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CHEMICAL ASPECTS OF REACTOR CONTROL

by

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PREFACE

The intention of this report has been to set forth an outline of the problem, describe briefly the difficulties which are likely to be encountered, and indicate the direction in which an experimental program should proceed. The possible applications of chemical control have been discussed with regard to both land-based reactors and mobile power plants.

The study represents about three and a half weeks of effort on the part of the Chemical Control Task Force, and thus cannot be considered to be a complete feasibility report. However, it may be possible to draw some conclusions from the work which has been done so far, despite its limited scope.

Many members of the Physics Department, Engineering Department and others from outside the Chemistry Subdivision have contributed to this study by way of conversations and reports which have been made available. The authors gratefully acknowledge all assistance which has been provided.

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I INTRODUCTION

The following definitions will apply throughout this report. By "chemical control" we shall mean a method of reactor control in which the power level is changed by variation of the concentration of neutron-absorbing poisons which are distributed more or less uniformly through the reactor core. Control of the reactor by variation of the moderating ability of coolant water might conceivably be considered a form of chemical control, but this method has received only cursory inspection in the study. For the purposes of this report, chemical control will be taken to have the narrower definition given above. The poison materials may be contained either in the coolant water or in a separate system of piping installed in the core. The two distinct methods will be referred to as coolant-water systems and auxiliary systems, respectively.

The major objectives of this study have been the following:

(a) To consider in general the requirements of a reactor control system and the capabilities and limitations of chemical control methods.

(b) To consider the chemical substances which might be used as poisons, particularly with regard to chemical compatibility and corrosion properties, availability, thermal and radiation stability, and residual radioactivity.

(c) To indicate the methods of concentration control which might be used in conjunction with the possible physical systems and chemical substances, considering these methods with regard to safety and reliability, space and power requirements, speed of operation, etc.

(d) To outline the experimental work which will be necessary to develop a practical chemical control system.

The Requirements of a Nuclear Reactor Control System

Any scheme or combination of schemes which is chosen for control of a power reactor must be able to perform a number of distinct functions. These are listed below approximately in order as to the rate at which the control system must add or subtract reactivity.

(a) "Shim" control - The control system must be able to compensate for such excess reactivity as is initially built into the core to insure long life of the reactor, and it must be able to decrease this compensation slowly over the lifetime of the reactor as the fuel becomes depleted and fission product poisons accumulate.

(b) Normal start-up and shut-down - A stable reactor will ordinarily have a negative temperature coefficient of reactivity, which means that the control system must be able to compensate for the extra reactivity the reactor will have in the cold condition over its reactivity while hot. In addition, for complete shut-down, the reactor is ordinarily made about five percent sub-critical. The rate of change of temperature for a reactor contained in a steel pressure vessel will not ordinarily exceed about 50°F per hour in order to avoid excessive thermal stresses. The maximum rate of addition of reactivity should not exceed 0.04% per second to insure a margin of safety under the rate which will make the reactor prompt critical.

(c) Changes in power level while operating - A change in demand for power will change the power level of the reactor automatically, by virtue of the negative temperature coefficient of reactivity. However, depending on whether the power plant is operated at constant cold leg temperature, constant average temperature or constant steam pressure, the control system will have to make some adjustment to maintain the desired conditions.

(d) Stabilization of the power level - A reactor with a sufficiently negative temperature coefficient will not require adjustment of the controls to compensate for small fluctuations in pressure, pumping speed, etc., which might affect reactivity.

(e) Xenon transient - If it is desired to over-ride maximum xenon, the reactor must be provided with a large amount of excess reactivity. The control system must be able to compensate for this excess reactivity when the xenon is not present, and it also must be able to add or subtract reactivity at a rate corresponding to the rate at which xenon is being accumulated or destroyed. The maximum rate required in this case is addition of negative reactivity to compensate for xenon burn-out when the reactor has been brought to full power after accumulating maximum xenon.

(f) "Scram" control - If any of the safety circuits incorporated in the reactor indicate that a dangerous condition is being approached, or if a mistake has been made by the operator, it is desirable that negative reactivity should be inserted at the maximum possible rate. The method of scramming the reactor should be such that recovery from the scrammed condition is relatively easy, inasmuch as false scrams are likely to occur.

Bases for Comparing Control Systems

The most important factors to be considered in judging possible methods of control are safety and reliability. Any system which can perform all the above functions satisfactorily must still be considered inadequate if it does not fail safe. Additional factors which must be considered are initial and operating costs (particularly for a commercial type power plant), space and weight requirements for the components and auxiliary equipment (especially

important for a mobile power plant), possible detrimental effects on other portions of the apparatus (corrosion of coolant system by chemicals, fouling of heat transfer surfaces, etc.) and, of course, the ability of the system to perform the function for which it is intended. In considering costs, one must take into account not only the cost of the control system but also any savings which may be effected by reduced core size, increased, efficiency of heat transfer, etc.

Rod Control vs Chemical Control

The art of reactor control by mechanical movement of absorbing rods has been brought to a high degree of development, yet certain difficulties are inherent in the method. Among the difficulties may be listed the following:

(a) The channels through which the rods move must occupy a portion of the core volume (about 2% for STR), thus limiting the compactness of the core structure,

(b) With rods partially inserted only part of the core is being used for production and transfer of heat,

(c) Headroom must be provided for the rods to be fully withdrawn,

(d) Mechanical difficulties are associated with the method, principally variation of frictional resistance to movement, and also including problems of shock sensitivity, inaccessibility, wear, etc., and

(e) The temperature coefficient of reactivity is dependent on rod position.

Some of the above difficulties would be eliminated by chemical control, as achieved either by varying the concentration of neutron absorbing poisons in the coolant water or in an independent system of pipes incorporated into the core structure (coolant and auxiliary chemical control systems, respectively). A disadvantage common to both types of chemical control is that thermal expansion of the solution reduces the poison concentration in the reactor, thus decreasing the negativity of the temperature coefficient of reactivity. However, it has been suggested that a desirable temperature coefficient could be preserved by designing the core with a higher metal-to-water ratio than would be used with rod control. Whether such a core design would result in a non-optimum shape, requiring more U-235 for criticality, will have to be determined by calculations.

Coolant vs Auxiliary Chemical Control

There are advantages and disadvantages associated with one or the other of the two possible methods of chemical control. Some of these are tabulated below:

	A. Auxiliary System	B. Coolant System
a	Piping occupies part of the core volume.	Permits most compact core structure.
b	Many different types of absorbing media possible since materials and solvents may be chosen to fit system.	List of possible poisons limited by requirement for use in water systems, compatibility with system materials, corrosion properties of the solution, etc.
c	Might be made to fail safe only with certain substances as poisons.	Fails safe.
d	Probably requires smaller total quantity of poison. (Continued)	Poison is dispersed throughout entire coolant system and probably requires larger total quantity of poison.

	A. Auxiliary System	B. Coolant System
	(Continued)	
e	High poison concentration required.	Low poison concentration suffices.
f	Small volume to be processed for reconcentration, indicating: i. Fast action, ii. Small storage requirements, iii. Low reconcentrating power requirements, and iv. Small size reconcentrating equipment.	Large volume must be processed for reconcentration, indicating: i. Slower action, ii. Either large storage requirements or iii. High reconcentrating power requirements, or optimum combination, and iv. Large size reconcentrating equipment
g	Poison chemicals cannot foul heat transfer surfaces, interfere with ion exchanger operation, water purification, etc.	Decomposition of poison chemicals could have deleterious effects, fouling heat transfer surfaces, interfering with water purification, etc.
h	If poison has high residual radioactivity, compact auxiliary equipment is easily shielded.	Entire coolant system requires extra shielding if poison has high residual radioactivity.
i	Mixtures, slurries and other heterogeneous sytems handled relatively easily.	Heterogeneous poison systems handled with difficulty.
j	Properties of coolant and moderating fluid are not affected by changes in poison concentration.	Changes in poison concentration affect properties of coolant and moderating fluid.
k	Requires piping to be brought through pressure vessel.	Requires no extra seals through pressure vessel.
l	Residual radioactivity does not prohibit access to boiler, pumps or coolant piping.	Residual activity of poison, if long-lived, limits access to coolant system.
m	Poison can be completely removed easily.	High flow rate through concentration system necessary to reduce poison concentration to low value in a given time.

An auxiliary system, if it can be made fail safe, would seem to be better for a mobile power plant in which weight and space requirements are important, frequent and rapid changes in power level would be expected, and minimum cost is sacrificed to some extent in favor of convenience, maneuverability, etc.

It would seem that a coolant water poisoning system would be preferable to an auxiliary system in the case of a land-based power plant where the price of storage capacity is not excessive, weight and size of equipment are not important (except as to cost) and the core structure should be the most economical one. This is particularly true of a base-load power plant which would require relatively infrequent changes in power level.

II METHODS FOR REACTOR CONTROL

A. Methods for Reactor Control by Neutron Absorption

Twenty-one methods for the control of nuclear reactors by the use of neutron-absorbing poisons are given in Table 1. Nine of these are applicable as direct additions to the primary coolant system, but all are applicable in an auxiliary system independent of the primary coolant system.

Variable "greyness" is possible with thirteen of these types. The remaining eight are inherently opaque ("black") to neutrons and hence control must be achieved by a change in physical position. The grey types achieve control by concentration changes, thereby utilizing the entire core for heat generation and heat transfer. A reactor which is controlled by blacking-out a portion of its core volume with opaque poisons is less desirable than one in which control is achieved by varying the overall greyness of the core environment.

TABLE 1 - METHODS FOR REACTOR CONTROL BY NEUTRON ABSORPTION

No.	System Type	Active Poison	Inactive Carrier	Physical Phases	Applicable to Primary Coolant	Grey (1)
1	Gas rod	Gas	None	1	No	Yes
2	Liquid rod (2)	Liquid	None	1	No	No
3	Solid rod	Solid	None	1	No	No
4	Duplex gas	Gas	Gas	1	No	Yes
5	Gas lift	Liquid	Gas	2	No	Yes
6	Fluidized solid	Solid	Gas	2	No	Yes
7	Soluble gas	Gas	Liquid	1	Yes	Yes
8	Foam	Gas	Liquid	2	Yes	Yes
9	Soluble liquid	Liquid	Liquid	1	Yes	Yes
10	Emulsion	Liquid	Liquid	2	Yes	Yes
11	Soluble solid	Solid	Liquid	1	Yes	Yes
12	Slurry	Solid	Liquid	2	Yes	Yes
13	Gas-filled particles	Gas	Liquid	2	Yes	Yes
14	Liquid filled "	Liquid	Liquid	2	Yes	Yes
15	Coated solid "	Solid	Liquid	2	Yes	Yes
16	Gas-filled rod	Gas	Solid	1	No	No
17	ditto	Gas	Solid	2	No	No
18	Liquid-filled rod	Liquid	Solid	1	No	No
19	ditto	Liquid	Solid	2	No	No
20	Alloy rod	Solid	Solid	1	No	No
21	Clad rod	Solid	Solid	2	No	No

(1) Methods which are "grey" to neutrons are capable of variable neutron attenuation.
(2) Reference to "black" rods are included for completeness.

Each of these methods has inherent advantages and disadvantages, which when considered together with the factors described above will determine the choice of the optimum system to fill the requirements of a particular application.

The first method given in Table 1 using a gaseous poison in an auxiliary system has the advantage of being simple to control. A gaseous neutron absorber, such as BF_3 or He-3 isotope, can be added to or removed from closed-end tubes within the core by the use of external pressure alone. Thus the greyness of the control tubes can be automatically controlled with ease. The disadvantage involved in losing poison due to an external leak in the system cannot be ignored, but can be minimized by conservative design.

Method two, the liquid control rod, was considered for STR, Mark I, using mercury in tubes. A complete report* covering this system was issued in 1950. This system, too, must be designed conservatively because an external leak could cause the loss of the poison from the core.

Solid control rods, Method three, are being used in STR, Mark I, and have been discussed above.

The duplex gas control, Method four, is similar to Method one, except for the addition of a gas which essentially absorbs no neutrons. The second gas could serve two functions. The total pressure could be adjusted to nearly equalize system pressure and thus require less tubing strength inside the pressure vessel. Furthermore, the second gas could be used as an indicator for leak detection, provided the poison-gas itself was not applicable to a conventional leak-detection scheme. Here also, the potential loss of

* WAPD-3 Preliminary Feasibility Report on Liquid Control Systems for the Naval Reactor, January 1, 1950.

poison through an external rupture must be considered a disadvantage.

The liquid poison and gaseous non-poison, Method five, represents a means by which mercury control rods, mentioned above, may be converted to a variable greyness type of control. Mercury, for example, in closed-end tubes located within the core, with helium gas bubbling into the mercury near the bottom of these tubes would have a variable effective density depending upon the rate of helium flow. The excess mercury would collect in a reservoir at the top of the tube, outside the core. Variable greyness control is thereby achieved coupled with the advantages that the helium could be used for leak detection, and loss of helium pressure through an external leak would cause a safe failure in that the mercury would return to its normal (blackest) density.

A similar scheme may be employed using a solid poison, in place of mercury, for Method six. Finely divided solids are fluidized by a high velocity gas stream, such that the mean density of the stream may be controlled by the gas velocity. Safe failure due to loss of gas pressure results when the fluidized powder collapses into its densest form.

Methods seven through twelve are applicable to either the primary coolant system as additions or to an independent auxiliary system. The liquid-soluble gas system, Method seven, is illustrated by the use of He-3 isotope in the water of the primary coolant system, or by BF_3 in a suitable solvent in an auxiliary system. In the primary system, the soluble gas concentration could be controlled by a degassifier-pressurizer, but interference due to the presence of hydrogen is a complicating factor which may lead to a complex design.

Method eight, using a two-phase gas and liquid mixture in which the gas is the poison, is most likely limited to application in an auxiliary system.

A foam in the primary coolant system would probably be undesirable because of unstable transients in the foam due to sudden pressure changes in passing through the pumps and sudden temperature changes in passing through the reactor. The use of a foam in an auxiliary system would seem to have no advantage over the use of the gas alone, or in combination with another gas.

Miscible liquids, Method nine, apply principally to an auxiliary system where the solvent is not limited to water. The use of high-boiling solvents could reduce the working pressure of the auxiliary system to atmospheric, thereby minimizing the loss of poison by flashing, in the event of an external leak.

Mixtures of insoluble liquids, or emulsions, Method ten, physically are more stable than foams, and as two-phase fluids are conveniently subject to changes in relative proportion by simple mechanical methods. For example, an emulsion of mercury in water could be considered for use in the primary system, providing all internal materials were resistant to amalgamation. Loss of control with loss of pumping power accompanied by settling-out of mercury from the core must be considered a disadvantage, even though loss of pumping power itself is serious. In an auxiliary system, loss of pumping power could be accommodated by design so that the poison settled into the core thimbles. If the operating pressure in such an auxiliary system was nearly ambient pressure, even a rupture of the auxiliary system could result in safe failure without loss of poison from the core thimbles.

Soluble solids, Method eleven, have been adopted for emergency shutdown of STR, Mark I. A concentrated solution of boric acid is manually pumped into the primary coolant system to poison the reactor. Reactor control can be achieved by varying the concentration of the soluble salt in the primary coolant

stream. Increases in concentration are obtained by pumping in a solution more concentrated than is required, while withdrawing an equal volume of system solution. The reverse effect is achieved by pumping in water, or a dilute solution of poison.

A variety of methods exist in industry for the fast or slow reconcentration of water-soluble solids. The choice depends upon other factors such as weight and space tolerances for large storage tanks compared to the high cost of performing rapid evaporations discontinuously. In an auxiliary system, a solution of a solid in an appropriate solvent would be particularly attractive if the operating pressure were low enough to preclude loss of poison from the core in the event of an external rupture.

Method twelve, the slurry system, has essentially the same features as emulsions, Method ten, except that in this case the problems of abrasion and erosion are encountered. Abrasion is serious only if the solid is hard relative to the surfaces under consideration, and erosion is serious only if the effective density of the solid is far from that of the liquid. Crystalline growth may occur since the loop is not isothermal. Solution will generally be at the expense of the smallest particles because of their higher surface free energy and growth will take place on the largest crystals by virtue of their lower (relative) surface free energy. By-pass monitoring through appropriately sized screens will remove excessively large particles; the size of the by-pass depending upon the rate of crystal growth. Particle deposition on the core surfaces, as seems to occur with corrosion "crud", would be unfavorable to good heat transfer.

A special case of Method twelve, slurries, consists of a poison surrounded by an inert solid protective coating to produce poison particles with altered physical and chemical properties which may be applicable under specialized conditions.

The fabrication costs of slurries for Methods thirteen, fourteen, and fifteen may be higher than can be justified compared to other available methods.

The final six methods, sixteen through twenty-one, are not defined as applicable to the primary coolant system because of the specification of the solid non-poison carrier. This infers the use of position-controlled rods, filled with a poison, or acting as a solvent for the poison, and as such are not considered to be chemical control.

In reviewing this list of twenty-one methods for reactor control with poisons, those eight methods which do not provide variable greyness control are eliminated from primary consideration because the improved heat generation and heat transfer achieved with greyness control represent prime objectives.

Foams, Method eight, are eliminated from primary consideration on the basis of their pressure and temperature sensitivity. The coated poisons, Methods thirteen through fifteen, are also eliminated from primary consideration because of their probable high initial cost, without compensating advantages.

The remaining nine methods warrant further consideration, pending the availability of elements and compounds suitable to the requirements of the particular system. These methods are the gas rod, duplex gas, gas lift, fluidized solid, soluble gas, soluble liquid, emulsion, soluble solid, and the slurry.

Poison Cross-Sections Required for Each Method

In considering Method one, compressed gas in an auxiliary system, the maximum poison concentration will be obtained at maximum pressure and minimum temperature. At 2000 psia and 81°F, an ideal gas has a maximum concentration of 5.5 gram atomic weights per liter. For a given reactor, a minimum microscopic absorption cross-section can be computed for use in the selection of a suitable gaseous neutron absorber. For example, shutdown of an STR-type reactor might require more than 0.02 cm^2 absorption cross-section per cm^3 core, and if the auxiliary gas is limited to 2 volume percent of the core volume, then the minimum microscopic absorption cross-section is 300 barns per atom. Thus, the separated isotope, He-3, and natural BF_3 can be considered for use in the control of such a reactor.

Since both of these gases can be used for leak detection with modified commercial leak-detecting devices, no need for a tracer gas involving the duplex gas method (four) is indicated. For this reason, type four methods may be eliminated from primary consideration.

To achieve greyness control in Method five systems, more than half of the auxiliary system within the core is presumed to be occupied by the carrier gas. A minimum macroscopic absorption cross-section for the liquid poison can be calculated for a given reactor and auxiliary system. Assuming, for example, that more than 0.02 cm^2 absorption cross-section per cm^3 core, and that less than 2 volume percent of the core volume is occupied by the auxiliary fluids, the macroscopic cross-section for absorption must be greater than 2 cm^2 per cm^3. Mercury satisfies this requirement, and can be considered for use in this type of control system.

Type six systems, fluidized solids in an auxiliary system, are subject to the same minimum macroscopic cross-section, and roughly to the same carrier-gas proportion. A wide selection is available for solids that exceed this minimum requirement. Boron, cadmium, gadolinium and many others in either elemental or solid compound form may be considered for this application. The changes in the character of the fluidized solids with inclination from the vertical may be disadvantageous to such a system in a mobile reactor.

Primary-coolant poisons may seriously affect the moderating effect of the water if the poison concentration exceeds one volume percent. To provide 0.02 cm^2 absorption cross-section per cm^3 core, at 0.5 water-to-core volume ratio, and not exceed the above mentioned concentration value, the macroscopic cross-section for neutron absorption by the poison must exceed 4 cm^2 per cm^3 based on the pure poison. Water-soluble compounds are known for several elements which exceed this assumed limitation. Lithium borate easily satisfies this example calculation and is under serious consideration for this application. If reduced moderation is tolerable, so that higher concentrations are permissible, more compounds are available from which to choose a primary-coolant poison.

Water-insoluble compounds, for systems using emulsions or slurries, need to have a macroscopic absorption cross-section in excess of 4 cm^2 per cm^3 based on the pure poison, as in the case of the soluble poisons. This minimum may be reduced in the event that more than one volume percent of poison may be tolerated in the primary coolant, as in the case of the water-soluble poisons.

The foregoing discussion illustrates that eight systems are worthy of further consideration for the control of nuclear reactors by neutron absorption. These methods are the gas rod, gas lift, fluidized solids, soluble gas, soluble liquid, emulsion, soluble solid, and the slurry.

The example calculations are overly simplified, but are included to illustrate the general type of computations that can be made, once the reactor characteristics are assumed, to establish minimum criteria to aid in the selection or rejection of otherwise acceptable materials as chemical control substances.

Since for some of the systems warranting primary consideration, more than one chemical material may be acceptable from the neutron absorption point of view, it is emphasized that certain experimental work may be carried forward, independent of the nuclear properties of the active material. For example, the effects of inclination from the vertical on the character of the fluidized-solid system might be shown to be negligible in tubes of less than a certain inside diameter, without reference to any particular chemical compound. Especially in those cases involving the separated isotopes, such as He-3 or B-10, the natural forms can be used for all studies not involving their nuclear properties.

Naturally, such studies which involve specific chemical or physical properties will have to be carried out with the true compound.

B. Methods for Reactor Control by Variable Moderation

Reactor control by neutron absorption is wasteful of neutrons in that some of these neutrons otherwise could be used for fast fission of U-238. Control in this situation is achieved by varying the moderator efficiency so that an adjustable proportion of neutrons are thermalized.

In water-moderated reactors, the reactivity of the core is decreased as the mass of water associated with the core is reduced. This reduction of water mass may be accomplished by several methods. An increase in water temperature decreases the density of water, such that the core contains less water mass and the reactivity is reduced. This negative temperature coefficient of reactivity is used in STR, Mark I, to provide stability against disturbing transients.

Additional control may be achieved by adjustable displacement of water from the core volume. Five of the previously discussed methods of control using poisons may be considered for control by moderation changes. These methods are the gas lift, emulsion, slurry, soluble liquid, and soluble solid, in which one component is water and the other component is a substance having a low neutron absorption cross section.

The gas lift, in an auxiliary system, could use helium bubbling through water, under pressure, such that more or less water is in the core volume depending upon the rate of helium flow. Steam may be used instead of helium, in which case circulation is not required. Control may be achieved by adjustment of the total volume in a relatively constant pressure system, so that an adjustable water density is obtained.

Emulsions, slurries, pellets, soluble liquids and soluble solids may be considered for use either in an auxiliary system or in the primary coolant.

Emulsions, slurries and pellets would probably introduce fewer engineering complications in an auxiliary system, and are considered primarily in this respect.

Soluble liquids and solids in the primary coolant system for moderator control would have to be present in concentrations appreciably greater than one volume percent to produce a significant degree of control. The effects on pressure drop and heat transfer of such concentrations would require detailed investigation to evaluate possible deleterious effects.

The disadvantage associated with the use of a poison in an auxiliary control system wherein poison is lost from the core in the event of an external rupture is eliminated with non-poisonous moderator control. An external rupture would permit expulsion of moderator by flash evaporation and would result in a decrease in reactivity.

An internal rupture could result in an unsafe condition if primary coolant replaced the inert material used to displace moderator. The resulting increase in moderation would presumably increase the reactivity of the core.

In the interests of overall neutron economy and potential advantages in reliability and simplicity, the use of an adjustable moderator for control would seem to warrant further consideration.

III CHEMICAL PROPERTIES OF NEUTRON ABSORBERS

It has been estimated above that the gas lift and fluidized solid methods would require a minimum macroscopic absorption cross-section of 2 cm^2 per cm^3 of poison material. This is the minimum value for all the methods discussed, and is based upon the given assumptions for an STR-type core.

A. Natural Elements and Compounds

Twenty natural elements have macroscopic absorption cross-sections in excess of this minimum, and are listed in Table 2. Separated isotopes will be considered below where specific advantages may warrant the increased cost.

Gas Rod

The chemical suitability of a particular neutron absorber depends upon whether it is to be considered for use in the primary coolant or in an auxiliary system; whether it is supposed to be soluble or insoluble, etc. The pressure-controlled gas rod has been discussed, and gaseous BF_3 has been shown to have adequate cross-section at 2000 psi. The dry gas is not corrosive to steel, copper, or brass, and is stored and shipped in commerce in contact with these materials at pressures up to 2000 psi. Mercury is used for liquid seals, and petroleum oils are used for lubricants in systems handling gaseous BF_3. Satisfactory thermal stability is indicated by the fact that BF_3 is produced at about 1000°C.

However, BF_3 reacts with water to produce an acid solution, so that an internal system rupture would produce a corrosive primary solution.

TABLE 2

NATURAL ELEMENTS HAVING MACROSCOPIC ABSORPTION CROSS SECTIONS IN EXCESS OF 2 cm^{-1} @ 0.025 ev.

Element	Symbol	Cross Section	
Gadolinium	Gd	1380 cm^{-1}	(1)
Samarium	Sm	180	(1)
Cadmium	Cd	110	(4)
Boron	B	100	
Europium	Eu	93	(1)
Dysprosium	Dy	35	(1)
Iridium	Ir	31	(2)
Mercury	Hg	16	(2)
Rhodium	Rh	11	(2)
Indium	In	7.6	(3)
Rhenium	Re	5.7	(2)
Gold	Au	5.5	(2)
Hafnium	Hf	5.1	(3)
Lithium	Li	3.6	
Silver	Ag	3.5	(2)
Cobalt	Co	3.1	
Erbium	Er	2.8	(1)
Thulium	Tm	2.7	(1)
Holmium	Ho	2.4	(1)
Lutecium	Lu	2.4	(1)

(1) Rare earth, type 4f.
(2) Easily reduced noble metal.
(3) Easily hydrolyzed in hot water.
(4) Neutron absorption increases with core temperature. Very desirable.

Gas Lift

The gas lift method of control was discussed above with reference to mercury as the absorbing liquid. The cross-section of mercury is adequate for this application. The resistance to amalgamation of possible materials of construction needs experimental verification. Helium is well suited for the motive gas, having low neutron absorption, and excellent chemical inertness and thermal stability. The increase in the size of the helium bubbles as they rise through the mercury, due to the decrease in hydrostatic pressure, can be minimized by operation at a high total pressure. Otherwise the tube would be much darker grey to neutrons at the bottom than at the top. Aqueous lithium borate might be substituted for mercury.

Fluidized Solids

Most of the elements listed are suitable for use as fluidized solids, and many compounds are available to suit this application. The choice may better be narrowed by other considerations such as cost, residual radioactivity, abrasiveness and density. Powdered boric acid might be very suitable, depending upon other factors. Generally, the compounds are cheaper than the elemental forms.

Solutions, emulsions and slurries may be considered for application either in the primary coolant system or in an auxiliary system. With reference to the primary coolant system, chemical compatibility between system materials and the poison is complicated by the fact that some of the structural materials have no equivalent substitute. For example, all poisons which attack zirconium may be eliminated from primary consideration because of the desirable use of zirconium in the core structure. Fluorides are in this category.

The structural material for an auxiliary system is not predicated upon previous development, although in addition to being strong and chemically compatible with the poison, the absorption cross-section preferably should be low for that portion within the core volume.

Soluble Gas

In the above list of elements, suitable for chemical control by neutron absorption, none is a gas at normal temperature and pressure, and only boron has compounds which are gases at normal temperature and pressure, according to data given in the HANDBOOK OF CHEMISTRY AND PHYSICS. Table 3 lists the gaseous compounds of boron with pertinent data affecting their suitability as nuclear poisons.

TABLE 3 - CHEMICAL PROPERTIES OF GASEOUS COMPOUNDS OF BORON

Formula	Melting Point °C	Boiling Point °C	Remarks
$B_2 H_5 Br$	-104	+10	hydrolyzes: $HBO_2 + HBr + H_2$
BCl_3	-107	+12	hydrolyzes: $HBO_2 + HCl$
$B_2 H_5 Cl$	-	-78 (18mm)	highly unstable
BF_3	-127	-101	hydrolyzes (hot): $HBO_2 + HF$
$B_2 H_6$	-165	-92	hydrolyzes: $HBO_2 + H_2$ Thermally unstable
$B_4 H_{10}$	-120	+18	hydrolyzes: $HBO_2 + H_2$ Thermally unstable

These data indicate that none are suitable for addition to the primary coolant. It is concluded, therefore, that no suitable gas is available for use in solution with the primary coolant, separated isotopes excepted.

937 026

In an auxiliary system, the halide gases may be applicable providing a compatible combination of solvent, structural material, and gas can be found. The hydrides decompose at core temperature. The possible number of combinations is so great that comment is deferred until a detailed investigation is warranted.

Soluble Liquid

Mercury is the only suitable element which is liquid at normal temperature and pressure. Rhenium oxychloride (ReO_3Cl) and five compounds of boron are liquids under these conditions, according to the above reference. Table 4 lists these liquids and their pertinent chemical properties.

TABLE 4 - CHEMICAL PROPERTIES OF LIQUID NUCLEAR POISONS

Formula	Melting Point °C	Boiling Point °C	Remarks
BBr_3	-46	90 (740mm)	hydrolyzes: HBO_2 + HBr
B_5H_9	-47	0 (66mm)	hydrolyzes: HBO_2 + H_2 Spontaneously flammable in air
B_5H_{11}	-123	65	hydrolyzes: HBO_2 + H_2
B_6H_{10}	-65	0 (7.2mm)	hydrolyzes: HBO_2 + H_2
B_2H_5I	-110	0 (78mm)	hydrolyzes: HBO_2 + HI + H_2
Hg	-39	360	Insoluble in water
ReO_3Cl	4.5	131	Unknown - probably hydrolyzes to acid solution

These data indicate that none are suitable for solution with the primary coolant. Again, the large number of possible combinations of a non-aqueous solvent, structural material, and liquid poison preclude rigorous comment without detailed investigation.

Emulsions

Mercury, being insoluble in water, and a liquid poison may be considered for use as an emulsion. The resistance to amalgamation of proposed system materials, and the stability of such an emulsion are unknown and require experimental determination. Emulsions in an auxiliary system, where the continuous phase is not predetermined may offer a wide selection. Here again, the fairly large number of possibilities indicates more investigation.

Soluble Solids

In the primary coolant, where water is the solvent, the sulfates of the rare earths and of cadmium, indium and hafnium would hydrolyze and form an acid solution. Compounds of the noble metals, being easily reduced, would probably be unsatisfactory, since deposition might take place in the core and produce an irreversible shutdown. These arguments reduce the list of suitable elements to cobalt, lithium and boron, but experimental determinations may show that compounds of cadmium, for example, do exist which do not hydrolyze to produce an acid solution, and which meet all the other requirements. Thus, although boron seems to be suitable and desirable, other useful compounds should not be ignored in the developmental program.

Soluble solids for use in an auxiliary system offer a wider choice because of the possible large number of solvents which may be useful. Furthermore, the pH is not restricted to that of the primary coolant.

Slurry

The use of a slurry either in the primary coolant or in an auxiliary system immediately raises the problems of abrasion and erosion. Slurries have a compensating advantage in that the control of concentration can be

accomplished with devices which consume little power. Such devices as a centrifugal separator, filter, or settling tank may be used to reduce the concentration of poison in a slurry. Increases in concentration are easily accomplished by flow reversal or similar low-power means to reintroduce a slightly thicker slurry into the control stream. For these reasons, it seems to be advisable to continue consideration of slurries, at least for an auxiliary system where fewer moving parts are subject to abrasive wear. An ultra-fine slurry may be stabilized against rapid settling out upon flow failure.

B. Separated Isotopes

The main objections to the use of separated isotopes are their higher cost and their limited availability. Further consideration is warranted because lower concentrations may be important, and in certain cases, the natural mixture does not have a high enough cross-section to be considered at all.

Since fourteen natural elements have microscopic cross-sections (@ 0.025 ev) in excess of 100 barns per atom, only separated isotopes having cross-sections in excess of this value will be considered. Table 5 lists those isotopes which have microscopic cross-sections above this limit, and above the cross-section of the natural mixture.

In addition to the isotopic cross-section, the abundance of the isotope in the naturally occurring mixture, and the cross-section of the natural mixture are tabulated. From these values, the ratio of the isotopic to the natural cross-sections, and the proportion of the natural cross-section contributed by the normal presence of the isotope are given.

TABLE 5 - SELECTED HIGH CROSS-SECTION ISOTOPES

Isotope	Isotopic Cross-Section	Abundance % of Natural	Natural Cross-Section	Cross-Section Ratio Isotopic: Natural	Cross-Section Due to Isotope % of Natural
He-3	5,000	0.00013	0.008	625,000	100
Li-6	950	7.5	70	14	100
B-10	4,000	18.8	750	5.3	100
Cd-113	25,000	12.3	3,100	8.1	99
In-115	197	95.8	190	1.0	99
Nd-143	290	12.2	44	6.6	80
Sm-149	50,000	13.8	10,000	5.0	69
Eu-151	8,400	47.8	4,300	2.0	93
Gd-155	70,000	14.7	36,000	1.9	29
Gd-157	160,000	15.7	36,000	4.4	70
Dy-164	2,700	28.2	1,000	2.7	76
Yb-168	11,300	0.14	36	310	44
Hf-174	~500	0.18	120	~4.2	~0.75
Hf-177	370	18.4	120	3.1	57
Ir-191	950	38.5	440	2.2	83
Hg-196	2,500	0.15	360	6.9	1.0
Hg-199	2,000	10.0	360	5.6	56

Note: Cross-sections for 0.025 ev neutrons.

A high ratio of isotopic cross-section to natural cross-section is favorable toward the use of the isotope, and a high proportion of the natural cross-section due to the given isotope further enhances its value. For example, helium-3 has 625,000 times the cross-section of natural helium, and 100 percent of the cross-section of natural helium is due to helium-3. This makes the use of helium-3 very attractive. Both factors must be considered. In this case, natural helium has far too low a cross-section to be an effective neutron absorber.

The only elements which this list includes which were not considered as natural elements are helium, neodymium and ytterbium. Thus, the choice of elements has not been greatly increased, but rather, the necessary concentration of the element has been reduced.

Neodymium and ytterbium, being type 4f rare earths are expected to have chemical properties similar to the other type 4f rare earths, gadolinium, in particular. The cross-section of natural gadolinium (36,000 barns per atom) is higher than that of any other natural rare earth and higher than the separated isotopes of any of the rare earths except samarium-149 and gadolinium.

Helium-3 is uniquely attractive but the low abundance and strategic importance dictate against its practical application.

IV RADIATION AND RADIOACTIVITY PROBLEMS IN CHEMICAL CONTROL

A. Residual Radioactivity

There are two problems which occur as a result of residual radioactivity of poison substances as induced by exposure to the neutron flux. These are: (1) the possible necessity for additional shielding of equipment external to the reactor but containing the circulating poison, and (2) the possibility that the interval between the time of shutdown of the reactor and the time when access can be had safely to the reactor compartment or system piping ("accessibility time") may be increased.

Table 6 lists some of the materials proposed for use as poisons and the approximate saturation level of radioactive disintegration activity to be expected from each of these. The activities given in the table are best used in comparing the substances with one another and cannot be taken as absolute values. The activities were calculated approximately by assuming a cycle time of ten seconds, one second of which is spent in-pile. If the half-life of the radioactive nuclide is several times the cycle time, the approximate saturation activity can be calculated simply as that which the substance would assume in a continuous flux given by the average of the

actual flux over the cycle period. The values of activity which may be encountered in practice will depend on the various times involved*, and more detailed computations will become desirable when the values of these parameters have been specified.

It may be pointed out from Table 6 that only B, Na, K, Li and He-3 show activities per cc of coolant lower than the combined O-19 and N-16 activities of the coolant water, and hence these substances will certainly pose no additional shielding problems. (Sulfates and nitrates also are not likely to be troublesome in this respect.)

Unless it is possible to drain the active poison solution to a storage tank and decontaminate the drained system easily, the accessibility time will be proportional to the half-lives given in Table 6 Again it may be pointed out that Li and B alone pose no additional problem in this respect, as the half-lives of their active species are less than those of either O-19 or N-16. Sodium and potassium might be easily flushed from the system, however, and thus might be used in a situation where storage tanks are available or where dumping is permitted. The low-energy beta activity of tritium from water or He-3 is no problem unless the tritium is constantly being ingested for long periods of time (as could be the case on a submarine).

* The saturation level of activity for a given species in a cyclic activation and decay process is

$$\text{Disintegration rate (per cc-sec)} = \frac{R(1 - e^{-\lambda\tau}) e^{-\lambda t}}{(1 - e^{-\lambda(\alpha+\tau)})}$$

where R = rate of activation of the species per cc-sec in the reactor, τ = transit time in the reactor, λ = decay constant of the species, α = decay time outside reactor, t = time after last pass through reactor.

TABLE 6 - RELATIVE SATURATION ACTIVITIES OBTAINED FROM PROPOSED POISONS

(1-gram of element in 10^{14} slow flux; 1 second in flux; 9 seconds out of flux)

Element	Atomic No. Radioactive Nuclide Z	Mass of Radioactive Nuclide A	Half Life $t_{\frac{1}{2}}$	Cross-Section Barns	Abundance Parent Nuclide %	Saturation Activity β/sec-gm	Activity per cc of Coolant for Σ_a of .022 cm^{-1}	γ_1 (Mev)	γ_2 (Mev)	γ_3 (Mev)	β (Mev)
Rhenium	75	186	90h	102	37	120×10^{10}	7.3×10^{7}	.137	.76	.63	
Indium (d)	49	114	49d	61	4.28	14×10^{10}	0.76×10^{7}	.72	.55		
Boron	5	12	.027s	.05	81	2.2×10^{10}	0.003×10^{7}				13
Oxygen	8	19	29s	2.2×10^{-4} (a)	.20	1.5×10^{5}	0.24×10^{7} (b)	1.6			
Sodium	11	24	14.8 hr	0.5	100	13×10^{10}	0.03×10^{7}	2.76			
Potassium	19	42	12.4 hr	1.0	6.7	$.98 \times 10^{10}$	$.005 \times 10^{7}$	1.5			
Lithium	3	8	.86s	.033	92.5	2.6×10^{10}	$.002 \times 10^{7}$				13
Hydrogen	1	3	12.5 yr	$.46 \times 10^{-3}$	.016	2.06×10^{10}	3.7×10^{7} (b)				.019
Sulfur	16	35	91d	.26	4.15	$.19 \times 10^{10}$	5.9×10^{4} (e)	.166			
Nitrogen	7	16	7s	2×10^{-5}	0.38	-	$\sim 1.1 \times 10^{6}$ (c)	6.13	7.10		10.3, 4.3, 4.8
Cadmium	48	107	6.7h	1.0	1.21	$.07 \times 10^{10}$	$.21 \times 10^{7}$	.85			
		111	49m	0.2	12.8	$.01 \times 10^{10}$	$.03 \times 10^{7}$	.149			
		115	43d	0.14	29	0.21×10^{10}	$.03 \times 10^{7}$	.5			
		115	2.3d	1.1	29	1.7×10^{10}	5×10^{7}	2.4			
		117	2.8 h	1.4	7.6	0.55×10^{10}	1.60×10^{7}				1.7
Gold	79	198	2.7d	96	100	203×10^{10}	3.6×10^{11}	.4.			.95
Silver	47	108	2.3m	30	51	87×10^{10}	1.4×10^{11}	1.4	.94		2.8
		110	24s	100	49	257×10^{10}	4.2×10^{11}	.7			
		110	270d	2.3	49	6.2×10^{10}	0.1×10^{11}	1.5			
Iridium	77	192	1.5m	260	39	316×10^{10}	1.2×10^{11}	0.2			
		194	19h	130	62	249×10^{10}	1.0×10^{11}	1.4			
Rhodium	45	104	4.4m	12	100	69×10^{10}	4.2×10^{10}	.08			.03
		104	42s	137	100	800×10^{10}	4.6×10^{10}				2.5
Helium 3	1	3	12.5 yr	5×10^{3}	1.2×10^{-4}	2×10^{10}	0.1×10^{7}				.019
Gadolinium	64	153	240d	125	0.21	1×10^{10}	$.38 \times 10^{7}$	0.1			
		159	18 hr	1.1	25	1×10^{10}	$.38 \times 10^{7}$	.35			.85
		161	3.6m	0.17	22	0.14×10^{10}	$.05 \times 10^{7}$	.37			
Samarium	62	153	47h	6	22.5	5.3×10^{10}	7.3×10^{10}	.27			
		155	23h	1.1	0.90	$.053 \times 10^{10}$	7.3×10^{8}	.3			2.0
Mercury	80	197	64 hr	3.1×10^{3}	0.15	0.4×10^{10}	20×10^{7}	.06			
		199	43m	60	23.3	42×10^{10}	20×10^{9}	.37			
		203	47d	2.4	29	2.1×10^{10}	1×10^{9}	.28			
		205	5.6m	.34	6.7	$.067 \times 10^{10}$	3×10^{7}				1.6
Hafnium	72	181	46d	10	35	11.7×10^{10}	1.7×10^{7}	.13			.46

Notes:

(a) Based on S at in $CdSO_4$

(b) Figures on Coolant at 100% H_2O

(c) N^{16} by (n, p) on O^{16} (for 100% H_2O)

(e) Indium 116

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B. Radiation Chemistry

Table 7 lists some of the kinds of radiation chemistry problems that might occur for any material used as a chemical control poison. In the body of the table are given guesses as to the probable result under each problem for the poisons considered.

Since there exists data in this field only for H_3BO_3 and $LiBO_2$, it is clear that experimental work is required. Substances must be tested with regard to the problems listed in Table 7 before they can be passed upon with regard to usefulness in water in a radiation field. Hence an extensive experimental program, using mocked-up reactor radiations and, insofar as possible, the conditions and materials of the practical application should be undertaken. A part of the program is underway at present.

TABLE 7 - RADIATION PROBLEMS

Material	Radiation Stability	Increased Water Decomposition	Effect on Corrosion	pption of solute?
Rhenium (salts)		Probable	High Ox pot.	
Indium Nickel (salts)	Unknown			
Boric Acid	Good at low concentration	Possibly good at low concentration	Increases due to complexing?	Improbable
Li Borate	Probably	Good	Inhibitor Probably	Possibly
Na Borate	Similar to Li Borate			
K Borate	ditto			
Li(OH)	Good	Probable	Inhibitor	No
Sulfate	Fair			
Nitrate	Poor (make up required)			
Li_2CO_3	Poor (ditto)			
Boron Hydrides	Poor			
BF_3 gas	Unknown			
$Cd SO_4$	Unknown	Probable	Deleterious?	Probable
Cd Complexes	Unknown	Probable		Exception - acid soln.
Gold Metal				
Silver Metal				
Iridium				
Rhodium				
Helium^3	Excellent	None	None	None
Gadolinium) Salts Samarium)	Unknown	Probable	Unknown	Possible
Mercury (dispersed in H_2O)				
Hafnium (Boron hydride or salt in bi phenyl)	Unknown	None	Probably none	Unknown

Notes:
Blank space - unknown and/or presumably unimportant in any case.
Unknown - not known but presumably important for design, i.e., information required.
Probable - similar cases (analogy) give result indicated.
Good - little or no effect expected but information generally not available.
Excellent - information available indicates no bad effect.
Poor - information available indicates bad effect.
? - Guess
Possible - possible in theory

Aggregation of Colloid	Change in Valence*	Change in Chem.Type	Effect on Crud	Susceptible to free radicals?	Slurry effect of radiation	Thermal Stability
	Probable	Unknown	Increase?			Good
	Probable			Yes		
	Possible		May effect deposition			Good
		Probable Li disappears	"			Good
			Inhibitor			Good
	Probable	Probable				Poor
						Good
	Probable	Probable	Increase	Unknown	Unknown	Fair
	Probable	Probable		Unknown		Poor
Probable				Unknown		
Probable				Unknown		
Probable				Unknown		
Probable				Unknown		
None	No	No	None	No		Excellent
						Good
Possible					Poor?	
						Probably Good

*steady state Ox-Red

V METHODS OF CONCENTRATION CONTROL

The means used for controlling the concentration of poison in the reactor will depend on the poison and the physical system chosen. However, we can make the following point in general with regard to the percentage change in poison concentration required to bring about a given percentage change in the multiplication constant of the reactor.

According to the simple theory for a homogeneous reactor, the variation of the multiplication constant of the reactor, k_{eff}, with poison concentration, Σ_p (cm^2/cm^3 of core volume), can be written as

$$k_{eff} = \frac{a}{b + \Sigma_p} \quad , \quad k_{eff} \sim 1 \qquad (1)$$

where a and b are constants roughly independent of Σ_p if the poison atoms have no important resonance peaks between 2 Mev and thermal energies. We are neglecting here any change in the moderating properties of the coolant as a result of its poison content. This will be permissible if the concentration does not exceed one volume percent. We can then write for the ratio of a fractional change in Σ_p to the resulting fractional change in k_{eff},

$$\frac{d\ln \Sigma_p}{d\ln k_{eff}} = \frac{-(\Sigma_p + b)}{\Sigma_p} \qquad (2)$$

We can get an approximate idea as to the order of magnitude of this ratio if we insert numbers based on STR in the clean, cold condition with a loading of 18 kg U. The constant b is 0.053 cm^{-1} in this case and Σ_p is 0.018 cm^{-1} for k_{eff} equal to one, giving

$$\frac{d\ln \Sigma_p}{d\ln k_{eff}} = -3.95 \text{ for } k_{eff} = 1.$$

This means that to increase the k_{eff} by a given percentage we must decrease the poison concentration by four times that percentage. It also can be seen that if the Σp required to hold the reactor in the critical condition is decreased, the percentage change in poison concentration required to bring about a given change in reactivity will be increased.

A. Aqueous Solutions

A number of the possible chemical control systems involve the use of aqueous solutions of non-volatile solutes. Consequently, we will consider these in some detail, particularly with regard to coolant-water poison systems. Among the standard operations used to change concentration in aqueous solutions, we may list the following: dilution ("feed and bleed"), evaporation, electrodialysis, ion exchange, electrolysis, crystallization and precipitation.

1. Rates of concentration change required

Each of the above methods has the characteristic that the percentage reduction of poison concentration per unit time is directly proportional to that rate at which poison solution is withdrawn from the circulating system. That is to say, processing a given fraction of the system volume per unit time will reduce the poison concentration by the same fractional rate. Inasmuch as it is generally more difficult to reduce the poison concentration than to increase it, we will consider first the rates at which it may be required to reduce poison concentration. The situation which requires the maximum rate in this case is that of over-riding xenon. We can estimate the rate by requiring that we hold k_{eff} equal to one while removing poison to compensate for xenon formation. Then

$$1 = \frac{a}{b + \Sigma_x + \Sigma_c} \qquad (3)$$

where Σ_x is the macroscopic xenon cross-section, Σ_c is the macroscopic cross-section of controllable poison, a and b are constants and $\Sigma_x + \Sigma_c = \Sigma_p$, where Σ_p is the total macroscopic cross-section of poison required for criticality. Thus we can write

$$-\frac{d\ln \Sigma_c}{dt} = \frac{1}{\Sigma_c}\frac{d\Sigma_x}{dt} = \frac{1}{(\Sigma_p - \Sigma_x)}\frac{d\Sigma_x}{dt} \qquad (4)$$

for the fractional rate at which Σ_c must be decreased to hold $k_{eff} = 1$. If we compute Σ_x and $d\Sigma_x/dt$ as functions of time for the period during which Σ_x is increasing, we can plot curves of $-d\ln\Sigma_c/dt$ against time for various values of Σ_p. A set of such curves, based on STR parameters, is given in Figure 1. Curve A is for the new reactor when $\Sigma_p - \Sigma_x$ (max) is at its largest. Curve E is for the end of life when $\Sigma_p - \Sigma_x$ (max) is zero, and curves B, C and D are for the cases when $\Sigma_p - \Sigma_x$ (max) is 1/2, 1/4 and 1/8 its value for the new reactor. It can be seen that the percentage change in Σ_c per second (proportional to the rate of withdrawal of solution from the circulating system) goes to infinity as the concentration of controllable poison goes to zero. (Actually, the maximum rate necessary for complete removal of poison is equal to the flow rate in the primary system.) Evidently then, for any rate of withdrawal at less than full flow of primary coolant, there must be a certain amount of poison left in the system when the xenon concentration reaches its maximum. How much poison is left, and how much additional uranium is necessary to compensate for it (for a given useful life of the reactor) will depend on the volume of the system, the rate of withdrawal of poison

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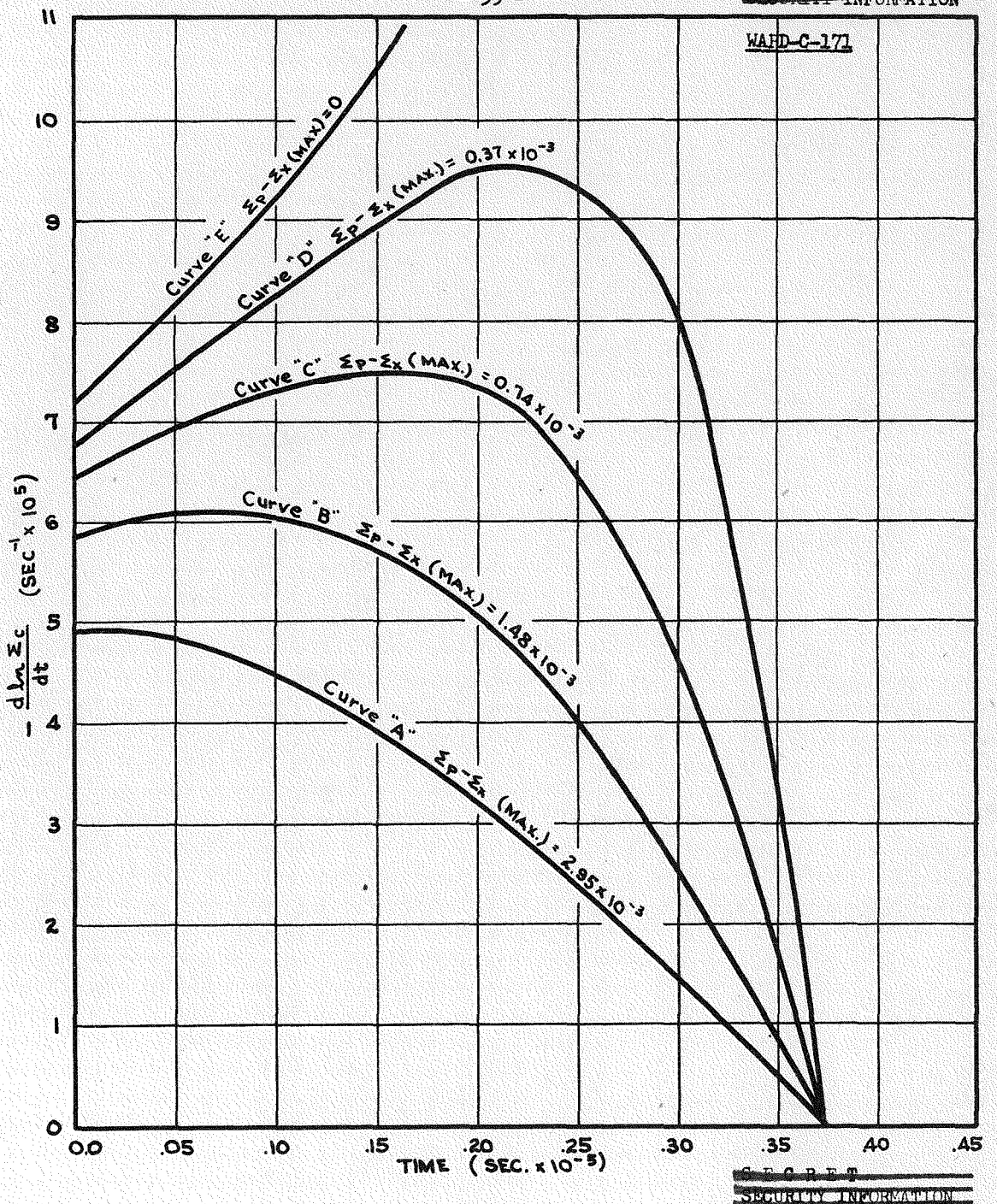

FIG. 1
RATE OF REMOVAL OF CONTROLLABLE POISON NECESSARY TO COMPENSATE FOR XENON FORMATION
(100% REDUCTION FROM STR FULL POWER)

solution which can be achieved and the way with which the ε_p necessary for criticality varies with time during operation of the reactor.

It should be emphasized that the curves given in Figure 1 are independent of the particular poison used, so long as the solution is dilute enough so that its moderating properties are essentially that of pure water. The results are also applicable to either auxiliary or coolant-water poisoning systems. As a numerical example, suppose we can achieve a flow rate of 20 gpm as a practical limit. Then, with the STR volume of 457 ft^3, the maximum rate of percentage change of poison concentration will be 9.8×10^{-3} percent per second. Then, by Figure 1, we see that for a given loading of U-235 there will be no periods during which the reactor will be inoperable until slightly more than about 7/8 of the excess reactivity associated with length of life has been consumed. For a given lifetime, the amount of U-235 would have to be increased. Just how much extra uranium would be needed depends on the core design.

2. Methods of operation

A generalized schematic diagram of a concentration-control system is shown in Figure 2. A part of a coolant stream is withdrawn from the main loop through a at the same time as an equivalent volume of solution of different concentration is injected from b. The sizes of the storage tanks and other equipment of Figure 2 will depend on the rate and type of concentration process chosen. In the "feed and bleed" process the concentrator is not necessary. For another process, if the concentrator is able to handle the full stream from a, the hold-up tank W is not required and the sizes of tanks C and D are greatly decreased as the

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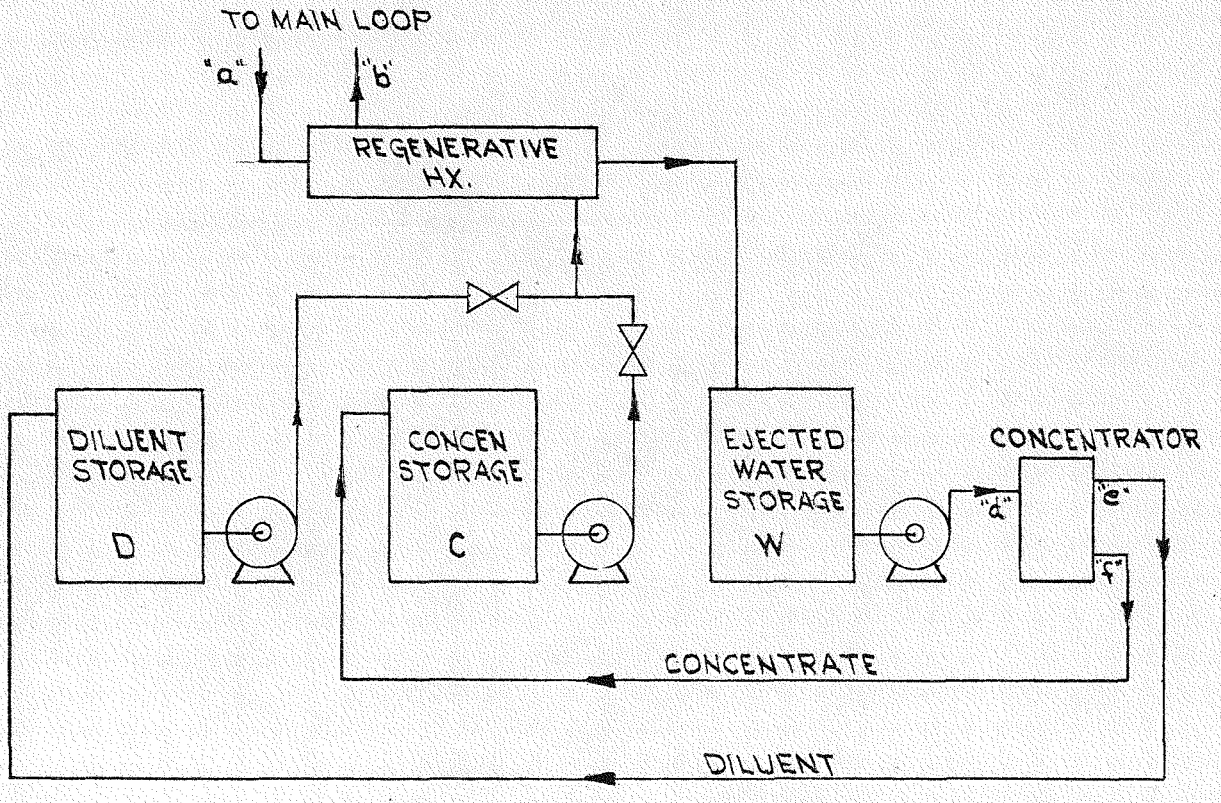

FIG. 2
POISON LOOP SCHEMATIC FLOW DIAGRAM

stream to b need be only partly drawn from storage. In evaporation, if the concentrate is held in the evaporator, the concentrator and tank C are the same piece of equipment.

The time variation of poison concentration in the reactor will depend on the points of the main loop where the connections a and b are made and on the relative cycling periods for liquid in the main loop and in the poison loop. For instance, if the time required for solution to pass through the concentrating equipment is long compared with the time required for the coolant to make a complete circuit through the main loop, the poison concentration in the reactor will vary in a step-wise fashion if the flow from the poison loop is started in a time short compared with the main loop cycling period. Figure 3 illustrates this argument. If it is desired to avoid step-wise changes, it is necessary to increase the flow from the poison system during a time equal to or greater than the main loop cycling period. Step-wise changes can be tolerated if the size of the step is such that the period with which the flux builds up as a result of the change is about 20 times the main loop cycling period. Then the change of flux with time will be essentially uniform.

(a) Probably the simplest method of changing the poison concentration is to add concentrated solution or diluent water from make-up tanks while ejecting an equal volume of solution of the concentration existing in the coolant loop. If the poison is relatively inexpensive, there is no point in concentrating the ejected solution, except that it possibly may be desirable to reduce its volume for convenience in storage or disposal.

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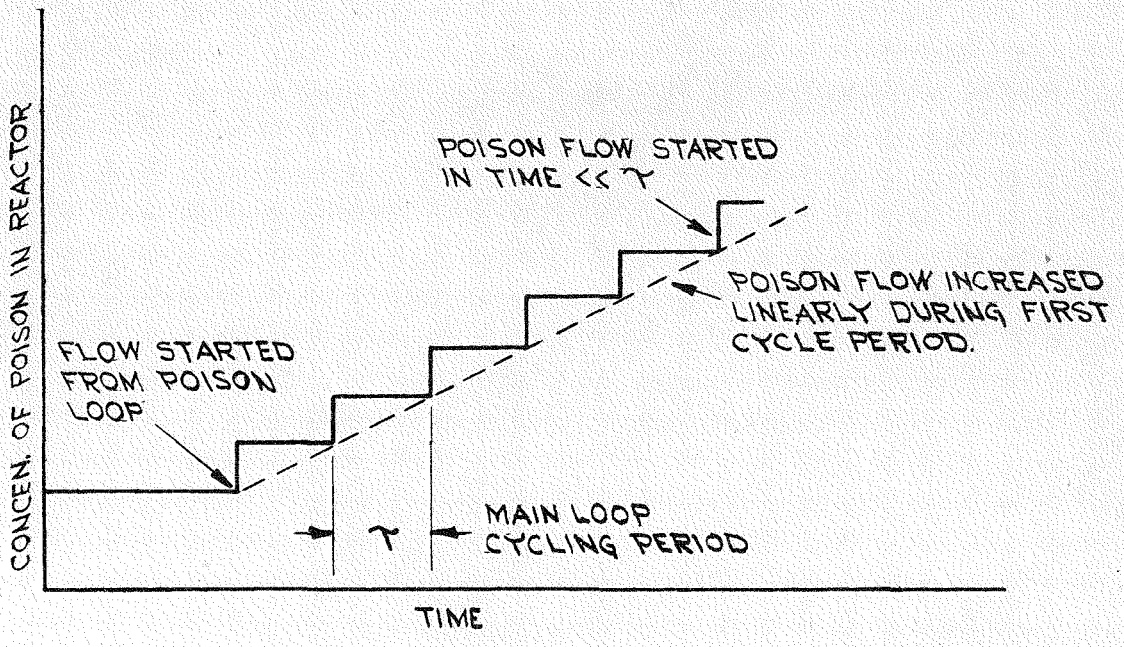


FIG. 3
POISON CONC. IN REACTOR VS TIME
(POISON LOOP CYCLE PERIOD >> MAIN LOOP CYCLE PERIOD)

We shall call this type of operation "feed and bleed". The method requires a storage tank for make-up water, a source of water, a tank for making up concentrated solutions of poison, metering pumps for injecting and ejecting solutions at a controllable rate and a regenerative heat exchanger for recovery of heat from the ejected solution. Storage capacity and a source of pure water are no problem for a land-based power plant. A sea-going plant must be able to make its own pure water from sea water. It is suggested that the Ionics, Inc. electrodialyzer, to be discussed later, be used to provide additional capacity for the fresh-water plant. This device should be satisfactory if used in an accessible location so that maintenance can be performed on it. The storage requirement for pure water will be a function of the pure-water recovery rate. Storage requirements for poison solution should not be too great, as the saturation concentration of the solution will be many times the highest concentrations employed in the loop. Solid chemicals for make-up can be stored dry in a small volume, the total quantities involved being quite small.

The power required for injecting 20 gpm against 2000 psi is 23.3 HP or 17.4 KW, assuming 100% efficiency. One possible method for injecting the poison would be the use of a pair of two-stage rotary gear pumps or a pair of displacement pumps, poison being added to the system with one pump while being withdrawn from the system through the other. If the shafts could be coupled together, the second pump could be used as a hydraulic motor to help drive

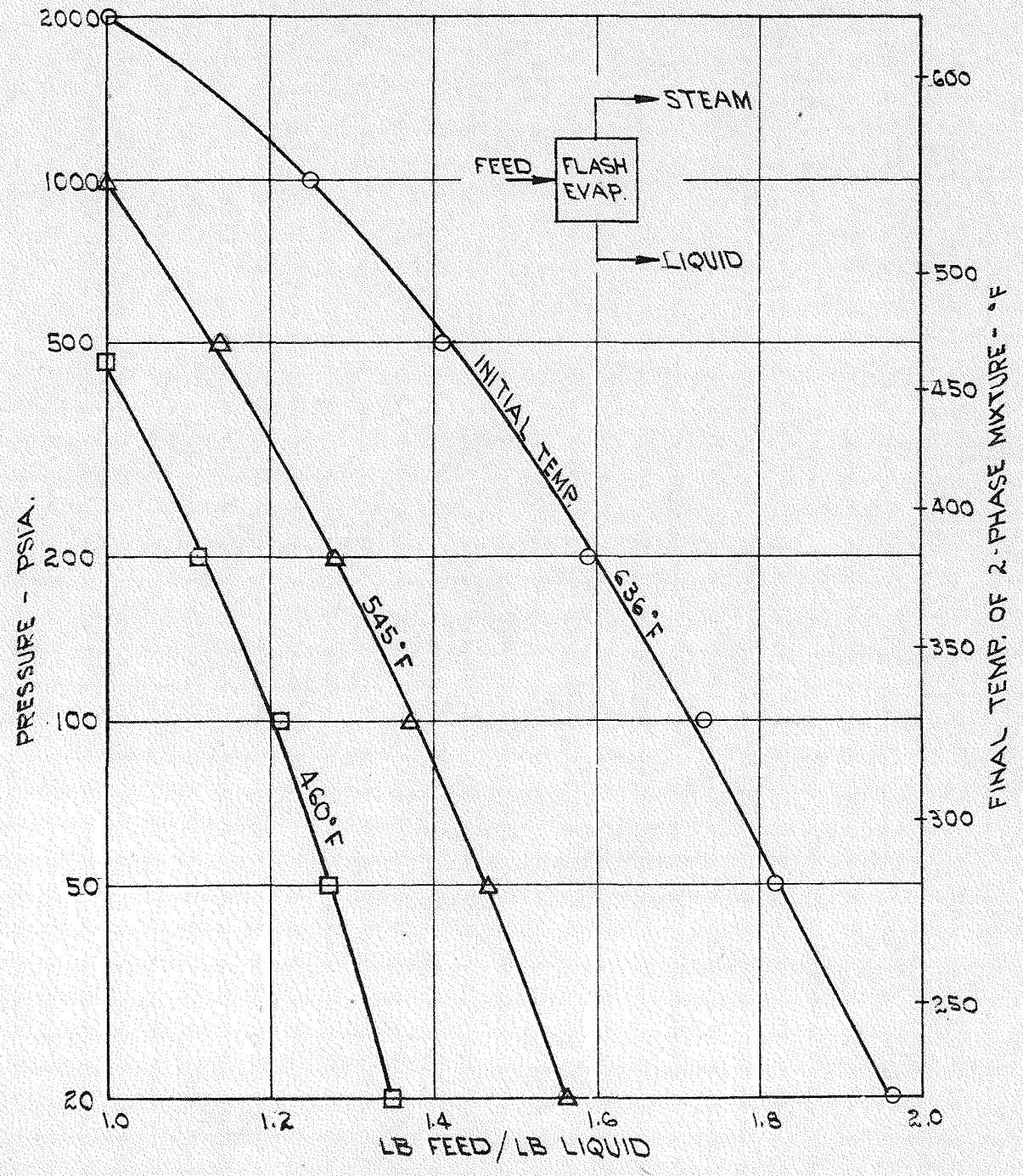
the first and to insure that the volume of solution in the coolant system remains constant. Additional power would be required only to make up for the inefficiency of the pumps. Counting both recovery of power by the hydraulic motor and the efficiency of the pumps, driving power probably would not exceed the figure quoted above for 20 gpm at 100% efficiency. The pumping and metering system would fail as-is. It would be operated at low temperature.

A liquid-liquid heat exchanger for cooling the ejected poison solution would be expected to recover at least 80% of the heat content of the effluent stream. On this basis, the power loss would be 212 KW for 20 gpm. Since this 212 KW is low-grade heat power and is required only intermittently, this figure is comparable with the approximately 20 KW of electrical power required continuously by a full set of control rods.

(b) Evaporation is one of the most widely used methods of concentrating aqueous solutions. It can be done at constant pressure or by flash expansion at constant enthalpy. The second of the two methods can be made much more rapid than the first, the rate of evaporation being limited only by the area provided for transport of vapor from the liquid phase. In principle, however, flash evaporation will require more power, as the expansion is an irreversible one. In Figure 4 are plotted the saturation temperature and pressure of water against the ratio of the weight of water fed to a flash evaporator to the weight of liquid phase remaining. (The elevation of boiling point by dissolved material has been neglected, but we will be dealing only with quite dilute solutions,

Fig. 4

FLASH EVAPORATION OF PURE WATER FROM 2000 PSIA AND SEVERAL INITIAL TEMP.



so the arguments here are approximately correct.) It can be seen that, if the initial temperature of the water is 636°F, the concentration can be increased by a factor of 1.4 before the temperature of the liquid has fallen below 460°F. Thus, one method of operation could be to take the feed to the evaporator from the system pressurizer, maintaining the temperature of the flash chamber at that of the coolant. The vapor could be condensed by isothermal compression. The stream which is to be fed back to the coolant would already be at system temperature, but it would have to be injected against a considerable pressure head. The power for this process would be supplied by the pressurizer heaters and the injection pumps.

High pressure evaporation has the advantage that the heat of vaporization per pound falls off rapidly as one approaches the critical point. Evaporation at constant high pressure could be carried out in the part of the apparatus which functions as the degasifier, pressurizer and surge tank. An abbreviated diagram of a pressurized water reactor, including the degasifier loop, is shown in Figure 5. The similarities between the degasifier loop and the poison concentrating scheme shown in Figure 2 are apparent. If the direction of flow through the degasifier is reversed, so that vapor passing from the liquid phase is condensed externally, the liquid would become more concentrated in dissolved solids, thus removing the material from the main loop. The latent heat of vaporization could be used to furnish the heat losses from the equipment by passing the vapor through a jacket surrounding the evaporator

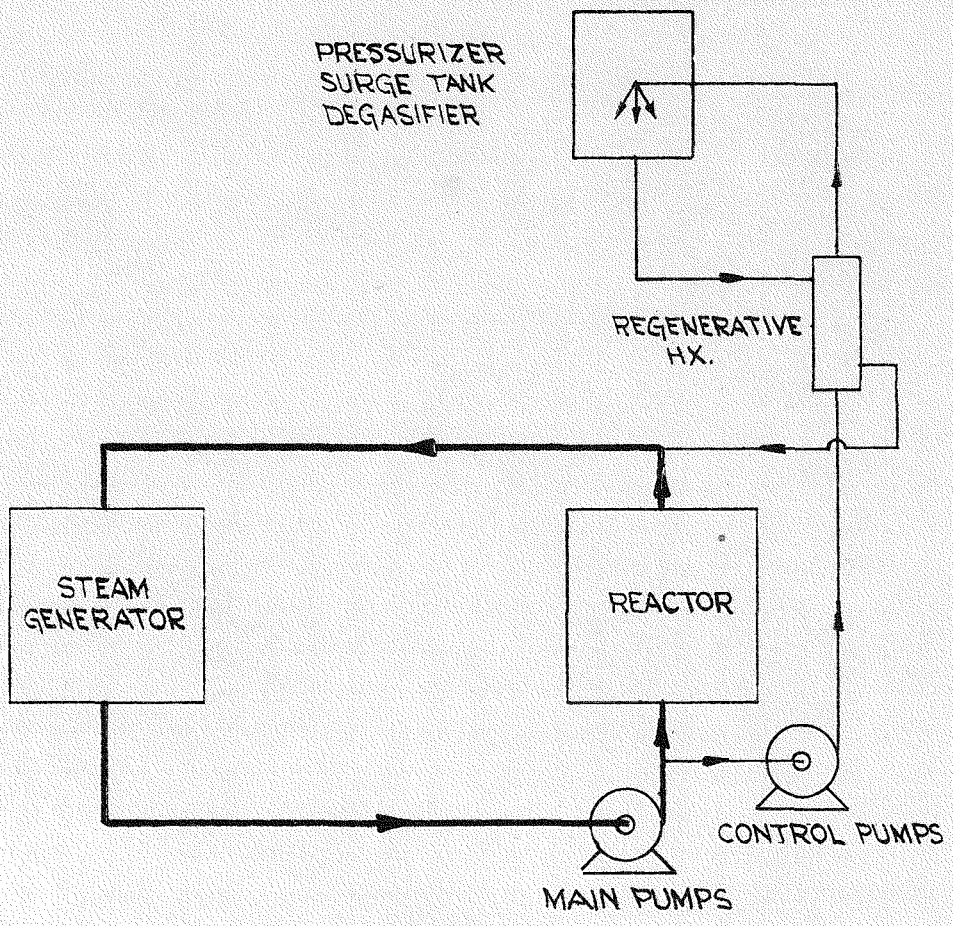


FIG. 5
STR MAIN LOOP & PRESSURIZER LOOP DIAGRAM

and through the shell side of the regenerative heat exchanger. The hot condensate and the remaining uncondensed vapor would be bled into the hot leg of the main coolant stream for ultimate recovery of the heat input to the evaporator. The increased power consumption over the present degasifying unit would be the result of degradation of that part of the electrical power input to the evaporator heaters which is recovered as heat power in the coolant stream. A disadvantage of this method is that surges from the tank might cause mixing of concentrated poison with the more dilute main stream unless adequate volume is provided in the surge line. To use the device as a degasifier during reactor operation at constant power level, it would be necessary to mix the condensate stream with solution drawn from the liquid phase in the tank so that the concentration of the solution fed back to the coolant stream would be equal to the concentration existing in the coolant. The potential advantages of the method are the combining of functions with existing equipment, the impossibility of withdrawing poison from the reactor at an unsafe rate, and the fact that sufficient poison to shut down the reactor is actually in the system and can be added rapidly to the main body of coolant water.

(c) A concentration device has been developed by Ionics, Inc. using their electro-dialysis apparatus. Studies have been made on the transfer of lithium borate from dilute to concentrated solutions as a function of the lithium to borate ratio of the solution, concentration of solutions, temperature, and method of recirculation and flow rate. The method has the disadvantages that the apparatus

must be kept at a low temperature and that the lower limit of the volume of the concentrate storage tank is set by the properties of the dialyzing membranes, which limit the concentration that can be achieved. The reliability of the equipment, particularly of the experimental dialyzing membranes, is an unknown factor. The device should not be used in an inaccessible location without first undergoing an extensive series of tests using actual plant conditions and crud-containing water. The advantages of the device are relatively low size and power requirements and considerable ease of control.

(d) Ion exchangers do not look promising in view of their need for regeneration or replacement (limited capacity), the requirement that they be kept at low temperatures, and the low velocity of liquid which must be maintained in the exchanger. STR would require a minimum of 50 ft^3 of resin for each over-riding of maximum xenon.

(e) Crystallization methods are also not considered to be too attractive, particularly because there exists the possibility that supersaturation may occur. The rate of growth of crystals is slow at best, solutions sometimes requiring days or weeks to reach their equilibrium solubility after having been cooled from a temperature at which they were not saturated.

(f) Electrolytic and precipitation methods will depend specifically on the chemistry of the substances used as poisons. There are very few possibilities as precipitating agents suitable for borate poisons, cadmium and barium being the most likely. Cadmium poisons could be precipitated as CdS even if present as complex ammines or cyanides. The precipitate would have to be

removed by a filter. The solubility and tendency toward colloid formation of the precipitates at operating temperature must be known before such methods can be evaluated.

(g) Only inert gaseous poisons are compatible with water. The degasifying apparatus could be used conveniently to control the concentration of gases soluble in the coolant water.

B. Other Systems

1. Suspensions, slurries and other coarsely disperse systems can be separated quite easily by filtration, settling or centrifugation. Filtration is probably the simplest and most workable method and is the only method that can be used when there is little difference in density between the poison and its carrier.

2. The methods of changing concentration in gas-lift and other fluidized-solid systems are inherent in the system and do not need separate discussion here.

3. Auxiliary systems, in general, present few difficulties, as the volume to be processed will be quite small. This means that the rate of change of concentration can be made more rapid than in coolant-water poisoning systems, and it is practical to consider chemical scram and recovery from scram in addition to the other control functions. Electrolytic methods are quite feasible for amalgams and molten salt or alloy systems. Evaporation is practical for any aqueous solution. Non-aqueous solutions, such as diphenyl boric acid in biphenyl, can be distilled with low power requirements. If gases are used in an auxiliary system, control of density by varying the pressure is the most convenient method.

VI CONCLUSIONS AND RECOMMENDATIONS

The potential feasibility of chemical control of nuclear reactors is indicated by the following facts:

(a) The neutron capture cross-section of many chemicals is sufficiently high that reasonable amounts within the core will control the excess reactivity of an STR-type core.

(b) Several physical methods are proposed which will provide a fluid system within the core in which the nuclear poison concentration can be adjusted automatically. Both the primary coolant system and an auxiliary system have been discussed as suitable to accommodate the poison.

(c) An adjustable concentration can be achieved with well-known methods of the chemical industry.

(d) The radiation hazards which require shielding and delayed access for personnel protection have been discussed. A few nuclear poisons present no hazard greater than that already existing due to the neutron bombardment of water.

(e) Proposed system materials are chemically compatible with some nuclear poisons.

(f) Safe failure of chemical control is inherent in several of the possible methods of application.

The development problems related to chemical control consist chiefly of the following:

(a) Thermal expansion of most chemical control fluids opposes the self-regulating effect of the negative temperature coefficient of reactivity of a given core. The cross section of cadmium increases uniquely with

core temperature to offset this disadvantage.

(b) Peak power requirements in adjusting chemical control may exceed the power requirements of mechanical control.

(c) Some desirable nuclear poisons are chemically incompatible with proposed system materials.

(d) Some nuclear poisons undergo rapid and undesirable changes in the neutron field, and some poisons increase the rate of dissociation of water.

(e) The use of chemical control may require extra fissionable material if (a), above, cannot otherwise be accommodated by core design alone.

(f) Recovery from chemical scram may be slower than with mechanical scram.

(g) Difficult core design problems may be created if both mechanical scram and auxiliary system chemical control are required.

(h) The problem of maintaining the coolant water free of corrosion products and fission products to minimize shielding and access delay may be complicated by the use of a nuclear poison dissolved in the primary coolant.

These serious problems are not insurmountable, and the attendant advantages of chemical control are not beyond practicality. It is therefore concluded that the development of chemical control should proceed with every reasonable effort.

937 053

The arguments and data contained in the body of this report point to lithium borate, dissolved in the primary coolant, as the chemical control method which is most promising for both PWR and SFR.

The unique advantages associated with the use of lithium borate are that no extra shielding or additional access delay is required.

Improved flux distribution, simplified core design and safe failure are unique advantages associated with the use of the primary coolant as the vehicle for the poison.

The one major disadvantage associated with this method is the necessity for changing the core to a less optimum configuration in order to preserve a desirable temperature coefficient of reactivity. Such a change in core design would probably require a greater loading of uranium.

Cadmium is the one material which does not suffer from the above disadvantage, since its microscopic cross-section increases with temperature as a result of a resonance peak at about 0.17 ev. This increased cross-section is just about sufficient to offset the expansion of water, with a net result of no change in the reactor temperature-coefficient. Cadmium, however, suffers from the major disadvantages that most of its water-soluble salts hydrolyze to an acid solution and are not compatible with stainless steel, and additional shielding and access delay are required. Nevertheless, there does exist the possibility that sufficiently stable cadmium compounds may be found which are applicable to a coolant-water poisoning system. Alternate materials later might be developed which are compatible with cadmium solutions.

For PWR the additional shielding would be inexpensive, but for SFR the additional mass would be objectionable. Furthermore, access delay would be more serious for SFR than for PWR.

937 054

An auxiliary poison system for cadmium would minimize the mass of shielding required, and would increase the access time of a smaller portion of the reactor plant. Materials which are compatible with cadmium compounds can be used for an auxiliary system without affecting those of the primary system. Heterogeneous mixtures can also be accommodated by appropriate design.

An auxiliary system is not inherently fail-safe, since control can be lost through an external rupture. However, with proper design and application, an auxiliary system may be made to fail safely.

Faster response to control demands is possible with the smaller volume of the auxiliary system. Chemical scram is possible, thereby avoiding the complicated core design which otherwise would result from the simultaneous application of auxiliary system chemical control and mechanical rod scram.

Moderator control for nuclear reactors has the unique advantages of higher neutron economy which results from avoiding the use of a nuclear poison and the elimination of the problem of extra core loading.

On the basis of the foregoing discussions, the Chemical Control Task Force submits the following recommendations:

(1) Lithium borate in the primary coolant should be developed as a chemical control method for PWR and for SFR.

(2) In the interests of economy of fissionable material, a parallel effort should be made on the development of a method employing cadmium.

(3) Research on the use of an auxiliary system is proposed to achieve faster control response.

(4) The possible applications of moderator control are recommended for further study on the basis of enhanced neutron economy.

Although in making the above recommendations, it was recognized that the development of a reliable method in time for its application to PWR and SFR should take precedence over the development of a potentially more ideal method which may not meet the schedule, the same recommendations would have been made in the absence of such a restriction.

QUANTITY OF POISON FOR CRITICALITY*

Element	G.A./1000cc Σp=.022 (1)	G.A./1000cc Σp=.0108(2)	g/1000cc Σp=.022(1)	g/1000cc Σp=.0108(2)	G.A./1000cc per sec. (3)	lb/457 cu ft per sec. (3)
Ag	1.50	.975	161.8	105.2	244. x 10^{-6}	.750
Au	.957	.619	188.7	122.1	155.	.871
B	.12	.078	1.30	.844	19.5	.006
B^{10}	.0225	.0147	.225	.147	3.67	.001
Cd	.0268	.0108	3.01	1.21	2.69	.009
Dy	.081	.0534	13.16	8.68	13.4	.062
Er	.54	.351	90.29	58.69	87.8	.419
Eu	.0199	.013	3.02	1.98	3.25	.0141
Gd	.002	.00133	.314	.209	.33	.0015
He^3	.0173	.0112	.0519	.0336	2.81	.00024
Hf	.778	.507	138.95	90.55	127.	.647
Hg	.236	.154	47.34	30.9	38.5	.22
In	.473	.307	54.28	35.23	76.8	.251
Ir	.204	.133	39.39	25.68	33.3	.183
Kr^{83}	.439	.285	36.44	23.66	71.2	.168
Li	1.28	.829	8.86	5.74	207.	.041
Li^6	.0986	.0643	.592	.386	16.1	.0028
Lu	.827	.542	144.7	94.85	135.	.673
Re	1.07	.696	199.4	129.7	174.	.924
Rh	.597	.39	61.44	40.14	97.5	.285
Sm	.0093	.0048	1.4	.722	.12	.0051
Tm	.764	.494	129.4	83.68	123.	.593
Xe^{131}	.749	.488	98.1	63.93	122.	.455

(1) Pile cold; 20 kg undepleted fuel; all rods out.
(2) Pile 460°F; 20 kg undepleted fuel; all rods out.
(3) Gram atoms poison per 1000 cc per second and lb poison per second for Mark I to compensate for Xe burnup during startup at maximum Xe concentration. (Temperature 460°F)

*This information appeared as Table III in WAPD-C-159, Chemical Control, by Paul E. Brown.