
P_2O_5-Blown[a] (Lion Oil Co.)	184	58	515	1×10^9	14.3	1.7	1.7×10^{-3}	65	(a) Expanded cone-shape and honeycombed throughout.
Roofing (Lion Oil Co.)	190	24	525.6	5×10^8	11.4	1.4	2.8×10^{-3}	58	(a) Expanded cone-shape and honeycombed throughout.
Gulf-Seal Canal Lining[b] (Gulf States Asphalt Co.)	---	---	157.3	5×10^8	4.7	2.9	5.8×10^{-3}	0	(a) No expansion. (b) Durometer hardness decreased 12%.
Asbestos Pre-Fab Liner[c] (Johns-Manville)	---	---	99.3	5×10^8	0.35	0.34	0.7×10^{-3}	0	(a) No expansion. (b) Durometer hardness increased 17.4%.
Membrane-W/25% $BaSO_4$ Filler (Lion Oil Co.)	183	57	593.4	7.3×10^7	3.3	0.131	1.8×10^{-3}	46	(a) Expanded cone-shape and honeycombed throughout.
Membrane-W/25% Slate Flour Filler (Lion Oil Co.)	188	52	621.1	1.17×10^8	4.54	0.46	3.8×10^{-3}	53	(a) Expanded cone-shape and honeycombed throughout.
Membrane-W/20% Micro-Wax (Paraffin) Filler (Lion Oil Co.)	230	71	493.3	8.35×10^7	4.78	0.6	7.3×10^{-3}	4.5	(a) No indication on surface of honeycombing.

(a) Leakage of off-gas and/or recombination may have occurred in this specimen.

(b) Prefabricated membrane containing roofing scraps, felt, and vegetable fibers.

(c) Prefabricated membrane containing asbestos fiber.

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