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ORGANIC NUCLEAR REACTORS:

AN EVALUATION OF CURRENT DEVELOPMENT PROGRAMS

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ORGANIC NUCLEAR REACTORS: AN EVALUATION OF CURRENT DEVELOPMENT PROGRAMS

I. SCOPE AND PURPOSE

Following a request on January 25, 1961, of the Chicago Operations Office of the Atomic Energy Commission for an organic reactor study, a committee was formed of staff members of Argonne National Laboratory to review the technology of organic nuclear reactors. The Committee was requested to prepare, within a short time, a report with the following two principal objectives:

- to provide a critical evaluation of organic reactor technology, and
- to provide recommendations of areas of research and development work now lacking or inadequate for the successful development of this reactor concept.

The Committee commenced its work immediately by collecting and reviewing reports in the field and by visits to and discussions with organizations concerned.

Underlying assumptions in the study were that organic reactors are indeed feasible, and that both economically and practically the general concept is sufficiently attractive to justify an extensive development program. Consequently, principal attention was directed toward understanding the possible applications of the concept, determining the problem areas, and estimating the significance of work accomplished, under way, and planned. It was not attempted to evaluate the worth of the organic reactor concept either per se or relative to other types of reactors (i.e., to water or other reactor types); neither was it attempted to compare advantages and disadvantages or power costs of organic reactors with other reactors. Similarly, in making recommendations for further work, it will not be implied that such work should be undertaken in preference to work on other reactor concepts.

For the purpose of this survey, an organic reactor was broadly defined as any nuclear reactor which utilizes an organic chemical compound or mixture of such compounds, primarily in the liquid state, to convert usefully the energy of the nuclear reaction. Included under this definition are rigid and fluidized solid-fueled, organic-cooled reactors, both boiling and pressurized liquid types, moderated with organic or inorganic materials, as well as nuclear reactors used to effect chemical reactions. All reactors in this group have significant characteristics in common relating to the effects of radiation on organic materials. Members of the study group were all scientists and engineers affiliated with Argonne National Laboratory. Although the members were generally representative of the technological areas associated with organic reactors, none had recently been closely associated with the development program relating to organic reactors, and most of the group has no previous formal contact with this subject. The basis for the group's review was primarily information obtained during the short period of the study. The opinions evolved in general represent the authors' jointly considered review.

II. SUMMARY

The status of the program aimed to develop useful and economic nuclear reactors cooled by means of organic liquids, such as the polyphenyls,was surveyed. The concepts considered included organic, heavy water, and graphite-moderated reactors producing power by direct or indirect cycles. Although the principal focus of U.S. effort is centered at Atomics International, Canada has embarked upon a major program leading to a heavy water-moderated reactor, and twelve other organizations in the U.S. are or have been significantly engaged in the program. A number of European groups are now actively considering or working toward the development of organic reactor concepts

Organic reactors are characterized by major and generally irreversible decomposition of the coolant. An upper limit to the temperature at which coolants can be used is less than 530°C. Heat transfer coefficients are generally below those applying to other systems. Nonetheless, organic reactors could utilize low cost materials for fuel and for system structures. In most applications, low system pressures, low coolant activation, a need for extensive coolant processing, and some advantageous nuclear safety characteristics are also significant. Organic reactor concepts appear to have a broad potential for development if adequate attention is given to the chemical nature of the systems. For the concepts presently emphasized, the principal problems are concerned with decomposition and reclamation of coolant, fouling, heat transfer limitations, fuel element materials stable at high temperatures, and demonstration of overall system performance.

Coolants may be selected from organic materials of adequate thermal stability on the basis of superior radiation stability, low cost, or easy regeneration. Although the mechanisms of protection from radiation damage are not well understood, aromatic hydrocarbons, particularly the polyphenyls, have been found to be most stable, as well as otherwise suitable as coolants. A number of other types of compounds could be used, but they have not been adequately studied. Changes in the properties of mixtures of terphenyls are not excessive under reactor conditions if part of the polymer is continually removed. Much more work is needed to find out what happens when organic compounds are irradiated. Some searches being made for cheaper coolants, and for additives useful in inhibiting decomposition or in overcoming its effects, could well be extended. Consideration should be given to coolant systems best adapted to other specific applications or to ready reclamation.

Operation of the Organic Moderated Reactor Experiment (OMRE) has shown that terphenyl mixtures can be used to cool a nuclear reactor for extended periods. The heat transfer characteristics are acceptable and are reasonably well understood for a moderate range of conditions in which forced convection is used. Much more information is needed to establish limitations and capabilities for reactors of high performance and for conditions adaptable to concepts other than the organic-moderated reactor. Fouling deposits, developed from particle contamination or otherwise, may limit performance.

Particles in the coolant may result from system corrosion. It appears essential to develop means to prevent their formation or to remove them. Tests of appropriate equipment for particle removal are under way, and the mechanisms of particle formation and deposition are being extensively investigated. The development of systems for removing water, gases, and polymeric materials from the coolant, and for disposing of wastes, has been carried out only to a limited extent. The performance of such equipment largely remains to be demonstrated. Laboratory experiments indicate that there is a good possibility of developing means of reclaiming a major fraction of the polymerized terphenyls now being discarded, but process feasibility and economic advantage are not yet demonstrated.

The two principal fuel elements being considered for reactors of moderate to high performance are aluminum-clad uranium-molybdenum alloys and UO₂ contained in high-strength aluminum (APM) tubes prepared by the techniques of powder metallurgy. Uranium alloys containing enough molybdenum to provide adequate high-temperature strength appear to be undesirably diluted with absorbing materials for neutron economy, require development of a satisfactory diffusion barrier, and have unknown burnup capability. The development of other potentially useful uranium alloys for use at high temperature appears to have been neglected. The technology of producing, forming, and fabricating satisfactory APM materials to contain UO_2 is as yet unreliable and undemonstrated, but this problem appears to be resolvable if the work now going on is continued, particularly if it is adequately coordinated. Suitable zirconium alloys may be developed to contain UO_2 . Carbon steels are generally satisfactory as construction materials for organic reactors, but corrosion rates, though low, are inadequately defined.

The neutron physics data and calculation methods required for the design of organic reactors and for system optimization closely resemble those needed for analogous water systems and generally appear to be adequate. Discrepancies in evaluation of fast neutron effects on coolant damage are now substantially resolved. Physics calculations for D_2O -moderated, organic-cooled reactors indicate the desirability of developing uranium alloys of low parasitic absorption for use as fuels.

Although considerable commercial background is available in the petroleum and chemical equipment industries for fabrication of components for organic reactor systems, the units required usually fall beyond manufacturers' normal experience in sizes, temperatures, cleanliness, and leaktightness. In most cases, however, equipment has been fabricated through extension of commercial practices.

Development work is required in the field of gasket, packing, and sealing materials, and in simplification of fuel-handling techniques. The use of compact heat exchangers should be investigated. Operating experience with organic reactors soon to be completed will probably reveal areas in which initial component specifications can be relaxed for future plants, and may demonstrate the possibility of increasing power ratings for given plants. Consequently, some reductions in unit power costs for future plants might be realized.

Nuclear safety aspects of well-designed organic-moderated reactors are generally favorable. Although somewhat different considerations apply to organic-cooled reactors moderated with heavy water or graphite, such reactors can be made safe through careful study of all factors involved. It has been demonstrated that under normal operating conditions the addition of water to an organic-moderated reactor does not result in dangerous nuclear or pressure excursions. Transient conditions which might result in void formation in pressurized cores are under study.

The safety of organic reactors is also affected by the flammability, volatility, and toxicity of the coolant. Additional data are needed to evaluate fully the effect of radiolysis products on these properties. No exothermic reactions between coolants and other reactor materials have been found.

Containment for organic reactors is under study. Pressure-holding containment does not appear necessary. Investigations are in progress to devise methods of making conventional buildings sufficiently gastight to contain organic reactor systems safely at low cost.

Current interest in the use of organic materials as working fluids in fossil-fueled power plants suggests the need for detailed evaluation of possible direct-cycle concepts with organic reactors. The extension of thermodynamic data for organic fluids, including mixtures containing products of radiolysis, should be accelerated to provide an adequate basis for such studies. Although organic-moderated fluidized-bed reactors proposed to date do not appear overly favorable, the possibilities of using slurries of powdered fuels (including metallic powders) in organic fluids should be studied closely. Comparison of cost data for a number of large-scale power reactor designs incorporating the use of organic coolants with similar data for fossil-fueled plants indicates that the potential of the organic-cooled reactor for achieving competitive power in moderate to high-cost areas is much dependent on attaining the projected high performance. This requires continued intensive advancement of the technology. The most significant areas for possible cost reduction appear to lie in cost of fuel, coolant makeup, and accessory equipment.

Development programs for present organic and heavy watermoderated concepts appear generally adequate to reach specific goals which have been set. However, the narrow scope of the present organic reactor program should be broadened to assure coverage of areas where the application of novel principles might result in marked economic benefits. Further work, principally of a basic nature, is recommended in the fields of chemistry, processing, management, and thermodynamic properties of coolants, in fuel development, and in concept evaluation.

III. ORGANIC REACTOR CONCEPTS AND SOURCES OF INFORMATION

A. <u>Reactor Concepts using Organic Liquids for Cooling, Moderating, or</u> Absorbing Radiation

Because organic reactors have received only a fraction of the attention of nuclear scientists and reactor designers up to this time, relatively few concepts have evolved. Initial consideration was limited to the use of an organic material as a source of moderating hydrogen in place of water, and accordingly concepts were adapted from those used for watermoderated reactors. The principal question was to determine if there were organic liquids, suitable for use as coolants, which could withstand a radiation environment of high intensity for a time significant in the operating life of a reactor. Once this problem had been resolved favorably for a particular group of organic compounds, more attention could be given to effective utilization of the peculiar characteristics of these and other suitable materials in nuclear reactors. This has led to broadening of the nature of the concepts considered for organic reactors beyond that which existed originally.

1. <u>Pressurized Organic Reactors</u> - The pressurized organic reactor represents a category of concepts analogous to pressurized water reactors. The organic liquid (coolant, moderator, or both) is maintained in a liquid state throughout the reactor and its associated heat transfer system. The reactor core is composed of rigidly fixed fuel elements. Depending on the nature of the coolant and on operating temperatures, system pressures may be low or high.

a. <u>Organic-moderated-and-cooled Reactors</u> - By far the largest amount of work has been done on reactors to be moderated and cooled by organic compounds. Advantages of simplicity and of economic construction are claimed in particular for this concept. It consists of a core of metallic or metal-clad fuel elements in fixed positions, designed to allow adequate neutron moderation by an organic coolant. The heated coolant, leaving the core below the temperature of bulk boiling at the system pressure, generates and may superheat steam in an external loop, and is then pumped back to the reactor vessel. This concept is associated with Atomics International, and is typified by the OMRE, (1) the Piqua prototype power reactor, (2) by Bechtel and AI commercial power designs, (3,4,5)U.S. (6) and German ship reactor studies, a process heat reactor study, (7)and the EOCR (8) and PRO (planned by CNEN) test reactors.

b. <u>Heavy Water-moderated</u>, Organic-cooled Reactors - Use of heavy water moderator allows the use of natural uranium as fuel. When coupled with an organic coolant radiation damage to the organic coolant is reduced by having it occupy only a very small fraction of the reactor core in narrow passages directly adjacent to the fuel elements. The heavy water is contained in an insulated calandria. Steam is generated and superheated in external loops. Use of an organic coolant of low volatility facilitates the use of pressure tubes for containing the organic coolant and fuel. This concept is exemplified by the Canadian OCDR design,⁽⁹⁾ Euratom's ORGEL project for the ESSOR test reactor, and the Danish DOR project.⁽¹⁰⁾

In a modification of this type, higher steam temperatures could be achieved by cooling a portion of the fuel with steam. (11)

c. <u>Graphite-moderated</u>, Organic-cooled Reactors - In one variation of a concept, involving the use of a graphite moderator and organic coolant, high-temperature fuel elements are inserted in graphite moderator blocks which are cooled by circulating organic liquid in narrow channels. This is part of a Marquardt concept, (12) the DCDR, which utilizes another technique for energy conversion. Graphite moderation coupled with organic cooling has also been studied by Hanford(13) and Brookhaven.

d. <u>Organic-cooled Reactors with Other Moderators</u> - In other studies, light water, beryllium, beryllium oxide, and zirconium hydride have been considered as moderators in conjunction with organic coolants.

2. Fluid-bed-fueled Organic-moderated-and-cooled Reactors - In this concept, high-temperature resistant fuel pellets (of UO_2 , UC, etc.), suitably clad to limit the escape of fission products, are suspended in the organic coolant-moderator by fluidizing with flowing coolant. Surface temperatures are reduced considerably by using a large ratio of fuel surface to fuel volume. Fuel density may be varied by adjusting the speed of the fluidizing medium; power regulation may be accomplished by this means, and fuel may be removed from and returned to the reactor in a stream of coolant. Work on concepts of this nature has been carried out by Westinghouse(14) and Martin.(15)

3. <u>Boiling Organic Reactors</u> - Boiling organic reactors, analogous to the various types of boiling water reactors, can readily be conceived. Many organic vapors superheat upon expansion, so that nuclear superheating may not be needed. Organic compounds generally have critical pressures less than $\frac{1}{3}$ that of water, and consequently such reactors will operate at lower pressures unless supercritical conditions are maintained. Since organic compounds are available with widely varying volatilities at a given temperature, a wide pressure range is therefore available for selection. Organic vapors can presumably be used in direct-cycle systems, (16) although relatively little is known about their behavior in turbines. It is understood that Westinghouse and G.E. are planning tests of this nature now.

a. <u>Rigidly Fueled Organic-moderated-and-cooled Boiling</u> <u>Reactor</u> - This concept would involve bulk boiling of organic coolantmoderator and use of the vapor either indirectly to produce steam, or directly in a turbine. So far as is known, little consideration has yet been given to a direct cycle reactor of this type, except for a Russian study.⁽¹⁷⁾ b. <u>Rigidly Fueled, Graphite-moderated, Organic-cooled Boiling</u> <u>Reactor</u> - In a concept proposed by Marquardt,⁽¹²⁾ only local boiling takes place in the reactor core, and a fraction of the circulating coolant is flashed outside the core to yield vapor which is passed to a turbine, while the bulk of the coolant is returned as liquid from the flash chamber to the core. In the core, the graphite moderator placed between the fuel and coolant reduces coolant radiation damage and permits higher fuel temperatures.

c. <u>Boiling Organic Slurry Reactor</u> - It has been conceived that boiling organic vapors, generated by a slurried fuel, might be used in direct or indirect power cycles, similarly to concepts considered for water systems.

4. Other Concepts - There is little indication of concepts for utilizing organic fluids in unique ways. One thought along this line has been that radiation energy could be used more efficiently for promotion of chemical reactions if all or part of the moderator or coolant were composed of the organic feed stream upon which it was desired to effect a chemical change.⁽¹⁸⁾ Another adaptation of aqueous concepts might be that of a homogeneous reactor, by utilizing a soluble uranium or plutonium organic compound. What is known of soluble organic compounds, however, suggests that they are likely to be oxygenated and of low radiation stability, and that the necessary solvents would also be oxygenated materials.

It is apparent that the direction of development of organic concepts has been largely to consider organic coolants in much the same way as inorganic coolants, i.e., without full recognition of their unique chemical potentialities and problems. A principal reason for this has undoubtedly been due to the need to bring into design considerations for such reactors a whole new field (new to the reactor engineer accustomed to nonorganic reactors) of organic chemistry and organic chemical technology, much of it as yet only little explored. Added to this is the concept of coolant systems which are subject to extreme variations in both initial and operating compositions and properties, as well as methods of treatment. An organic reactor has many of the aspects of a complex chemical plant. It may be possible to take advantage of the chemical occurrences in planning reactor systems, rather than to attempt their elimination.

B. Sources of Information

1. Organizations Known to be Active in the Field - Early work on organic coolants and reactor concepts was carried out at Argonne in the period 1953-55, and Mine Safety Appliances Company made studies and evaluations of biphenyl systems in the latter part of this period. Aerojet-General made measurements of polyphenyl burnout limits in 1956-57. Extensive studies of physical properties and in- and out-of-pile performance of polyphenyl coolants were carried out by the Hanford Atomic Products Operation in 1957-58. These organizations are understood to be relatively inactive in this field at present.

Atomics International has been carrying out extensive work in all phases of organic reactor technology previous to 1954. These investigations were concerned with pyrolysis, radiolysis, heat transfer, and treatment of coolant, fuels development, design and operation of a reactor experiment, reactor conceptual design and evaluation, and development of reactor system accessories. The California Research Corporation is continuing work started as early as 1948 on radiation effects of a wide variety of organic materials, and is now studying mechanisms of coolant radiolysis and evaluation of alternate coolants and inhibitors. A coolant loop was operated in the Brookhaven pile in 1955 by the Monsanto Chemical Co. Monsanto is continuing studies of coolant reclamation initiated in 1958, and has recently commenced studies of the physical properties and behavior in boiling loops for diphenyl. Atomic Energy of Canada, Ltd., at its Chalk River Laboratories and by contract with Canadian General Electric Co., is studying and developing the heavy water system in all aspects, including basic physics and decomposition effects, heat transfer and fouling in- and out-of-pile loop tests, fuel and systems development, and design of a reactor experiment proposed for Whiteshell in Manitoba. The Bartlesville laboratories of Phillips Petroleum Co. have recently commenced a study of coolant decomposition and fouling mechanisms, coolant reclamation and additives, and alternate coolants. MIT is initiating an in-pile loop study of coolant performance. ORNL has a small effort on a search for organic-soluble uranium compounds.

The Atomic Energy Division of Phillips Petroleum Company at Idaho Falls is preparing for operation of the EOCR in which it will operate loops for studies of fuel and coolant technology. Studies of reactor concepts have been prepared by Westinghouse, Martin, and Marquardt, and some work may be under way by these organizations.

The UK several years ago undertook studies of thermal and radiolytic decomposition of polyphenyls, and is continuing basic work at Harwell on radiation damage to organic compounds. Outside of the UK, studies have commenced only quite recently. Euratom is committed to constructing an organic test reactor under the ORGEL project, and has made a number of contracts for research, development, and design work on various phases of the proposal. The CNEN in Italy is planning an organic test reactor (PRO) and is commencing extensive studies of fuels, coolants, systems design, and loops. The Danish group at Ris¢ commenced work in the organic field in 1957 and is increasing its efforts aimed at a heavy water-moderated reactor. In Germany, two groups are studying, respectively, an organic-cooled-and-moderated power reactor and a similar type of ship reactor (AKS and Hamburg-GKSS).

2. <u>The Literature of Organic Reactors</u> - Over 200 references have been found having more or less direct application to the organic reactor field. Many of these have been utilized in this study, although no attempt has been made to prepare a complete bibliography. One bibliography on coolants is available in reference (19), and an extensive list of reports is presented in Appendix A.

3. Organizations with Whom Discussions Were Held - In the course of this study, discussions were held by one or more members of the group with the following organizations:

Atomics International (Idaho Falls and Canoga Park)
Phillips Petroleum Co. (Idaho Falls, Idaho, and Bartlesville, Okla.)
California Research Corporation
Monsanto Chemical Company (Everett, Mass., St. Louis, Mo., and Dayton, O.)
Atomic Energy of Canada, Ltd.
Canadian General Electric Co.
Massachusetts Institute of Technology
U. S. Mission to Euratom (Brussels)
Euratom (Brussels and Ispra)
CNEN (Rome)
Danish AEC (Risø)

In addition, members of the group attended information-exchange meetings on the organic field at Idaho Falls, January 30-31, 1961, and at Chalk River, March 21-22, 1961.

IV. GENERAL ASPECTS OF ORGANIC REACTORS

The physical and chemical behavior of organic materials contributes to various aspects or organic reactors, in some respects uniquely. Certain of these distinctive features can be seen in all types of organic reactors, whereas others are applicable only to particular types. These attributes generally make up the advantages and disadvantages of organic reactors relative to other reactor systems.

A. Aspects Common to All Concepts

The unique features of organic reactors are almost all a result of the chemical behavior of organic materials. Certain of the more significant aspects of this behavior are discussed below.

Effects of Radiation Damage - The most apparent characteristic 1. of organic reactors is the major importance of radiation damage to the organic material. This is a factor whose diversity and importance is comparable to that of radiation damage to nuclear fuel materials in general. In characterizing organic reactors, whether moderated and cooled or only cooled with the organic materials, a significant fraction (in the range from 1 to 10 percent, depending on the volume fraction exposed and on flux patterns) of the energy liberated by the nuclear reaction is absorbed by the organic material. Generally, only a small fraction of the absorbed energy is effective in a chemical way. The effect of the energy absorption is evaluated by a factor, G (molecules reacting to produce other chemical species per 100 ev absorbed) and by the nature and effects of the products of radiation damage. Values of the factor G vary within orders of magnitude for simple organic compounds. In contrast to radiation effects on inorganic reactor coolants (gases, water, liquid metals, and salts), the results of radiation damage to organic materials are generally quite complex and are largely irreversible. A major reaction product (if there is one) may be accompanied by a great variety of minor and secondary reaction products in widely varying yields. Minor products might conceivably exert significant deleterious effects on the operation of a reactor. Since reactions effected by ionizing radiation on organic materials are generally not simple dissociations, the starting materials may not readily be reformed. For example, whereas the principal products of water radiolysis are hydrogen and oxygen, which are readily recombined to water in a simple device, the sum total of the hydrogen, light hydrocarbons, unsaturated materials, rearrangement products, and polymers resulting from irradiating hydrocarbon mixtures cannot conceivably be completely reconverted to their original form without making use of very elaborate methods.

2. <u>Thermal Stability</u> - A second basic aspect of organic reactors is that due to the limitations upon the thermal stability of organic materials. In general, organic compounds become thermodynamically less stable toward decomposition and rates of reaction are rapidly accelerated as temperatures are increased. Although organic compounds vary widely in thermal stability, and the amount of material decomposed is a function of exposure time, it appears that, with minor exceptions, no organic compounds can be exposed for significant periods to temperatures exceeding about 530°C without serious decomposition. Therefore, this is about the upper temperature limit for fuel surfaces or other areas in a nuclear reactor to which an organic material is exposed.

3. <u>Materials Compatibility</u> - A third and somewhat less general characteristic relates to the relative chemical nonreactivity of organic compounds of interest with common and useful construction materials. Although there exists an enormous variety of organic compounds, many of which are highly corrosive to metals, the nuclear and thermal characteristics desired in organic reactor liquids have thus far limited the field of interest to hydrocarbons. The hydrocarbons themselves, in general, are relatively inert in contact with aluminum, mild and stainless steels, and most other metals, although reactions with impurities in the organic material may occur. This now permits the use of more economical and easily fabricated materials for both fuels and systems structure than generally may now be used for other types of reactors, and of relatively conventional heat and fluid transfer equipment.

4. <u>Heat Transfer Capacity</u> - Organic materials are characterized by having low heat transfer coefficients, low heat capacity, and small heats of vaporization, as compared with water and liquid metals. For both forcedconvection cooling and boiling conditions, this requires increased coolant circulation and heat transfer area, or greater temperature differences, as compared with what can be used with other materials. Correspondingly, the specific power of an organic reactor tends to be low.

5. <u>Flammability</u> - Another common characteristic of organic materials useful for reactors is that they are flammable and will burn under some circumstances. The effect of this danger may be reduced by choice of very high boiling materials, but radiolysis in general will cause breakdown and increase flammability.

B. Aspects of Specific Concepts

Some further aspects of organic reactors, which may represent significant advantages, are not characteristic of all organic concepts.

1. Low System Pressure - If an organic coolant of low volatility and high molecular weight is used in a reactor, the system pressure may be relatively low (of the order of 100-200 psi). This permits a reduction of reactor vessel, pump, and other component costs, and simplifies shielding considerations in that the primary coolant cannot readily leak into the steam-generating system, which would normally be at a higher pressure. Correspondingly, the possibility of leakage of water into the coolant system may result in corrosion problems and nuclear hazards. A hydrogenous organic liquid of low vapor pressure might also be used conveniently as part of the reactor biological shield.

The use of more volatile coolants in other concepts in which boiling occurs and vapor is passed to a turbine may somewhat negate the advantage of low system pressure (though not relative to water at similar temperatures).

2. Low Coolant Activation - The availability of relatively pure hydrocarbons at moderate cost, and the low activation cross sections of these materials, ensure that coolant activation will be considerably reduced with resultant simplifications in shielding and in system maintenance. This is a very significant advantage for nonboiling organic systems compared with most other liquid-phase coolants. However, such an advantage may be lost or diminished under the following circumstances:

- a. Use of fuel materials for which the probability of cladding failure is such as to require protection from effects of coolant contamination from fission product dispersal.
- b. Use of systems permitting corrosion, from structures in contact with the coolant, of chemical elements which can become highly activated.
- c. Use of low-cost impure coolants (perhaps from oilrefinery sources) which contain chemical elements, such as sulfur, which can become activated, or intentional use of additives or special coolants containing elements subject to activation.

3. <u>Coolant Processing and Disposal of Waste Coolant</u> - Since, in general, organic coolants are significantly decomposed in service, cannot readily be recombined to restore the original composition, and are too valuable to discard in large amounts, systems must be provided for separation, disposal of decomposed and polymerized material (gas, liquid, and possibly solid), and perhaps for reclamation of useful fractions. Such systems, though largely adaptable from existing organic chemical technology, can be more complex than required for other types of reactors, although the treatment of primary cooling water in a reactor is somewhat parallel in nature.

The disposal of potentially contaminated organic coolant, by combustion or otherwise, presents new and unique problems. Since coolant nature, purity, and stability can vary widely, so also can the magnitude of the disposal problem. It might be possible to devise a very special organic coolant system, selected specifically such that decomposition products could readily and completely be reformed to useful coolant, for which disposal systems would not be required. Further, reactor coolant systems may be devised for which rejected coolant has a byproduct or major product value (i.e., chemonuclear systems).

4. <u>Nuclear Safety</u> - It appears to be possible to design practical organic-moderated reactors, as well as certain other types of reactors, to take advantage of significant negative temperature, void, and power coefficients, and thereby to enhance nuclear safety characteristics. This behavior is presumably restricted to reactors which contain organic material in a sufficient, but not excessive, volume fraction in the core. Insofar as these effects are characteristic of organic liquids, they are not necessarily characteristic of reactors cooled by relatively thin films of organic materials and moderated by other materials.

V. EVALUATION OF ORGANIC COOLANTS

The unique component common to most organic reactors is the organic compound or mixture of compounds used to transfer heat from the reactor fuel. Coolants are selected on the basis of required physical and chemical properties and other desirable characteristics. Thermal and hydraulic behavior must be well known. Means for economic production must be available, and processes for treatment in the reactor system, reclamation, and disposal may be required. Its use in a reactor must not result in deposits which would adversely affect fluid and heat transfer.

A. Radiation Chemistry of Organic Reactor Materials

1. <u>Selection of an Organic Coolant</u> - The principal disadvantage of the organic-cooled-and-moderated reactor is the thermal and radiation decomposition of the moderator-coolant. In order to cope with this disadvantage one must consider several alternatives.

- a. Find the most stable organic material and accept the decomposition if the cost of the material results in an economically attractive system.
- Find a low-cost material which could have a relatively higher decomposition rate, but also provides low makeup costs.
- c. Consider a very low-cost material coupled with a very limited throughput time after which the material can be completely discarded or used for other purposes.
- d. Use selected coolants from which useful radiolytic products can be recovered.

All of the materials in the above four categories should preserve some of the inherent advantages of an organic coolant, namely:

- (i) low vapor pressure at operating temperatures;
- (ii) low levels of induced activity;
- (iii) low corrosion rates with low-cost materials of construction; and
- (iv) no danger of exothermic reactions with fuels, water, or other materials of construction.

It has long been recognized, from fundamental radiation studies of organic compounds, that compounds containing a resonant ring system, such as benzene and other aromatic compounds, exhibit a stability which is several times that of most other organic compounds with regard to total products formed. In addition, such substances are stable by one or two orders of magnitude more than nonaromatics with regard to production of gaseous products Although the mechanistic details of radiation damage to organic compounds is far from being firmly established, it does seem that two properties of the resonance-type compounds contribute to their relative stability, namely:

- l dissipation of the absorbed energy over many bonds (delocalization of energy), and
- 2. effectiveness as a scavenger, i.e., the ability to react with thermal free radicals formed in the initial radiation act, thereby preventing chain decomposition.

Early screening tests by California Research Corporation (CRC) have shown that biphenyl, o-, m-, and p-terphenyl, and naphthalene were the best of forty aromatics tested and that their thermal stabilities were reasonable up to 490°C.

The effect of fast electrons on the polyphenyls at 350°C is mainly to give polymer, the G for conversion of monomer being 0.05-0.5 and the G for production of gas being 0.003-0.03. Fast neutrons seem to produce 3-6 times as much polymer and nearly ten times as much gas per 100 ev absorbed. Fission fragments, in turn, produce several times the polymer produced by fast neutrons and amounts of gas greater by an order of magnitude. The polymer produced is mainly a dimer mixture of polyphenyls containing one or more phenyl groups more than the starting material. Complete analyses of the polymer are not yet available, but other products include partly unsaturated aromatics and alkyl aromatics. The formation of polymer causes the viscosity of the molten material to increase with dose, first at a constant rate, then at an increasing rate, culminating in the formation of a coke-like material. The melting points show a similar behavior. The rate of decomposition of polyphenyl decreases with dose, indicating the formation of radiation-resistant products. Also, the addition of small amounts of higher polymers of the polyphenyl type seem to protect radiolytically virgin material of the polyphenyl type.

The major efforts of those engaged in organic reactor design have been concerned with alternative <u>a</u> (use of the most stable material consistent with technological and economic considerations) and have culminated in the selection of some commercially available mixture of terphenyls, terphenyls and biphenyl, or biphenyl. An initial systematic screening of biphenyl and terphenyl was done by the CRC. Subsequently, Atomics International (AI) selected a commercial terphenyl mixture as a moderator and coolant for the OMRE, and the bulk of the available information on this material available stems from the operation of this reactor, as well as laboratory testing by AI. This contractor also has embarked on a much-needed program of fundamental research on the mechanism of radiation decomposition of organics. Atomics International is continuing its study of coolant technology in OMRE with core III and will have EOCR as an additional tool in which in-pile loops will be used to study coolant technology. Other contractors now engaged in or planning work in coolant technology and effects of radiation are: Phillips Petroleum Co. at Bartlesville and Idaho Falls, Monsanto Chemical Co., and Massachusetts Institute of Technology. In addition, studies are being carried out by AECL at Chalk River, by Canadian General Electric, by AERE at Harwell, and by Euratom.

The basic primary processes associated with the absorption of energy in an organic molecule are generally:

- a. the dissociation of energized molecules into radicals; the term energized molecules includes molecule-ions of the type observed in a mass spectrometer, as well as highly excited ions and excited molecules such as those important in the photolysis of these materials;
- b. the interaction of hydrogen atoms with an organic molecule;
- c. the interaction of aryl radicals with hydrocarbons; and
- d. molecular dissociation of excited molecules into stable products.

In addition to these possible reaction types, one must consider the effect of ionization density and the type of radiolysis products. This is referred to as the Linear Energy Transfer (LET) effect or fast neutron effect.

Studies of the radiation chemistry of the resonance-type molecules give evidence of all of these primary reactions. The extreme complexity of the reaction products indicate the importance of all of these reactions. For example, the polymer products of benzene radiolysis include biphenyl, phenylcyclohexadiene, phenylcyclohexene, dicyclohexyl, and a rather wide spectrum of products including 2 and 3-ring structures of various degrees of hydrogenation in one or all rings. Studies of biphenyl and the terphenyls indicate a similar or even a more complex spectrum of similar products. 2. Radiation Decomposition Experience in OMRE - The choice of alternative <u>a</u> (p. 16) with the selection of terphenyls as the coolant by AI for use in OMRE was an understandable choice based on the state of knowledge of radiation damage of organic materials and of coolant technology at that time. Operation of OMRE with both core I and core II has demonstrated in general, the expected rate of decomposition. The virgin coolant used in OMRE is a reactor grade of Santowax furnished by the Monsanto Chemical Co In general, the composition runs 1 part ortho-terphenyl, 5 parts meta-terphenyl, 3 parts para-terphenyl, and 1 part higher boiling components. Other grades of Santowax, consisting of varying isomers, biphenyl, and higher boiling components, are available.

The rate of decomposition of the coolant in cores I and II varied as a function of the high boiler concentration which builds up during operation. The high boiler fraction (HB) is described as those compounds less volatile than the original major coolant components (the least volatile of which is para-terphenyl). The G(-coolant) (that is, the number of molecules of coolant decomposed per 100 ev of energy absorbed, assuming a molecular weight of 230) varies from 0.25 at a HB of 11% to 0.12 at a HB of 40% This is based on a rate of absorption of radiation energy in the moderator-coolant of 78% of the total power of the reactor. At 30% HB concentration, this rate amounts to about 51-52 lb/Mwd(t) of coolant decomposed Extrapolating these data to a full-scale power reactor, such as Euratom AKS (150 Mwe), AI predicts an HB formation rate of 27 lb/Mwd(t) for reactor operation at an HB concentration of 30%. Under these conditions there is a corresponding gas generation of 8 SCF/Mwd(t).

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The decomposition rates were based initially on the assumption that gammas and neutrons are equally effective in decomposing the coolant. More recent data indicate a ratio $G_{(n)}/G_{(\gamma)} = 0.31/0.08 = 4$. However, AI claims that they did not properly assess the inelastic scattering effect in the larger reactor, and the increased inelastic scattering decreases the fast neutron energy absorbed in the coolant relative to the gamma energy, the two effects compensating each other and making the original extrapolation valid. The extrapolation to larger reactors involves, in addition to establishing the G values for gammas and fast neutrons, a proper assessment of the inelastic scattering of fast neutrons and a calculation of the fission energy absorbed in the particular reactor under consideration. This is a matter which should be investigated more thoroughly. The value of 30% HB is an arbitrary compromise between reduced decomposition rate and reduced heat transfer ability, and acceptable viscosity characteristics. It should be stressed that this is purely arbitrary and there is a possibility that further study will indicate a better alternative.

Other organizations have been studying rates of decomposition of terphenyl-type coolants under radiation. In general, the results reported are in agreement with those of AI except for two important exceptions reported by AERE. One is the G for neutrons versus G for gamma rays. This seems to have been resolved, as has been discussed above. The second point is a study by AERE of the effect of temperature on the radiation-decomposition yield of the coolant. They have observed that the G(-coolant) for Santowax R slowly increases with temperature until just above 400°C. Above this temperature, there is a rapid increase of G(-coolant) with temperature. At 404°C, the G(-coolant) is 0.79; at 415°C it is 1.19; and at 419°C it is 1.86. These results were obtained at a dose of approximately 2.4 watt-hr/gm. These results cannot be explained on a purely thermal basis but seem to be a combined radiolytic and thermal effect. This point should be examined further.

The changes of physical property under irradiation in OMRE have been measured by AI. The specific heat is found to decrease slightly with decomposition. For the 30% decomposed coolant at 311°C, the decrease is 4%. Thermal conductivity of OMRE coolant has been studied as a function both of temperature and radiation decomposition. The thermal conductivity is found to decrease with temperature, and for irradiated OMRE coolant containing 30% HB it is found to be about 4-9% higher than that of the unirradiated coolant. The viscosity of the coolant increases with decomposition about 100% as the HB content varies from 0 to 30 weight %. The density changes only slightly (~4%) in the same HB range. For all the coolants studied by AI, the melting point initially decreases with irradiation, and then gradually increases at high HB content. For example, the liquidus temperature of OMRE coolant (core I) was 96°C before decomposition, decreased to a minimum value of 57°C at 24% HB, and increased to 74°C at 41% HB.

Atomics International has also made studies of the chemical behavior of the coolant, including a) nature of the coolant decomposition products in the circulating liquid, b) gross chemical properties of coolant during reactor operation, such as acidity, olefin content, molecular weight distribution of higher boiler fraction, water content, and concentration of reactive species such as free radicals, and c) radioisotopic studies in the coolant.

AI has carried out experimental work on polyphenyl coolants irradiated in in-pile test loops in addition to OMRE polyphenyl coolant irradiations during $2\frac{1}{2}$ years of operation. Experience in both programs are quite similar with regard to gases, low boilers, intermediate boilers, and high boilers. On a weight basis, most of the decomposition products are in the high boiler fraction. The gaseous products account for about 1% of the decomposition products and consist principally of hydrogen together with lesser amounts of methane and higher hydrocarbons. The low boiler fraction is composed of those decomposition products more volatile than biphenyl and equal to or less volatile than benzene. During core II operation, this ranged from 0.13 to 0.95% concentration. Low boilers that have been identified are: benzene, toluene, and ethyl benzene. The intermediate boilers are defined as those having a volatility intermediate between byphenyl and the HB fraction. This group has remained at between 9-12% of the coolant during Core II operation. This appears to be the equilibrium value of this fraction for OMRE. Among this group is found fluorenone, phenanthrolene, 3-methyl biphenyl, and fluorene, with the first three constituting the main fraction, being at least 10 times that of the others. More than 95% of the decomposition products are in the HB fraction.

The average molecular weight of this fraction increases during reactor operation. During Core I operation the average molecular weight increased from an initial value of 400 to a final value of 600 over an elevenmonth period; the change seemed to be linear with time of reactor operation and gave no evidence of attaining a steady-state value. Tentative conclusions of AI from Core II operating data are that there is a possible leveling off at the end of this core operation at a value of 610 for the molecular weight. Data on the distribution of molecular weights cover values from 228 (triphenylene) to 3000 (39 phenyl groups), with the largest single contribution being for hexaphenyl. About 8 weight % of the HB has a molecular weight greater than 1500.

Water content of OMRE coolant has varied randomly in the range from 80 to 260 ppm. According to AI, a water content below 500 ppm 1s of minor significance.

Introduction of oxygen is more serious. It results in increased corrosion rates and pitting, with subsequent increased activity in the coolant, and production of oxidation products in the coolant. These products are themselves of a corrosive nature. Fouling films and some fractions of HB have been shown to contain carbonyl groups as well as phenol, alcohols, and water.

Olefinic unsaturation has been detected in the coolant at a molar concentration as high as 0.06 after 373 Mwd of operation, about 60-85% being in the HB fraction.

3. <u>Use of Additives to Decrease Decomposition Rates</u> - The use of and additive to increase the radiation stability of the coolant is an attractive possibility. An additive is generally believed to act as a scavenger, reacting with radicals produced in the solvent or effecting an exchange of energy between it and the solvent molecules. The polyphenyls are a class of compounds containing a great deal of resonance stabilization and act very efficiently as scavengers themselves. This fact together with our present state of knowledge of the basic chemistry involved suggest that the problem of finding an effective additive for this coolant may be a difficult one. Both CRC and AI have tested a large number of additives. Phillips Petroleum is also testing additives. All the results are either erratic or inconclusive. It should be emphasized that an additive might conceivably alter the nature of the reaction products, and in this way may have a possible bearing on fouling and on the physical and chemical properties of the polymeric product. There is just not enough detailed knowledge of the basic chemistry, the nature of the intermediates, reaction products, and rate constants for elementary reactions involved to make a really intelligent prediction.

One rather surprising result is some work reported by Monsanto. Radiation tests conducted at a level of 20-80 watt-hr/gm indicated that the temperature at which the radiolysis is conducted is an important parameter in the efficiency of a stabilizer. One group of compounds which Monsanto classes as scavengers, such as carbonyl, sulfide, disulfide and polyaromatic compounds, were quite effective at low temperatures in the range from 200 to 300°C. The second group, characterized by Monsanto as hydrogen donors, exhibited a reverse effect of temperature. They were good stabilizers at high temperatures but accelerated radiolysis at temperatures below 100°C. One material in a 10-volume percent concentration reduced polymer formation to one third and yielded very little hydrogen as a gaseous decomposition product. An example of this category was a partially hydrogenated polyphenyl mixture (presumably of compounds in which one ring was completely saturated). Monsanto also observed that, when radiation damage was plotted against degree of hydrogenation of a polyphenyl, the curve exhibited a minimum at low fractional hydrogenation, and that damage increased with further hydrogenation to a level exceeding that of the unhydrogenated compound. Montar (a material in the terphenyl range) hydrogenated to 60% of saturation appears principally to cleave on irradiation, indicating that hydrogenation of part of the ring may promote ring-bond breaking.

These data point out the fact that reactor Santowax OMP polymer (HB) is one of the most effective "stabilizers" tested. The polymer has a hydrogen content which is 20% of saturation; in a concentration of 21% in m-terphenyl the polymer has a hydrogen content of 4.3% of saturation. At this concentration it was effective in decreasing the total decomposition by a factor of 57%.

However, it was also pointed out that other hydrogenated additives may have more desirable physical properties for coolant use. These stabilizers are thermally less stable, but this decrease in thermal stability is more than compensated by the increase in radiation stability. The data indicate that this type of stabilizer at low concentration can protect 2.7 molecules/molecule of stabilizer and that the effectiveness decreases at higher concentrations. Some petroleum streams which were good protectors were also found. However, too little detail is known about these experiments to evaluate the results critically. If they are firmly established, it emphasizes our lack of knowledge of the basic chemistry involved in these compounds.

The results reported by Monsanto do bring out an interesting possibility. It is well known that mixtures of hydrocarbons, such as cyclohexane and benzene, protect one another from radiolytic decomposition by either an energy-transfer process or a scavenging action of one of the hydrocarbons. This has been studied for the production of gaseous products but not for polymer. It might be possible that a mechanism, such as the following, would be effective in reducing polymer yield:

 $C_6H_{12} \longrightarrow C_6H_{11} + H$ H + C_6H_6 \longrightarrow C_6H_7

There is some evidence that C_6H_7 may be a relatively stable radical and that it might live long enough for the following reaction to occur:

 $C_6H_{11} + C_6H_7 \longrightarrow C_6H_{12} + C_6H_6$

This effectively promotes a back reaction and prevents C_6H_7 from reacting to form polymer. A similar mechanism may possibly hold for the hydrogenated terphenyl and terphenyl mixtures studied by Monsanto. A study of the yields of polymer in mixtures of hydrogenated terphenyls and terphenyl under radiolysis may indicate such an effect. Further examination of the polymer yields on the simpler mixtures of cyclohexane and benzene and extending the gas-production studies could possibly yield basic information on this point.

Other Low-cost Coolants (Alternative b, p. 16) - AI, CRC, Monsanto, and Phillips Petroleum are also concerning themselves with testing alternative low-cost coolants. AI has results indicating that alkyl phenanthrene-type stocks from refinery streams look promising. These are obtainable at a price between 5 and 8 cents per lb. Although the G_{gas} of these refinery streams relative to the terphenyls is increased by a factor of 10, the yield of polymer (the more troublesome material) is 0.6 that of the terphenyls. Anthracene oils from coal tar hydrocarbons $(3\phi/lb)$ also show promise. Both CRC and Phillips are also looking at various low-cost refinery streams. Previously, CRC had found a number of refinery streams which, even after minimum refining (necessitated by lowering sulfur content to less than 100 ppm), indicated a cost of 2 to $3\phi/lb$ compared to $15\phi/lb$ for Santowax. This program was curtailed because of a conflict with the AEC on proprietory rights, but is being reactiviated. Phillips has been screening petroleum refinery streams. One fraction was found to be highly aromatic and has a boiling point range from 288° to 538°C. It consists of alkyl naphthalene, anthracenes, and phenathrenes. These compounds cost approximately $1 \frac{\phi}{1b}$ but will require some refining.

Low-cost Material with Limited Throughput (Alternative c, p. 16) - This alternative is being looked at by both CRC and Phillips Petroleum. Phillips is examining aromatic cycle oil stocks which are of interest to carbon black manufacturers. It may be possible that the residue after passing through the reactor and after extraction would still be useful for carbon black manufacture.

Use of Selected Coolants from Which Useful Radiolytic Products Can be Recovered - CRC is also reviewing the possibility of producing commercially useful material by the effect of radiation on the organic coolant and moderator. Since radiation attacks all organic molecules, resulting in cleavage, polymerization, and cross-linking reactions thereby forming a wide variety of compounds whose structures differ from that of the starting material, it may be possible that useful compounds can be formed in this way from selected coolants. Their recovery and sale could be a cost credit to the reactor plant. One such material chosen for study is the group of polyphenyl ethers, which now cost about \$20 per lb. These compounds have been the object of intensive research for use as high-temperature lubricants. They might be prepared cheaply from diphenyl ether and might be used as a coolant or as additives to presently used polyphenyl or other coolants. CRC is looking at this possibility. There is a wide field of investigation for the simultaneous production of these products from possible coolants, and their products could materially lower effective coolant costs.

4. <u>Research and Development Program on Organic Coolants</u> - The problems of interest to a productive research and development program may be divided into two groups: those of interest in current coolant technology, and those of importance to the development of an advanced coolant technology.

In the field of current coolant technology, AI is continuing its study of the terphenyl coolants, using the OMRE principally as the radiation source. It is of course important to establish within as close limits as possible the physical and chemical properties of the terphenyl coolants under reactor operating conditions, to determine makeup costs, and to establish specifications for control of coolant properties. In addition, the EOCR will soon be available as an additional facility for these studies. The several problems in current technology which seem to have an urgent priority are:

1. To establish firm G values for coolant decomposition by different types of radiation, particularly by fast neutrons and γ -rays, and the possible effect of energy on the G values. This should be coupled with more sophisticated calculations of the distribution of energy dissipation in the moderator so as to assess more accurately the decomposition rate in proposed full-power reactors. Such a program has started.

2. To study the effect of temperature on the radiation-produced yields in terphenyl coolants. This should be done particularly to establish the upper temperature limits of operation and the possible effects on the nature of the products produced This is particularly emphasized in view of the recent Harwell results.

3. Examine the role of the coolant, under radiolysis and at high temperature, in the fouling problem. The possible use of additives to alter the rate of fouling or the nature of the fouling film should be examined.

4. Pursue the possibility of additives to minimize the rate of decomposition or to change the nature of the radiolytic decomposition products in order to produce an equilibrium coolant mixture with more desirable properties.

5. To develope analytical tools for analysis of polymer composition. Gas chromatographic techniques have shown a surge or productive development and should be further exploited. Mass spectrometric techniques for analysis for polymer components should be further developed. CRC has devised a low-voltage ionization technique which simplifies the mass spectra pattern. Another technique along these lines is the possibility of using α bombardment (with a polonium source) as the source of ions. This has been developed at Oak Ridge and yields greatly simplified mass spectra patterns. Use of the osmometer to determine molecular weights is now being carried out at Phillips and should make available more complete and accurate analysis of molecular weight distributions in the polymer.

6. To study basic radiation chemistry of the polyphenyls and other aromatics. The detection of unstable intermediates and the kinetics of their formation and disappearance is a requisite for this study. It should be emphasized that as complete an analysis of reaction products as possible should be one of the major efforts of such a kinetic study.

Problems in advanced technology should include:

1. Fundamental mechanisms of radiolysis of a broad spectra of organic compound types.

2. Development of new coolants, both of synthetic and industrial materials.

3. Examination of economically useful materials which can be produced in the coolant under radiolysis.

4. Development of new stabilizers.

Several companies have under way or are planning programs covering these topics. Further efforts along these lines of research and development should be encouraged.

B. Sources and Costs of Polyphenyls

Up to now the material which has received major attention as an organic coolant and moderator is the commercial terphenyl mixture sold under the trade name Santowax by the Monsanto Chemical Company. Santowax OMP is a mixture of ortho, meta, and para-terphenyls. Santowax R is the same material with the addition of about 11% of higher boiling components. Terphenyls are also produced by Dow Chemical Company and by a few European concerns, but the total production of these is said to be less than half that of Monsanto.

Terphenyl is produced by pyrolysis of benzene at 700 to 850°C. The process also produces biphenyl and higher polyphenyls, which are separated by distillation. Currently, terphenyl is a byproduct of biphenyl production. Since the present market for terphenyl is small, the process conditions are adjusted to produce a minimum of terphenyl. However, should the demand for terphenyl increase the process conditions can be adjusted to produce terphenyl as the major product.

Monsanto has guaranteed that they could supply up to 10 million pounds per year of Santowax OMP at a price of 15 cents/lb, exclusive of containers and shipping cost. The price of Santowax R would be about $14\frac{1}{2}$ cents/lb. In larger amounts, of, say, 50 to 100 million lb/yr, the price would decrease to possibly 12 cents/lb. All of these prices are conditioned on a cost of the raw material (benzene) below 40 cents/gal. The current cost of benzene is 35 cents/gal. For easy handling it is recommended that the terphenyl be shipped in flaked form in bags. The cost of flaking and bagging would be about $\frac{3}{4}$ cent/lb.

Santowax contains a small amount of sulfur impurity which is derived from the starting benzene. Due to activation of sulfur in the nuclear reactor, the degree of difficulty encountered in the disposal of waste gases during reactor operation may be influenced by the amount of sulfur present in the coolant. Sources of benzene of low sulfur content are known, and it has been stated by Monsanto that the selection of benzene of low sulfur content would probably have only a slight effect, if any, on the price of terphenyl. Biphenyl has also been considered as a coolant for nuclear reactors, particularly for direct-cycle concepts. Since biphenyl is produced by the same process, its price is approximately the same as that of terphenyl.

C. Coolant Processing and Reclamation

The coolant for an organic reactor must be purified continuously to remove the radiolysis and corrosion products which form during reactor operation, and these waste products must be disposed of in a safe and economical manner. The following processes are currently considered necessary for this purpose:

- (1) removal of insoluble impurities from coolant;
- (2) separation of high boiler from coolant;
- (3) separation of water and decomposition gases from coolant;
- (4) disposal of waste gases; and
- (5) disposal of waste solids, including insoluble impurities and nonrecoverable coolant residues.

1. <u>Removal of Insoluble Impurities</u> - It is generally believed that the presence of particulate matter, specifically of iron oxide corrosion products and possibly other inorganic or organic particles, will lead to fouling of fuel elements if allowed to accumulate in the organic coolant. Consequently, it is important to keep the concentration of particulate matter in the coolant at a very low level. The precise concentration of particles which can be tolerated is yet to be determined, but, on the basis of OMRE operating experience, Atomics International has established a goal of 10 ppm of particulate ash or less to achieve an acceptable fouling rate.

Experimental work on coolant cleanup has included tests in the OMRE as well as laboratory and pilot-plant work. The methods which have been studied include filtration, centrifugation, magnetic and electrostatic separators, adsorption, and distillation. The efficiency of these methods has been evaluated by determination of the coolant ash content before and after treatment. The work done to date has indicated several methods which might be adapted to clarification of organic coolants. A program is under way at OMRE to study various types of equipment for this service. It has been suggested that the optimum installation may incorporate two devices - one for a first step treatment to remove larger particles, and the second for removal of very fine particles. The very small size of the particulate matter and the high coolant temperature make the problem of coolant clarification somewhat more difficult than the common filtration problem. However, the removal of small-size particles from liquids (even down to colloidal size) is not an uncommon problem in industry, and a number of methods are available for such clarification operations. The problem of clarification of the organic coolant therefore resolves itself primarily into a selection and testing of the most desirable equipment and method.

Knowledge of the particle size of the insoluble matter is highly important to the proper selection of a process for coolant clarification. Particle size distributions have been obtained by AI for five samples of OMRE coolant obtained during various operating periods of the reactor. These determinations are subject to appreciable uncertainty because of experimental difficulties, so that the results can be considered as only tentative. The preliminary data indicate that the size of the particles is in the range from 0.01 to 12 microns. The size distribution and ash content for one sample of OMRE coolant are as follows:

| Particle Size (μ) | Weight Percent of Total Particles | Ash Content (as Fe ₂ O ₃ , %) |
|---------------------------|--------------------------------------|--|
| >5 | 18 | 70 |
| 5 to 0.22 0.22 to 0.01 | 7 75 } | ~5 |

The particles larger than 5 microns contained almost all of the iron in the particulate matter. These data indicate that the bulk of the particles are of colloidal size (below 0.1 micron). Additional work is required to establish firmly the size distribution of the insoluble matter.

The above data on the size spectrum of the particulate matter and the iron content of variously sized particles introduce considerable uncertainty in the interpretation of the results of previous coolantclarification experiments conducted by AI. Many of the data which have been reported on the particulate-removal efficiency of various devices are of doubtful value, because these efficiencies are generally based on the effectiveness of the devices for removal of inorganic material from the coolant. Since the inorganic content of the small particles below 5μ is only about 5%, the change in inorganic ash content of the coolant after treatment would be a measure primarily of the efficiency for removal of particles of large diameter, which constitute only a minor fraction of the total insoluble material. This situation is realized by those studying the fouling problem at AI, and in recent work a pyrolytic capsule fouling test is being employed to supplement the criterion of ash content as a measure of the effectiveness of coolant clarification. If it is found that the small particles, constituting some 75% of the total particulate matter, must be removed to avoid fouling problems, then a number of the clarification devices currently under study are of questionable value except as primary separators solely for removal of larger particles. For example, magnetic separators might not be effective because of the low iron content of the fine particles. Likewise, centrifugation would be of doubtful efficiency with very small particles unless adsorbents or filter aids were first added to the coolant. Since the data indicate that the larger particles represent only about 25% by weight of the total particulate matter, the value of a primary separator appears questionable, because it would not materially reduce the load on the secondary unit.

Another important aspect of the problem is the rate of formation of the particulate matter in the coolant under normal operating conditions and under abnormal conditions, such as following periods in which air or moisture has been admitted to the reactor. This information is important for the proper choice of equipment size and method for clarification. Data have been reported on the concentration of particulate matter in the coolant during operation with the OMRE Gore II. Particulate ash concentrations up to 200 ppm were observed during periods of reactor shutdown. It has been reported by AI that the total particulate mass produced in the OMRE operating under normal conditions is less than 25 grams per day. On the other hand, a single shutdown of several days duration has apparently been responsible for the production of several kilograms of solids in the coolant. The available information on rate of formation of insoluble particles is, however, very meager and very approximate.

For particles of very small size it is common industrial knowledge that a clarification method based on adsorption would have the most likelihood of being successful. The adsorbent could be in the form of filter aid which is added to the coolant, then removed by a mechanical filter; or the adsorber might be comprised of a packed column of some industrial adsorbent. Certain treated clays have been in widespread use for such filtration of petroleum fractions. Both types of adsorbents, filter aids and clay beds, are included in the experimental studies at AI and have shown promising results.

One disadvantage of the industrial adsorbents, such as clays, is that their capacity is frequently low. This means that the amount of clay required may be unduly large, constituting a waste problem and a handling problem because of the presence of radioactivity. However, it may be possible to regenerate the clay by burning off the adsorbed material, as is commonly done in the petroleum industry. This points out the need for firm information on particulate generation rates, in order to establish the required capacity of such equipment. It is entirely possible that a unit, such as an adsorbent column, would be suitable during periods of normal operation, and this could be supplemented by another type of filtration unit for abnormal periods, such as when the reactor is opened for fuel unloading. It is also possible that the distillation column used for high boiler separation would be adequate for particulate matter removal during normal periods, even though the feed rate to the purification unit is low.

There appears to be every likelihood that the current experimental program will result in a suitable method for coolant clarification. Sorely lacking, however, is information on the extent of the particulate problem for a reactor which is operated under conditions approaching that of a large power reactor. To supplement the information which will be obtained from OMRE operation, it is essential that information on the particulate problem be obtained from more advanced reactors, such as the EOCR and the Piqua reactor. Until this data is obtained the selection of the optimum method of coolant cleanup may not be possible.

2. <u>Removal of Decomposition Products and Gaseous Impurities</u> -During reactor operation, the organic coolant undergoes certain changes which require that it be continuously treated. Radiation and heat induce polymerization and "cracking," resulting in the formation of "high boilers" (organic fractions with boiling point above para-terphenyl), light organic compounds, and hydrogen. In addition, possible leakage in the heat exchanger may introduce water into the organic coolant. For the organic reactor systems currently under consideration, it is planned that the high boilers be removed from the coolant continuously by vacuum fractionation. The water, hydrogen, and other light gases are to be continuously removed by flashing in a large vacuum tank.

Experience at the OMRE, as well as laboratory data, have shown that the higher boilers can be removed easily by distillation with very good (99%) separation of high boiler from reusable coolant. This experience was obtained with a batch-operated still. Automatic, continuous purification units have been designed for OMRE, EOCR, and the Piqua Reactor. None of these have yet been operated, but a full-scale prototype of the Piqua purification unit was in operation during 1960 in a brief test program with OMRE coolant. Purification is accomplished by feeding the hot coolant into a flash chamber maintained at a pressure of about 1 psia. Most of the coolant is flash-vaporized into the coolant condenser. The remaining coolant enters a stripping column where the last bit of coolant is removed from the HB. As a result of operating the prototype unit, several design changes were made, and the test program is continuing. At the present time the design of the purification unit cannot be considered proven since only a small amount of operating experience has been obtained. However, fractionation of organic materials is a well-known technology, and no serious problems are to be expected in this operation.

The method which has been selected for removal of gases from the coolant depends upon the principle of a reduction in the solubility of the gases with decreasing pressure. Two experimental degasifier loops have been operated, the first being a small-scale unit and the second a quarter-scale prototype of the Piqua degasification system. Operation of these units permitted a study of mechanical problems, which finally resulted in moderately satisfactory mechanical operation. However, the amount of operating experience which has been gained on the degasification operation is small, and the amount of useful data which has been reported is also small. Considerable difficulty was encountered in the analyses of coolant for water content before and after degasification, so that the reported data are not highly convincing. More recently, an improved sampling technique was developed, and the analytical results obtained are said to be reliable. Recent degasification runs indicated that the water content of the coolant was reduced from several hundred ppm to 100 ppm, which is considered adequate on the basis of OMRE experience. No data are reported on the removal of gases other than water vapor and nitrogen. The principle upon which the degasification operation is based is theoretically sound, but additional practical experience and more quantitative data are desirable to prove out the method.

3. <u>Waste Disposal</u> - The polymerized high-boiling residues which are removed from the coolant by continuous distillation might be disposed of by the following methods:

- (1) regeneration to usable coolant and return to the reactor;
- (2) burial at land or sea; and
- (3) combustion of the high boiler and burial of the ash.

Regeneration of the high boiler to usable coolant is the most desirable method from many standpoints, but a large amount of development work must be done before the economic feasibility of this method can be established. Experimental work on coolant reclamation is discussed in the next section of this report. Even if successful, it is likely that a portion of the high boiler would not be recoverable and would require disposal. The choice between burial and combustion of the high boiler is not an obvious one unless uncontained burial in terrestrial pits is possible. A considerable amount of additional study would be required to establish the feasibility of such burial.

Most of the experimental work on high boiler disposal has been concentrated on combustion. The technical feasibility of such combustion has been fairly well established in prototype-scale equipment for the Piqua reactor. A commercial burner designed to handle asphaltic type wastes appears to be suitable for this purpose. Operation of the Piqua reactor will provide additional experience on this disposal method. The gases produced by combustion of the high boiler will contain radioactive particles from activated corrosion products, as well as volatile radioactive gases, such as carbon and sulfur oxides. The current plan is to remove particulate matter by dry filtration, using an automatic bag filter followed by an absolute filter. Experimental studies on prototypescale equipment indicated that particle removal to the extent required for the Piqua reactor system (99% removal) could be obtained. However, further work needs to be done to test filter media capable of withstanding the high gas temperatures.

The volatile radioactive gases and any remaining particulate are to be diluted in a stack to permissible levels. Since the design of the Piqua plant includes a decay tank capable of holding a six-month output of high boiler (above the melting point), the problem of gas disposal is reduced because of decay of short-lived isotopes. For a full-scale power plant, the holdup of molten high boiler for this period of time would be obviously undesirable because of the large storage volume required (of the order of $\frac{1}{2}$ million gallons for a 300-Mw plant). It appears probable, however, that a much shorter cooling period will be possible if a special method for removing S³⁵ is developed. Laboratory studies have been carried out on removal S³⁵O₂ from gas streams, but the process is still in a preliminary stage of development. The requirement for decay storage of high boiler would probably be eliminated if a feasible method of regenerating coolant is developed.

In addition to these gases, the waste gases from the organic coolant degasifier and the purification unit must be disposed. These are estimated to be composed of about 63% hydrogen together with hydrocarbon gases, such as methane, ethane, and propane. The volume of these gases is only a small fraction of that produced by burning high boiler. Under normal conditions of reactor operation, the safe release of these to the atmosphere requires only dilution of the radioisotopes to permissible limits and the removal of offensive, and possibly dangerous, organic vapors. However, in the event of a rupture in one of the fuel elements, radiokrypton and -xenon would contaminate the decomposition gases and complicate the disposal problem. Delay line systems containing activated carbon have been investigated for adsorption of the fission gases for periods long enough to allow decay. The hydrocarbon gases are also adsorbed on the activated carbon, and the presence of these adsorbed hydrocarbons necessitates larger amounts of carbon for adsorption of the fission gases. For large reactors, removal of the hydrocarbon decomposition gases has been recommended in order to reduce the size of the delay line required. Experimental work is needed to develop procedures for removal of the hydrocarbons by combustion or other methods so as to permit smaller delay lines and thus reduce the overall cost of disposal of waste gas.

Other problems of waste disposal include waste water from process steam jet ejectors, solid ash from high boiler combustion, and solids generated by removal of particulate matter from coolant. These wastes probably constitute no unusual problems and can be handled by known methods.

4 Additional Development Work Required on Coolant Processing -Because of the many operations required for coolant processing, the overall facility, including the numerous auxiliary items of equipment, becomes very complex Even though automatic operation is possible for most process units the costs of operation and maintenance of such a complex system will probably be high. Therefore, development work to combine and integrate the various operations into a simpler system would be worthwhile. For example, the single operation of distillation is capable of performing the three functions of high boiler removal, particulate matter removal, and degassing Unfortunately, however, the flow rates considered necessary for the latter two functions are considerably higher than for the former function. Nevertheless, it is possible that conditions might be found under which these operations could be combined into a single step. If not, it should at least be possible to integrate some of the auxiliary units into a simpler system

Consideration should also be given to the possibilities of eliminating the need for some of the process operations. Assuming that particulate matter results only from introduction of iron corrosion products into the coolant, it is possible that the cost of avoiding corrosion by reactor designs which eliminate atmospheric contact with coolant might be comparable with the cost of the required cleanup equipment Work is needed to determine whether insoluble organic material will form in the absence of corrosion products before this alternative could be considered.

The necessity for degassing is largely due to the possibility of water leakage into the coolant. The additional cost of leak-proof heat exchangers should be compared with the cost of degassing.

5. <u>Coolant Reclamation</u> - The cost of replacing the decomposed terphenyl coolant with fresh coolant is an important factor in the operating costs of organic reactors. In addition to the cost of the coolant (current market price 15 cents/lb) there is an estimated cost of 1.5 to 1.8 cents/lb for disposal of the high boiler by burning. These costs provide a large incentive for developing a process for regeneration of the high boiler into useful coolant.

There are two general types of reclamation processes under study. One is based on physical methods, which effect a separation of high boiler into higher and lower molecular weight fractions. For example, it appears possible to separate and reject those polymers having molecular weights above 460 (hexaphenyl), so that the lower molecular weight fraction might be reused as coolant. One of these processes, "solvent distribution," has been demonstrated successfully on a laboratory scale. Approximately 60% yield of the lower molecular weight fraction can be achieved. In addition to this method, both Monsanto and AI are studying molecular or highvacuum distillation as a means for effecting such separations. These physical methods may be applied to either the separated high boiler or to the coolant itself.

The other type of reclamation process is based on chemical transformations, which are aimed at either converting the polymers into lower molecular weight compounds or reducing the viscosity of the polymer without change in molecular weight. At least five different chemical methods have been studied with varying degrees of success. The program is currently in the small bench-scale stage of investigation. The experimentation has been of an exploratory nature to screen out those methods worthy of more intensive study.

Chemical methods which appear promising on the basis of preliminary experiments are hydrocracking, partial reduction, and redistribution. In hydrocracking studies, yields of products having physical properties similar to terphenyls have ranged above 60%. However, coke formation on the catalyst is a problem which must be solved before the process could be usable.

Experiments on the partial reduction of high boiler with hydrogen at high pressure indicate that substantial reduction in viscosity can be achieved. Preliminary irradiation experiments indicate that the products, when added to terphenyl, may impart some resistance to radiation damage.

Studies on the redistribution reaction have shown that terphenyl can be reacted with benzene to yield diphenyl as product. However, the same technique applied to high boiler rather than terphenyl has not yet been successful because the thermal decomposition rate was too high relative to the redistribution reaction rate.

The research which has been carried out on chemical methods of coolant reclamation has been devoted almost exclusively to treatment of the high boiler after it has been separated from the coolant. It is possible that certain chemical treatments, such as partial reduction or redistribution reactions, might be more effective when applied to the whole coolant rather than to separated high boiler. For example, the continuous partial reduction of a coolant side stream might be able to maintain low viscosity and low (room temperature) melting point; or a similar side stream might be treated by the redistribution reaction, using benzene to permit low-cost benzene to be used as coolant makeup material.
In order to estimate the value of some of the potential coolant reclamation methods, the program should be coordinated with a "coolant management study." The latter is desirable because of the complexity of the coolant mixture after irradiation and because of the complex interplay of factors which affect the economics of operating power reactors. For example, the effect of coolant viscosity on heat transfer must be balanced against the cost of providing low viscosity by high boiler removal or other treatment of the coolant. A coolant management study would seek to establish the most economic coolant makeup material - terphenyl, diphenyl, or mixtures of these. It would also attempt to establish the most economic lower limit for the molecular weight of the high boiler fraction, that is, whether the separation of high boiler from coolant should be made at a boiling point above terphenyl, quaterphenyl, or hexaphenyl. Without such a study it is not at all obvious that the physical methods of coolant reclamation, such as solvent distribution, can accomplish any improvement over the current practice of high boiler removal and disposal. The work on coolant reclamation should not be directed solely toward regeneration of high boiler, but should be closely integrated with the treatment of the whole coolant.

Since the process for production of terphenyl (or biphenyl) is a simple one, any coolant reclamation process must also be simple to be competitive with virgin terphenyl. It is only for large power reactors that the costs of coolant reclamation could conceivably be low enough to justify installation of a reclamation plant for high boiler alone. However, a plant at one reactor site might also serve for other nearby reactors. The economics might be more favorable if the coolant treatment process produced other benefits, such as lower melting point or improved resistance to radiation.

Preliminary estimates of costs for the various potential reclamation processes would be desirable, even before loop studies of reclaimed coolant are made and certainly before any pilot-plant work is undertaken. The potential gain from the research and development on polyphenyl coolant reclamation should be weighed continually against the progress in the development of new, cheaper coolants.

D. Fouling

Because of the thermal and radiation decomposition of organic coolants there has always been concern about buildup of a solid polymeric material on the surfaces in the reactor system, particularly on the surfaces of the fuel elements. Because both radiation intensity and temperature are maximum there, it would be expected that this is the location of maximum buildup. In addition, these surfaces are the most sensitive to such solid material formation because of the heat transfer requirements. Initial results during heat transfer experiments and during first operation of OMRE were encouraging in that only thin films, well under 1 mil thick, were formed. However, after the reactor had been shut down and opened to the air for a period, there were indications of impediment to the flow of heat from fuel elements to the coolant. Examination of fuel elements at times subsequent to this showed the existence of varying amounts of solid deposits on the fuel element surfaces. In a few instances there was blockage of coolant channels by such deposits. Analyses of these deposits showed them to consist of polymeric organic material plus varying percentages of inorganic matter. The inorganic content depended on the thickness of the deposit, being of the order of 25% for the heavy (several mils thick) coatings. Its composition was sometimes largely iron carbide and sometimes mostly iron oxide.

Electron microscopy showed the inorganic material to be in the form of small particles, mostly below 1 micron in diameter. Subsequent examination of the coolant showed that it too contained particles, each consisting of an inorganic particle coated with a layer of organic polymer. Analysis of the coolant during Core-II operation of OMRE showed a buildup in the inorganic particulate content which was apparently related to shutdown periods. Following shutdowns involving opening to the atmosphere, a higher concentration of the inorganic material was always noted. During normal operation this ash content decreased, presumably due to removal in the bypass distillation equipment used to remove the high boiling fraction and by deposition in the system. From September, 1959 to the end of April, 1960, the ash content varied between about 70 ppm and 220 ppm.

The similarity between the deposits on fuel elements and the particles in the coolant led to the belief that the mechanism by which the films were formed was the deposition of existing particles in the coolant, followed by the buildup of polymeric organic material in spaces around deposited particles. There seemed to be correlation between rate of film buildup and particulate concentration in the coolant. The rate of film formation was strongly increased in the radiation field and apparently slightly increased by increasing temperature, at least for the range of temperatures encountered in OMRE. Most of the fuel elements consisted of a stainless steel-UO2 dispersion, jacketed with stainless steel. In addition, there were some experimental fuel elements composed of uranium-molybdenum alloy, clad with aluminum (alloy 1100). All of the instances of severe fouling occurred on the stainless steel, highly enriched fuel elements. It has not been clear whether this difference in performance is a result of the specific influence of the material on which deposition occurs, or of the difference in surface temperature and nature of the radiation which is present for the two cases. Because the temperature has appeared to be a rather unimportant variable in fouling rate, the choice of explanation seems to be between the nature of the surface and the difference in currents of beta particles leaving the fuel element surfaces. For some time the AI people felt that the important effect was the magnitude of the beta current, and that the difference in the nature of the metal surface was not important. More recently however, they have come to believe that aluminum is not fouled as rapidly as stainless steel.

At the present time the relative importance of these two variables must be considered as not determined. The Canadians are planning (spring, 1961) to put into their in-reactor organic loop some experimental fuel elements which should clearly indicate how much influence the beta current has on film deposition.

A number of experiments have been performed in loops and in static systems containing heaters. The static tests performed by AI are called pyrolytic capsule fouling tests (pcft). Many of these have been run, sometimes using OMRE coolant and sometimes using synthetic mixtures. The amount of deposit formed on the hot surface has shown the same kind of dependence on variables as has seemed to be the case for fuel surfaces in OMRE. In these tests, fine particles caused more rapid growth of deposit than coarse ones (at constant ash content), and lowering the particulate concentration decreased the amount of fouling deposit. Largely as a result of these tests, AI has estimated that film built up in the reactors with which they are concerned will be below maximum permissible thickness if the ash content of the organic is 10 ppm or less and if the standard pcft produces less than 5 mg of deposit. These specifications were derived from the "average" fouling rate during Core II operation of the OMRE, the average pcft result on that coolant, and the estimated tolerable fouling rate in reactor.

These criteria might not be sufficient to ensure adequately low fouling rates in reactor. The specification of ash content seems likely to be of limited value. The smaller particles have the smaller content of inorganic constituents, suggesting that the smallest particles might have very little or no ash content. Yet tiny particles do cause rapid fouling, as judged by the pcft. It is apparently not known whether the rapid fouling occurs because there is a relatively large amount of the finest particles, or because they deposit (and stick) more rapidly than do the larger particles. In any case, so long as the correlation between in-reactor fouling and the pcft result is good, the specification of less than a 5-mg deposit in the standard test should be valuable.

All studies have indicated the importance of maintaining low concentration of particulate matter in reactor coolants. For this reason OMRE is being fitted with two filtering systems and a centrifuge, all for the purpose of maintaining particulate concentration at a low level. It remains to be demonstrated, first, that this particular reactor system can be maintained at the low particulate concentration in the coolant, and, second, that if this is done the film buildup will be essentially negligible.

Because of the amount of inorganic material in the coolant in OMRE, all the particles present seemed to have inorganic nuclei, and this would suggest that the particulate buildup would be markedly reduced by avoidance of access of air to the mild steel system. However, there has always been a "normal" water content of terphenyl coolants, and it can be anticipated that in some cases there will be continuous requirement for removal of the product of corrosion of steel by this water. The magnitude of the anticipated buildup of purely organic particles in the coolant is not yet known. Presumably such particles would also cause fouling, although there has been no experience by which to judge. In any event, there is reason to be optimistic that systems with very low particulate contents will cause very little film buildup. In addition, the aluminum which is chosen as the fuel jacketing for all future reactor designs is perhaps resistant to particulate film buildup in organic systems. These things suggest that the fouling problem can be managed by means being developed. This remains to be demonstrated.

It is believed that the relationship between fouling rate and the size and composition of particles has not been established. It seems important to know this, most particularly if the means now being developed and put into use do not provide satisfactorily low fouling rates in reactor. A research program with the objective of determining this relationship is recommended. In order for such a program to be effective it will probably be necessary to develop a reliable method of sampling and of analysis of particle size.

E. Heat Transfer and Fluid Flow

Introduction - The function of an organic fluid in a reactor 1. system may be to moderate neutrons, to cool the reactor core, and to serve as a heat transport medium. In performing these tasks, the physical characteristics of organic fluids with regards to hydrogen content, low vapor pressure, and low corrosion rates in low alloy steel systems play important roles in producing a system of low capital cost. As an aid in reducing capital and fuel-cycle costs, it is natural to strive for high performance. In an organic-cooled reactor, the maximum thermal efficiency is restricted because of the upper temperature limit of organic fluids, and this imposes a limit on power cost reductions through increased thermal efficiency. In other reactor and fossil-fueled power plants the maximum thermal efficiency is expected to continue to increase as technological advances are made, and lower power costs should result. Therefore, it is extremely important for an organic system to strive for maximum performance in order to counterbalance the advantages and advances of other systems.

2. <u>Current Status</u> - The removal of heat from a reactor core through the use of an organic coolant has been successfully demonstrated by the operation of the OMRE. In this reactor, cooling by forced convection is accomplished by pumping an organic coolant between fuel-bearing plates. This method of extracting heat is conventional within reactor technology and presents no unique heat transfer or hydrodynamic problems because an organic coolant is used. The power capability of an organic-cooled reactor core can be determined by established methods of analysis for the case of singlephase, forced-convection cooling The information required for a heat transfer analysis consists of power distributions of the operating core, estimates of "hot spot factors," flux variations within fuel clusters, data on physical properties, core geometry data, and correlations for predicting pressure drop and the film coefficient of heat transfer. Given all these items, reactor technology is sufficiently advanced to determine the power capabilities of an organic-cooled core provided little or no fouling occurs. The difficulty connected with analyzing the heat transfer potential of the core stems from the lack of a firm understanding of the allowable design limits for organic-cooled reactors. Fouling as a function of fluid and surface temperatures, radiation, coolant velocity, coolant composition, and channel geometry must be determined in order to predict confidently the power capability of an organic-cooled reactor core.

Current designs are based on negligible fouling, but the coolant and surface temperatures of each design are substantially different These conditions are listed in Table I.

Table I

DESIGN LIMITATIONS OF OMRE, EOCR, OCDRE AND PIQUA

| Reactor | Fuel Cladding | Coolant Temp, °C | Maximum Surface Temp, °C |
|---------|------------------------------|------------------------|-----------------------------------|
| OMRE | Stainless steel and aluminum | 315-330 | 400 |
| EOCR | Stainless steel | 260-275 | 455 |
| OCDRE | Sintered aluminum powder | 320-370 | 480 |
| Piqua | Aluminum | 273-302 | 400 |

(Coolants: Mixed Terphenyls with 30% HB)

The OMRE has experienced gross fouling at the conditions shown in Table I, but the fouling has been attributed to particles in the coolant. At the present time, there is no conclusive proof that fouling will be avoided at the OMRE conditions in spite of the fact that the OMRE operated for 950 Mwd with Core I with very little fouling. The OMRE has not operated for a sufficiently long time under equilibrium conditions to ascertain long-term effects. At the present time, considerable effort is being expended to prove that fouling will not occur at the OMRE, EOCR, OCDRE, and Piqua conditions and, therefore, the conditions cited in Table I cannot be considered as firmly established, but rather as goals which it is hoped organic technology will attain. With regard to the analytical determination of the power capability of an organic-cooled core, the most important requirement for the analysis is the prediction of the film coefficient of heat transfer, h. At the present time, h for irradiated coolants can be found within $\pm 25\%$ through the use of the Dittus-Boelter correlation. This correlation can be used for finned plates as well as for flat plates and round tubes.

Pressure losses within a system using forced convection can be predicted by using standard engineering methods.

Vapor blanketing (burnout) is not a problem at steady-state conditions in reactor designs that utilize single-phase, forced-convection cooling. Burnout correlations are necessary, though, for evaluating the hazards associated with operating a reactor. Burnout correlations are available for pure fluids. There is a definite need for burnout studies, however, with complex, multicomponent, irradiated organic coolants in order to properly assess the hazards associated with an organic-cooled reactor.

The heat transfer coefficient for irradiated coolants can be predicted within $\pm 25\%$, and this is within the same range of accuracy of other fluids used in reactor technology. Since organics are poor heat transfer media, there is an incentive in continued work to increase the reliability of the predicted heat transfer coefficient.

Since the performance of cores operating at high temperatures is greatly affected by the degree of coolant mixing that occurs within a fuel subassembly and very little experimental work has been done on this problem, accurate predictions of core performance are not possible. Work is needed in this area to provide a basis for judging the degree of mixing that will occur in fuel subassemblies.

In conclusion, it can be stated that the current status of organic-cooled reactors is limited to the forced-convection regime in which no changes in the fluid phase occur. Adequate analytical information is available to predict the power capability of an organic-cooled reactor, but it is necessary to establish the operating limits of fuel spacing, velocity, mixing, coolant temperature, and surface temperature to control fouling.

While adequate data are available for analysis, there still is a need to define better the accuracy of the film heat transfer coefficient, burnout, the hot spot factors, and the method of applying the hot spot factors to the analysis of the reactor core.

3. Problems of Advanced Concepts - The trend in organic reactor technology is toward the use of a coolant phase change within the core to increase performance and, therefore, to lower costs. This is possible

because higher core power density and coolant temperature are permissible when a coolant phase change is employed as a heat transfer mechanism.

In a design utilizing an organic phase change during operation, consideration must be given to the heat transfer and hydrodynamic problems associated with an organic phase change because burnout of the heat transfer surface and flow instabilities may result. Coupled with these phenomena are the problems associated with the formation of voids within the coolant. The study of these items with proper consideration to geometry and radiation is complicated by the very nature of an irradiated, multicomponent organic. It is expected that the establishment of methods of predicting heat transfer and hydrodynamic behavior for situations directly applicable to an organiccooled reactor will be very difficult.

4. Current Research and Development Programs - Currently, AI is studying the heat transfer and hydrodynamic problems associated with their concept of an organic-cooled-and-moderated reactor. The paramount effort related to heat removal is to understand and control fouling. Most of the basic mechanism work is being done out of pile. Essentially every heat transfer and hydrodynamic investigation will yield some information on fouling as a byproduct.

AI is continuing its studies in out-of-pile facilities with forcedconvection cooling in which no phase change occurs to determine the effects of fouling and other pertinent parameters on the film coefficient of heat transfer. This work should establish a good design correlation and help to explain some of the observed anomalies in the OMRE bypass heater tests.

Additional work is under way in the local boiling and boiling regimes aimed at providing data to be used in evaluating hazards in a single-phase, forced-convection-cooled power reactor. Void data will also be obtained in this work.

AI plans to use the loop facilities of the EOCR for determining the effect of radiation on fouling and on heat transfer mechanisms with and without a coolant phase change. Although work should be done with irradiated coolants and the influence of radiation should be investigated, it is not expected that a radiation environment will affect the heat transfer mechanism to any significant degree.

The effort of Atomic Energy of Canada Limited (AECL) in the field of heat transfer and hydrodynamics is devoted to investigating fouling, local boiling, and critical heat flux characteristics of Santowax OM with 30% HB for the OCDRE project. Work will be done in-pile to determine allowable geometrical spacings and fouling. These tests are not sophisticated heat transfer tests, but applicable heat transfer information will be obtained. An out-of-pile heat transfer loop is also being used to study fouling; however, most of the OCDRE heat transfer and hydrodynamic studies will be done under AECL contract with the Canadian General Electric Company (CGE) at Peterborough, Ontario, Canada.

CGE plans to investigate forced convection at Reynolds numbers in excess of 2.5×10^5 , the onset of local boiling, burnout heat flux, and fouling. The work is concerned primarily with the OCDRE conditions, but will be done in out-of-pile facilities.

CGE is mainly concerned with the effects of gas content, of individual organic constituents, and of foreign material and water content on the onset of local boiling and the burnout heat flux. There are no plans to obtain void data. Geometrical variations, such as eccentricity of fuel, skewing of fuel rods, and spacer, will be investigated. All the work will be done with unirradiated Santowax OM with 30% OMRE HB. In all the tests, CGE plans to measure the physical properties of the coolant.

Phillips Petroleum Company (PPCO) at Idaho Falls, Idaho, is responsible for the operation of the Experimental Organic Cooled Reactor (EOCR) and the experimental loops associated with the reactor. The reactor will be a helpful tool for developing organic reactor technology. At the present time, PPCO does not have an established program for studies of heat transfer and fluid flow, but it is recognized that those interested in developing organic technology will make full use of this tool. PPCO will have a loop available in the EOCR for investigations in the field of interest.

The EOCR will provide information on fouling, coolant technology, fuel elements, and also heat transfer. The EOCR could be used effectively to establish the influence of irradiation on heat transfer and hydrodynamic mechanisms, and certainly some limited in-pile studies should be made.

The Massachusetts Institute of Technology (MIT) program is primarily concerned with coolant technology studies. An in-pile loop for irradiating a coolant will be operable soon in the MIT reactor. This will be the first US organic in-pile loop to operate since 1958. The loop will be used to study coolant behavior by using Santowax OMP and allowing decomposition to occur without changing or processing the coolant. At a later date, it is planned to study one of the more promising inexpensive coolants, such as alkyl phenanthrene.

In conjunction with the coolant technology studies, out-of-pile heat transfer data in the forced-convection regime will be obtained from a round tube. These tests should be helpful to define better the heat transfer film coefficient. European organizations are also planning to support heat transfer investigations. The DOR project in Denmark is a study and research and development program for large heavy water-moderated, organic-cooled power reactors. A heat transfer loop is available, and vibration studies of fuel rods in water have been made. Flow distribution problems are of concern in the DOR project and will be investigated analytically and experimentally.

The EURATOM, ORGEL project places its emphasis on developing adequate information to decide on the most promising organic concept. Studies of coolant mixing within fuel bundles have just started. Two small out-of-pile heat transfer loops will be used for measuring heat transfer coefficients in rod bundles. Burnout and fouling studies will also be conducted in another loop now under construction. A materials testing reactor facility, ESSOR, will be built by ORGEL for evaluating heavy watermoderated, organic-cooled reactors. This facility will probably be used for heat transfer and fluid flow studies of closely spaced rod bundles within a pressure tube.

The PRO project in Italy involves plans for a 30-Mw(t) organicmoderated-and-cooled reactor. The reactor is similar to the US facility in Idaho, EOCR. The Italian reactor will contain an 18-cm diameter loop and a 2.5-cm diameter loop. Research and development work is planned on fouling, heat transfer, fin design, and nucleate boiling.

5. Required Research and Development - It should be realized that the research and development work that has been done in the past and is planned for the future is generally associated with specific reactor concepts. There has not been a general research and development program to develop organic reactor heat transfer and fluid flow technology over a broad range of possible conditions of reactor operation. An extensive program is necessary to make organic technology effective.

a. Physical Properties of Organic Coolants - In any heat transfer and fluid flow investigations aimed at correlating data for reactor designs, it is imperative that sophisticated measurements be made of the physical properties of the organic coolant used in the tests. It would also be advantageous to identify the coolant composition in a detailed manner rather than by specifying the high boiler content, but this is considered impractical because of the great deal of effort required.

For forced convection, the physical properties of density, specific heat, thermal conductivity, and viscosity are important. Data are available for unirradiated and irradiated terphenyls, but the data for irradiated coolants are too meager. Further work is considered necessary. The successful understanding of organic phase-change heat transfer may depend on such physical properties as vapor pressure, gas solubility, and surface tension. Adequate data of this type for irradiated terphenyls are not available.

b. <u>Single-phase Forced Convection</u> - There is a need for continued work to refine the correlation for the heat transfer film coefficient in order to improve reactor performance. Since there is a tendency to use finned fuel elements in organic-cooled reactors, future studies in the singlephase forced-convection regime should concentrate on this type of geometry. An extension of data to higher Reynolds numbers is also warranted.

Phase-change Heat Transfer and Fluid Flow - Extensive с. investigations into the heat transfer mechanism for the formation of voids and hydrodynamic phenomena associated with an organic coolant phase change are definitely required to develop the full potential of organic-cooled reactors, using either an indirect steam power cycle or a direct organic power cycle. The areas for study are local boiling, bulk boiling, burnout, and the fluid flow characteristics associated with a phase change. It is expected that a thorough understanding of this phase of organic reactor technology will be difficult and time consuming because of the complicated nature of irradiated organic coolants and because of the many variables believed to be important Coolant composition, fluid and surface temperatures, surface roughness, and the presence of foreign substances are expected to affect characteristics of organic coolants during a phase change. Another item that should be checked is the effect of radiation on the mechanisms of heat transfer for a phase change. Geometry is expected to be very important, but the effects of heat flux levels, power distribution, and time should also receive attention. Studies involving these parameters are necessary to improve the performance of organic reactors.

6. <u>Summary</u> - Current US commercial organic reactor technology is limited to designs of single-phase forced-convection-cooled reactors because of a lack of data and experience applicable to high-performance systems in which a coolant phase change is utilized for increased heat transfer ability. Within the realm of current technology, work is necessary to define better the heat transfer film coefficient, to determine the degree of mixing within flow channels, and to determine the design limitations in order to increase the power capability of organic-cooled systems.

In order for organic-cooled reactors to achieve and maintain an economic position, increased performance is mandatory. This could be accomplished by utilizing a coolant phase change. As a consequence, investigations applicable to the heat transfer and hydrodynamic conditions of organic-cooled reactors for indirect and direct power cycles are necessary. Although work is currently being done, the program is too restrictive and will require a long time. Greater effort must be expended to obtain adequate design information for organic reactors utilizing a mechanism involving a phase change of the coolant.

VI. MATERIALS

The general requirements for materials in organic-cooled reactors and reactor systems are that they be resistant to corrosive attack by the organic coolant, that their dimensional stability in operation be good, and that they have and retain reasonable mechanical properties.

Resistance to corrosion is required to avoid penetration of container or barrier materials. There is also a more stringent requirement than this. It is necessary to avoid the formation of too much solid corrosion products which might be carried into the coolant stream in the form of particulates. The importance of such particulates in determining the fouling characteristics of the coolant has already been described.

Fuel materials especially are subject to conditions which cause dimensional changes and swelling due to the formation of fission products. The requirement for stability is in no way peculiar to the organic reactor system; the important parameter is the operating fuel temperature.

Requirements with respect to mechanical properties are in many respects similar to those for other types of systems. Materials must be fabricable, and must have and retain sufficient strength and ductility at the temperature and under the irradiation conditions of each application.

A. Fuel Elements

1. Fuel - The fuel materials at present under serious consideration include uranium alloys and uranium dioxide. The relatively low uranium atom density and low thermal conductivity are unfavorable features of UO_2 as fuel for thermal reactors. For this reason Atomics International has been interested in developing uranium alloys with acceptable properties. Although the alloys which have been considered do not seem to be the choice of the AI people with respect to ultimate potential, they are felt to be good fuel for specific applications.

Uranium reacts very little with Santowax. In a number of tests run for one week at 200°C, apparently very little attack was caused by the water present in normal amounts as an impurity. The amount of oxidation corresponded to the amount of oxygen that was in the system. Reaction rates have not been measured, and no tests have been performed at temperatures higher than 200°C. Higher temperatures in a reactor radiation field, however, apparently do not cause substantial reaction. One of the $U-3\frac{1}{2}$ w/o Mo elements in OMRE had channel blockage such that some of the aluminum cladding melted; the resultant contact of the uranium alloy with the coolant caused no apparent reaction, and there was little coolant contamination.

Among the alloy systems which have been considered for this application, uranium-molybdenum seems to show the most promise for high-temperature stability. Tensile properties of binary, ternary, and a few quaternary alloys have been obtained at molybdenum contents of 1.5, 3.5 and 10% (by weight). At temperatures higher than 550°C ultimate tensile strength and yield strength increase as the molybdenum content is increased. At this temperature and below, the strongest alloys (ultimate tensile strength) contain 3.5% Mo; a quaternary containing 0.1 Al and 0.3 Si was the best, followed by a ternary containing 0.1 Al. At 600°C, these alloys had lost approximately half of their tensile strength and had become weaker than the 10% Mo binary alloy. Considering the yield strength, which might be more important than the ultimate tensile strength, the 10% Mo alloy was superior between 480 and 550°C. The choice of alloy probably will depend on fuel temperature. At the highest temperatures for organic reactors, it is likely that the alloys with higher molybdenum content will be desirable, and perhaps necessary.

Within the range of permissible temperatures and total burnup, strengths and ductilities should be sufficient to maintain dimensions and support the fuel element structure. Wrought aluminum alloy jackets are therefore contemplated for metallic fuel.

Uranium dioxide has been tested in Santowax at 400° C with a 50 to 100 psi nitrogen overpressure. Small weight gains occurred, apparently due to the absorption of some of the organic. These amounts of organic material could be removed by treatments with solvent, so that final specimen weights were about the same as before initial exposure. Equally instructive was the behavior of the fuel during an in-reactor defected fuel test at Chalk River. There was no indication of interaction between the coolant and the UO₂, even though a considerable amount of coking occurred and the fuel temperature must have been high.

The stability of UO_2 to irradiation has been studied in considerable detail. There is little growth and little release of fission gases at elevated temperatures. This explains the general interest in this material as fuel for organic reactors.

Characteristic of its properties is high strength at elevated temperatures, but with accompanying brittleness. This brittleness, together with a low heat conductivity, makes it necessary to expect that UO_2 will fragment during operation, and perhaps especially during startup and shutdown. For this reason it does not seem possible to use monolithic UO_2 of significant thickness if the dimensions and configuration of the fuel element are to be retained. In such a case it would be necessary to use a jacketing material. Consequently, the use of wrought aluminum sheathing is not contemplated at temperatures of interest in organic reactors. Instead, aluminum products with higher strength, and particularly with better creep resistance, are being developed.

2. Jacketing - Among fuel-jacketing materials, interest has centered on metals of low neutron cross section. It is apparently believed that the use of steels in this application would make the reactor type economically unattractive because of their neutron absorption. The metals of general interest include beryllium, magnesium, zirconium, and aluminum. Beryllium has excellent corrosion resistance, but its technology is insufficiently advanced and its cost is high. There is apparently no interest in it at AI. Magnesium is badly corroded at temperatures of interest, apparently by the water content of the organic. Zircaloy-2 has shown considerable hydrogen uptake during exposure to Santowax at 400°C. After 1000-hr exposure, samples contained 2800 ppm hydrogen when a hydrogen cover gas was used and 460 ppm in the presence of argon. Anodizing specimens before exposure reduced the quantity of hydrogen absorbed by a factor of the order of 10 in tests of this duration, but the possibility of breaks and defects in such coatings (and subsequent hydrogen embrittlement) has caused the AI people to be pessimistic about the possibility of using zirconium-base materials. They apparently are planning no further work on these materials.

The situation is not as bad at lower temperatures. Zirconium and an alloy (containing Al, Sn, and Mo) were apparently in reasonably good condition after 5000 hours in OMRE coolant at 315 to 345°C. Samples exposed in an in-reactor loop at Chalk River showed no hydrogen pickup by Zircaloy-2 at 260°C. The possibility of using zirconium-base alloys cannot be entirely ruled out at this time, although prospects do not look good. Apparently the Canadians are planning to attempt to find means of making the behavior of zirconium alloys acceptable.

Aluminum has shown good corrosion resistance in laboratory exposure to Santowax R, during exposure in the NAA-20 loop (MTR), and in the OMRE both as fuel element jacketing and in the form of corrosion specimens. Laboratory tests at 400°C for one month have shown a sensitivity to water content of the organic. There has been little corrosion damage at normal water contents (less than 200 ppm). At 1000 ppm water, a weight gain indicating corrosion of the order of $\frac{1}{4}$ mil was observed. This increased to about a 2-mil penetration in one month at 2000 ppm water.

The relationship between water content and corrosion behavior has not really been determined. since the effect of exposure time has not been investigated. At reactor operating temperatures it is probable that amounts of water above some critical concentration will cause deep penetrating corrosion attack of both wrought and powder metallurgy aluminum. This critical water content might very well be below the 1000 ppm which has been used in laboratory tests. During exposure to water, the rapid attack is observed to occur only after relatively long exposure at temperatures which might be called threshold in terms of causing rapid penetrating attack. For this reason it is quite difficult to form an opinion about an upper permissible limit for the water content of organic coolants. The water present in the OMRE coolant seems to have been below such a maximum permissible level, as judged by the behavior of 1100 aluminum during 28 months of exposure. If the water concentration in a power reactor coolant can be kept at or below the level which has existed in OMRE, there is good reason to feel that the amount of corrosion will be acceptable.

The effect of coolant velocity on corrosion and/or erosion has been studied only in laboratory tests, where spinning samples gave calculated speeds up to 15 ft/sec. When no water was added to the organic liquid, there was generally no significant effect of the flow rate. In the presence of 1000 ppm or more water, consistent increase in the amount of corrosion (in one month) was observed as the speed of flow was increased.

The influence of particulate concentration on this effect has not been studied directly. The acceptable behavior of the aluminum cladding on OMRE fuel elements suggests that behavior is satisfactory at the flow rate (16 ft/sec) experienced there.

It is unlikely that there is a problem of dimensional stability for aluminum jacketing materials, either the weak wrought 1100 aluminum as bonded to a uranium alloy fuel, or a sintered powder product aluminum jacket containing free-standing UO_2 pellets. Irradiation experience has shown no cause for concern.

Because wrought aluminum alloys are weak at elevated temperatures, the contemplated sheathing for UO_2 fuel is a product produced by the techniques of powder metallurgy. The basic method is to compact aluminum powder into shapes and to fabricate them into final product. Certain extruded shapes and plate are believed to be commercially available: these have properties which are probably sufficient for use up to about 480°C (this depends on design stress). For a few years now, Alcoa and a few other companies have been developing processes for making good quality tubing of this general type. Although many pieces of tubing with acceptable dimensional tolerance, surface finish, and freedom from inclusions have been made, it cannot be stated that one could order and expect to receive with normal delivery time any substantial amount of completely satisfactory tubing. Efforts are under way at several locations to develop manufacturing procedures to a point which will allow predictable and reproducible quality tubing. Both AI and the Canadian General Electric Company are sponsoring such work at the Aluminum Company of America (in the form of purchase orders), and CGE sponsors this kind of development at two Canadian concerns. In addition, development work with similar objectives is being carried out by the Montecatini Company in Italy and by the Aluminum Industrie AG in Switzerland. Work with the same objective has apparently been started at the Research Center of the Trefileries et Laminoirs du Havre, France.

In consideration of the fact that some good tubing has been manufactured, it is reasonable to suppose that the amount of effort now being applied will be sufficient to make possible the routine production of satisfactory material. It is difficult to estimate the time required for this development; it is hoped that the problem will be solved in the reasonably near future (CGE reports it will be necessary to order tubing for OCDRE at the end of 1961 in order to meet the proposed construction schedule).

Based on limited experimental work performed within the last two years at the Armour Research Foundation, it is quite possible that powdered aluminum product tubing can be developed with substantially higher strength at elevated temperature than is possessed by Alcoa M257, the reference material so far chosen by both AI and CGE. The cost of stronger material might be greater, because of additional powder processing before fabrication.

The aluminum materials being worked upon are sufficiently ductile for the application and have shown no indication of loss of properties upon irradiation (although there have been little or no quantitative measurements).

3. <u>Bonding</u> - It is planned to clad uranium alloy fuels in aluminum. It is highly desirable to bond the aluminum sheath to the uranium alloy fuel, for two reasons. A temperature drop across the interface would increase the fuel temperature and is to be avoided. In the event of perforation of the aluminum, it is essential to minimize the buildup of polymerized coolant in a layer along the interface.

Unfortunately, direct bonding of aluminum to uranium can be expected to lead to interdiffusion and the formation of brittle compounds at operating temperatures. For this reason, a diffusion barrier is necessary, and considerable effort has been expended in developing such barriers, both for organic reactors and for others. Nickel can be used for this purpose if the temperature is low enough, and the methods of application and bonding have been worked out. The maximum temperature of use appears to be below 400°C, since a one-mil-thick layer disappears in six months at that temperature. One-fourth of a mil diffuses into the uranium alloy during only one hour at 540°C.

There are better diffusion barriers than nickel. Niobium is apparently quite good. During 4.2 months at 540°C, there was no observable diffusion of aluminum into niobium; after 7 months at the same temperature, $\frac{1}{4}$ mil had diffused. AI is now attempting to develop a method of applying a niobium layer. Application to the 10% Mo alloy is of most interest, since it is believed that it can operate at higher temperatures than can lower alloys. Relatively little success has been achieved to date. Some other metals are also promising as diffusion barriers. The ORGEL project (EURATOM) is sponsoring work on vanadium and chromium as well as niobium. They have looked at zinc, copper and nickel, but they believe these are not as good as the three chosen for study.

B. Structural Materials

The materials of construction of reactor vessel, mechanisms, piping, and external components are basically steels. The corrosion resistance of a number of steels ranging from straight carbon steel to stainless steel has been shown to be good in the organic coolants contemplated for use. The corrosion rate of mild steel (1018 or 1020) is of the order of $0.05 \text{ mg/cm}^2/\text{mo}$ (or 0.03 mil/yr), as an average during a 28-month exposure in OMRE at 315°C. Published data do not make it clear how the rate of reaction changes with time. AI people believe that this rate is relatively constant with time, yet laboratory tests of one-month duration showed an average corrosion rate up to about 7 times this value (at 400°C). The significance of these corrosion rates is their relation to the possible entrainment of particulate corrosion product and its subsequent influence on fouling. For example, the coolant in a reactor such as the PIQUA, which has an approximate steel surface area of 15,000 ft² might acquire metal oxide particles at a rate between 32 and 220 grams/day. In the 1520 ft³ of coolant this would correspond to 0.8 to 5.8 ppm/day.

Dimensional stability of the steels should be good and mechanical properties are well known for nonirradiation environments and fairly well known in reactor. There has been an indication of partial embrittlement of carbon steel samples in OMRE. It has not yet been possible to say whether this effect was partly due to hydrogen pickup from exposure to the organic.

C. Research and Development Required

In order to reach what is believed to be the potential of the reactor type for the production of economic power, it will be necessary to develop fuel elements which are capable of high burnup at temperatures as high as the limit of the organic coolant. It will also be necessary that the cost of the fuel elements and the neutron absorption of the materials used be acceptable.

It is generally believed that UO_2 offers the most promise of meeting the requirements for the fuel. If pellets are used, no significant development is required. Because of the cost of making UO_2 pellets, it is quite possible that alternative methods, such as vibratory compaction of powdered oxide in the fuel jacket, or swaging of the powder-filled jacket, would lead to significant cost reduction. These methods should be tried with sintered aluminum powder (SAP) jackets. This work might not be urgently required, because it would not prevent the construction of first model power plants. On the other hand, it might significantly influence later decisions as to whether to continue with development of the reactor type. Atomics International has no current plans for this type of work. As far as is known, only the DOR project (Denmark) expects to pursue this kind of development in the near future; they plan to try swaging SAP tubes onto powdered UO_2 . There are other studies on densification of powders. With the exception of the work of NUKEM (Germany) on UO_2 -carbon mixtures, their interaction and compaction, these studies do not appear to be easily applicable to the production of fuel elements for organic reactors.

At present the use of UO_2 fuel appears to require sintered aluminum powder jacketing. It is imperative that procedures for the manufacture of such tubes be developed. The work at a half dozen or more locations is testimony that all those in the field take this problem seriously. It appears quite desirable to maintain close contact between those working on this problem so as to ensure that progress will be as rapid as possible. There is danger that proprietary rights of some of the companies actually developing the processes will prevent the exchange of information which it is felt is highly desirable; the importance of successful development suggests that some effort be made to overcome this impediment.

In addition to learning the method of manufacture of tubes of material which now can be fabricated into other shapes, it is desirable to work toward the objective of obtaining better high-temperature properties. The yield strengths and creep rates of available material are barely adequate or are inadequate at the highest temperatures contemplated for organic reactors. The fact that some experimental materials having greater strengths and adequate ductility have been made suggests that it is feasible to develop commercial products with improved properties. It is not clear whether present efforts can be expected to carry development in this direction, but such a research and development program is recommended.

The closure of sintered powder jackets has been a serious problem. The development of hot pressing or eutectic bonding will perhaps solve the problem. Atomics International apparently feels that they have been entirely successful, but this has not been demonstrated. Continuation of the work already contemplated at AI and the Canadian General Electric Company seems likely to ensure at least eventual success. In addition, various welding techniques are being developed at AI and at several European sites. Quite possibly, the successful procedure will involve sealing in a plug at the end of the tube, followed by welding around the exposed bond line. The former should provide strength and the latter might ensure leaktightness. This solution of the problem does not seem to have been selected at this time by any of the organizations in this field.

An alternative material for jacketing might possibly be found in a zirconium alloy. Results to date suggest that hydriding of Zircaloy-2 precludes its use at temperatures of interest, probably even with some surface pretreatment. However, there is a considerable variation in the hydrogen absorption, during steam corrosion, caused by the addition of some alloying constituents. Using this result as a guide, it might be possible to develop alloys which would solve the jacketing (and perhaps pressure tube) problem as well or better than does dispersion-hardened aluminum.

If uranium alloys can be made strong enough at temperatures of interest, without using too much neutron-absorbing alloying constituents, it will probably be possible to make all-metallic fuel elements which are better than jacketed UO_2 . This might be true from the points of view both of cost and of fuel worth. To date, work has largely been restricted to the study of uranium-molybdenum alloys. The burnup potential of these alloys should be determined. Both AI and Saclay wish to do this, although the AI program has been suspended for lack of funds. This program should be supported, with any changes which are felt to be needed to ensure that the burnup limits will be established.

In addition, other alloy systems ought to be studied. The hightemperature properties of uranium-fissium alloys suggest investigation of their potential. The development of new alloys designed to provide strength by the simultaneous addition of small amounts of a number of alloying constituents should be investigated.

Another material of potential interest is the uranium-silicon compound U_3Si . Irradiation experience is felt to be inadequate to judge its potential performance, but its worth as fuel is good. The cost of preparation might be an adverse factor.

Work on other potential fuels seems justified. There has been interest at a number of sites in uranium carbide, and it is desirable to continue this. The development and evaluation of uranium-containing glasses and thorium-containing fuels seems worthy of support, if a uranium density which will not require an inordinately high enrichment appears capable of achievement.

There has been a hint that hydrogen absorption and embrittlement of steels might occur during exposure to (moist) terphenyl mixtures in the radiation field of a reactor. It seems essential to learn whether or not this will occur to a significant extent.

Organic reactors other than the PWR type would have materials problems not yet considered here. The behavior of available grades of graphite in organic liquids during irradiation might need to be determined. Methods of making and cladding very small fuel elements might need to be developed and demonstrated (fluid-bed concept). Whether any of these things should be done depends upon the desirability of pursuing the development of different reactor types. It is recommended that evaluations be made of the apparent economic (or other) potential of proposed new types before materials research and development is authorized.

VII. REACTOR SYSTEMS DESIGN

A Physics

Organic-moderated-and-cooled reactors are amenable to physics analysis which closely follow the analytical procedures for the physics of water-moderated reactors. The basic differences between organicmoderated and water-moderated reactors is that the former has a greater age and a greater thermal diffusion length.

At the present time adequate information is available for designing organic-moderated-and-cooled reactors, but there still is a definite need for further physics studies to permit more accurate determination of the characteristics of organic-moderated reactor cores. In particular, age measurements of irradiated coolants and representative reactor core compositions are advisable. It is understood that AI has embarked on this phase of the physics studies and will also measure the relationship of the thermal diffusion length and temperature.

Physics calculations herein refer to studies made by Atomics International on reactors using organic coolants and moderated by

- a organic material;
- b. D_2O ;
- c. graphite; and
- d ZrH

There are a variety of reactor studies with different power ratings, using an alloy of $3\frac{1}{2}$ w/o Mo-U or 10 w/o Mo-U, UO₂, or UC as fuel.

Two-group theory methods have been used for making parametric studies (such as for optimizing the ratio of moderator to fuel for a chosen burnup). Final design and checks of experimental data have been obtained by 16-group CURE or PDQ calculations. For core lifetime or burnup studies, zero-dimensional (no allowance for spatial variation) KINDLE code calculations have been made.

According to AI studies, the most promising reactors are organiccooled-and-moderated ones, in preference to units with organic coolants but moderated with D_2Q_i graphite, or ZrH. The D_2O and graphite-moderated reactors are relatively bulky and yield greater overall cost in mills per kw-hr The ZrH moderator looks favorable when small size and reduced organic ratio are of paramount importance. The fuel materials used in these studies are:

- 1) 3.5 w/o Mo-U;
- 2) 10 w/o Mo-U;
- 3) UO_2 ; and
- 4) UC.

Whenever high burnups are feasible, UO_2 is used. In a D_2O_1 moderated reactor, a unit using UO_2 fuel with 1.6% enrichment yields a burnup of 15,000 Mwd/T; another unit with 3.5 w/o Mo-U(nat)fuel yields 3-4000 Mwd/T exposure.

Use of organic coolant (instead of D_2O) in a D_2O -moderated reactor, resulting in greatly reduced operating pressure, is a decided advantage. To benefit fully from the use of D_2O , however, we should minimize parasitic absorption. In the 3.5 w/o Mo-U alloy, loss of neutrons by parasitic absorption by molybdenum is of the order of that of the coolant. It seems necessary, therefore, to obtain a U alloy with low parasitic absorption, for use in organic-cooled reactors.

Use of $U(nat)O_2$ instead of 3.5 w/o Mo-U(nat) alloy should retain the same thermal utilization, but increase resonance escape probability, and hence reactivity, by about 4%. AI claims that an increase in enrichment to 0.98% in the 3.5 w/o Mo-U alloy and an enrichment of 1.6% in UO_2 will result in burnup exposure of 7,000 Mwd/T, and 15,000 Mwd/T, respectively.

In this connection it is in line to point out to the high parasitic cross section in 10 w/o Mo-U alloy, which has been specified for possible use in AKS reactor moderated and cooled by organic. This amount of Mo would lower reactivity in the core by about 4%. A suitable substitute for this alloy could be 5 w/o fissium-U alloy, if rhodium (present to the extent of 0.29 w/o) could be eliminated from this alloy. The latter alloy would then reduce the parasitic poison in the 10 w/o Mo-U alloy by a factor of 2. Fissium-U alloy has stood irradiation at ~800°C at least as well as the Mo alloy.

The advantages of ZrH as a stable moderator possessing H densities approaching that of water is recognized, preference being given to the powdered hydride, which appears to be superior to the metal hydride from economic as well as nuclear considerations (greater scattering cross sections per zirconium atom as well as more efficient fuel utilization).

Some disadvantages are pointed out, such as the migration of hydrogen from the hydride at temperatures > 540 °C to regions of the core periphery, as the result of radial temperature gradients. Limited knowledge of the properties and cost of the hydride are also pointed out.

In summary, results of AI studies may point to the desirability for developing an alloy of U metal with sufficient stability but with lower parasitic absorption. Also, further studies of reactors moderated by D_2O and ZrH are desirable. Additional work to refine the physics by obtaining better measurements of age and diffusion length is necessary.

B. Plant Layout and Components

In general, pressurized organic-cooled-and-moderated reactors which have been proposed to date have followed design principles already developed for pressurized water reactors. The exceptions to this rule reflect the advantages offered by organic coolants: low vapor pressure at elevated temperatures, and absence of significant corrosion effects on low-cost structural materials, such as carbon steel. These factors have made it possible to design reactor vessels and primary system components for maximum pressures in the 200-300 psi range rather than the 1000-2500 psi range required for economical water reactors, and essentially to eliminate stainless steel as a structural material.

The basic primary system of the organic-moderated reactor consists of the reactor, coolant circulating pumps, steam generators and superheaters, a surge tank, and connecting piping. Side streams lead to the degasifier and to high boiler and particle-removal systems. The primary system pressure is applied and controlled by the degasifier system. The secondary steam system operates at considerably higher pressures than the primary system in order to achieve an economical steam cycle.

A great effort has been made to retain simplicity of design and to keep component requirements within normal commercial practice. For example, single tube sheets are used in steam generators, and the philosophy of permitting some water inleakage is accepted, although not specified. Indications are that the corrosion rate of the carbon steel primary system of OMRE is not significantly affected by changes in water content in the coolant up to 500 ppm. It is felt that normal operation of the degasifier system will remove small amounts of water inleakage without deleterious effects on the plant.

It has been found that components required for organic systems usually fall beyond the normal experience of vendors, despite the available commercial background in diphenyl heat exchangers and petrochemical equipment. Reactor components are generally of larger sizes, and must operate at higher temperatures than are normally encountered. Cleanliness and leaktightness have been particularly difficult to attain.

Some development work appears to be necessary in the field of gasket, packing, and sealing material. Valves with extended packing have been used in the OMRE system, but small leaks at valves continue to be a problem. Furthermore, difficulties have been reported in procuring a seal material for temperatures up to 455°C for the main coolant pumps for EOCR. Although it seems doubtful that combined pyrolytic and radiolytic damage rates will permit operation with present coolants at such high bulk temperatures, the possible development of improved coolants may justify research in this area.

With respect to heat exchange equipment, it is not certain that the approach of using standard components is best on a long-range basis. The direct adaptation to organic fluids of equipment fundamentally designed for water might exact economic penalties of a magnitude not fully appreciated at this time. Compact heat exchangers, in which coolant passage geometries are designed specifically to be consistent with coolant properties, appear to have potential application to organic reactors. By means of novel designs involving fins and other devices having extended surfaces, it might be possible to compensate sufficiently for the poor heat transfer characteristics of organics to make such units economically attractive. Development work in this field should be initiated.

Fuel handling in organic reactors has some problems not encountered in other reactor types. Difficulties are caused primarily by the fact that the coolant in the reactor must be kept above the melting point but below the flash point (about 150 and 190°C, respectively, for Santowax R) while reactor is open, and by the need for washing and further cooling of fuel elements before they can be placed in a conventional water-filled storage canal or pit. A rather complex system for accomplishing such fuel-transfer operations has been devised for EOCR, but extensive testing must be carried out before its success can be assured. Moreover, the EOCR procedure appears to be excessively time-consuming for application to power reactors wherein shutdown time must be minimized for economic reasons. A continuing development program on fuel-handling methods is therefore necessary.

With the exceptions noted, there do not appear to be any fundamental problems which would preclude the development of suitable components for large-sized organic-moderated reactors by relatively straightforward extension of present commercial practices.

Since OMRE, EOCR, and Piqua are the only organic reactors for which equipment has been purchased to date, there still appears to be considerable latitude for reconciliation of initial specifications with actual needs as determined by operating experience. Moreover, it is probable that the accumulation of such experience will permit some increase in power ratings, and consequent reduction in unit power costs, as has been the case with water reactors. It is significant to note that all organic-moderated reactor systems currently under design or construction are overdesigned with respect to temperature and pressure in order to permit experimental raising of initial operating conditions.

In the field of organic-cooled, heavy water-moderated reactors, additional problems include fabricating pressure tubes of a material having low neutron-absorption characteristics, providing thermal insulation between the organic coolant and the moderator, and devising leaktight connections between the tubes and the remainder of the coolant circulating system. Canadian and European project groups concerned with this reactor type are pursuing active development efforts on these problems. A close liaison should be maintained with these groups to insure that applicable solutions reached in these or the more general component problems are incorporated into the U.S. program.

C. Safety Considerations

The physics aspects of the safety of organic-moderated-and-cooled reactors are similar in most respects to water-moderated-and-cooled reactors. The reactivity coefficients for fuel temperature and for coolant and moderator temperatures and voids can all be made negative, provided the reactor core is designed on the under-moderated side and other conditions are met. (The OMRE has a positive coolant-moderator temperature coefficient in some temperature ranges, primarily because the reactor core is over-moderated to obtain maximum radiation exposure of the coolant, for experimental purposes.)

Some consideration has been given to the effect of water additions to organic systems, as could occur with the rupture of a steam generator tube. If an organic-moderated reactor core has a moderator-to-fuel ratio of 2-2.5, the addition of liquid water will increase reactivity, since water is a more effective moderator than organic liquids under current consideration. However, since the moderator-coolant would always be above 150°C when the reactor is under normal power operation, water injected would most probably flash to vapor upon entering the organic stream. Water vapor in the core would behave essentially as a void, and reactivity would decrease.

The non-nuclear effect of water addition on system pressure has been studied in experiments at AI. A quantity of cold water was injected suddenly into a tank of hot organic, in an effort to determine whether a pressure transient of explosive violence could thus be initiated. It was determined that the relatively slow heat transfer from the organic to the water caused the pressure rise to occur over a number of seconds, rather than in a fraction of a second. Such slow rises can be accommodated without danger of system damage.

A pressure-transient loop is under construction at AI to conduct safety studies. The facility includes a fully instrumented flow channel in which the void and pressure drop effects will be measured under conditions of local and bulk boiling in two-phase flow. The test section will later be expanded to obtain the effects of multiple flow channels. Loss-of-pressure and loss-of-flow tests are also planned in this facility. The nuclear effects of core voids are also under analytical study at AI. Mathematical models for use in both analog and digital computers are under development. It is anticipated that the results of these analytical studies will be verified experimentally in the EOCR by means of measurements with void-simulating materials such as Styrofoam.

Since, under reactor operating conditions, organic coolants are used above their flash and flame points (190 and 238°C, respectively, for Santowax R), particular attention must be paid to possible sources of ignition near the reactor primary system. Explosion-proof electrical equipment must be used. Although extreme care must be taken during refueling operations when the reactor vessel is open, it appears possible to carry on these operations with the coolant below the flash point, and the possibility of a serious fire resulting from small system leakages seems remote. Investigations have shown no exothermic reactions of organic coolants with other materials in the reactor. However, there are no data available at this time as to the effect of varying contents of radiolysis products on flash, flame, and autoignition properties of coolants. This must be studied.

Hydrogen constitutes the major fraction of the gas produced by radiolysis in organic reactors. However, since oxygen is not produced simultaneously in the reactor, the danger of gas explosions is perhaps somewhat less than in the case of water reactors.

The toxicity of diphenyl has been studied extensively. It was been judged that the maximum safe concentration for prolonged human exposure is considerably higher than the concentration detectable by odor. However, although coolants currently considered for reactor use are chemically similar to diphenyl, there is little direct knowledge of their toxicities. Moreover, there is evidence that some products of organic-coolant irradiations are of a carcinogenic type, so that further study of toxic properties is essential.

Elements in which high activity may be induced have been excluded from proposed organic coolants. As a result, it has been possible to design primary systems (except the reactor proper) without biological shielding, at a considerable cost saving. This principle has been demonstrated as safe in OMRE even under conditions of fuel failure. However, it remains to be proven whether this design philosophy will remain valid with advanced coolants, possibly containing additives, and with failures of more highly irradiated fuel elements.

Because of the low latent heat of vaporization of organic coolants, a major rupture of the primary system could result in the flashing to vapor of a large fraction of the coolant before the remaining liquid (and the system itself) had cooled below the atmospheric boiling point. This appears to be a serious problem for diphenyl (boiling point ~260°C), although not serious

for the terphenyls (boiling point ~ 370 °C). More complete thermodynamic data must be obtained for the various organic materials and mixtures before this problem can be fully evaluated.

The degree of containment necessary for organic reactor systems is under intensive study. It is evident that until a great deal more operating experience has been obtained with reactors of all types, some form of containment will be required except in remote areas such as NRTS. However, the low working pressures of organic reactors probably eliminate any need for pressure-holding containment.

In the absence of conclusive evidence permitting elimination, pressure containment has been specified both for the Piqua nuclear power facility and for the intermediate-size prototype (50-Mwe) plant now under consideration. It is significant, however, that for the latter plant a containment size of 60 ft in diameter and 78 ft high has been specified. In comparison, the containment for EBWR is 80 ft in diameter and 120 ft high, while the power capability of this plant is probably 25-28 Mwe at best.

An investigation is under way at AI to determine means by which relatively standard buildings can be made sufficiently gastight to contain organic reactor systems safely. This program should be pursued diligently and expanded if necessary to obtain conclusive results at an early date, since minimized containment cost is an important element in the potential economic advantage of organic reactors.

The safety aspects of heavy water-moderated, organic-cooled reactors are similar in most respects to organic-moderated reactors. Differences occur in the nuclear effects. Since in these reactors the organic coolant generally acts more as poison than as additional moderator, the coefficients of reactivity for coolant temperature voids and loss are positive. However, it is expected that reactivity coefficients for fuel and moderator temperature can be made sufficiently large to compensate for the positive effects, at least in the case of slower transients, and to provide a negative overall power coefficient.

The planned construction and operation of the ECO facility, a part of the EURATOM ORGEL program, should contribute important knowledge in this field. Experiments will be directed toward determining the effects of independently varying moderator and coolant temperatures on such parameters as neutron spectrum in a core. Close liaison should be maintained with this group.

VIII. ADVANCED CONCEPTS

Several advanced reactor concepts have been proposed to utilize organic moderators and coolants. These concepts have involved the use of liquid-vapor phase changes for the transport and conversion of energy, or the subdivision of fuel to various extents.

A. Direct Cycle Organic Reactor Systems

More than twenty years ago diphenyl and similar materials were considered as working fluids for fossil fueled power-generating cycles. The use of organic coolants as working fluids in direct-cycle nuclear energy conversion systems has been studied by AI and by the Marquardt Corporation. A boiling organic, direct-cycle reactor system was studied briefly by AI, but was dismissed on economic grounds. In the Marquardt study, a conceptual design was developed for a forced-circulation, diphenylcooled reactor in which a portion of the coolant was flashed to vapor in an external drum and passed through a turbine, regenerative heat exchanger, and condenser. The design also employed fuel material embedded in graphite blocks. These blocks were intended to serve also as moderator, in order to reduce the volume of coolant in the core and thereby to minimize radiation damage to the coolant.

In view of the sketchy information available as to the thermodynamic properties of the various organic coolants and coolant systems under development, it does not seem possible to make a proper evaluation of either of these concepts at this time. The conclusions of the different groups in the field appear to be in conflict on a number of points. For example, AI has rejected the use of boiling organics because of low heat of vaporization and consequent high vapor-transport requirements, although Purdy,(16)in a study for the Chrysler Corporation, has indicated that under certain conditions the turbine exhaust volume flow for a diphenyl turbine, per unit of energy, is about the same as that for a steam turbine. On the other hand, the Marquardt study predicts considerably higher plant efficiencies than appear justifiable from the Purdy work.

Several major suppliers of power plant equipment have recently expressed interest in the use of a direct diphenyl cycle in conventional fossilfueled power plants. This should serve as an indication that the use of such cycles in nuclear plants should not be dismissed lightly without thorough evaluation. The current AEC-supported Monsanto effort to extend the thermodynamic data on organic fluids should provide an excellent basis for such evaluation. A broad study should be initiated to determine the potentialities and limits of direct-cycle organic reactor systems. The study should include both forced and natural-circulation systems with boiling within the core, as well as with flashing cycles. Experimental work should be included where necessary, particularly in the field of heat transfer and fluid flow, to provide a sound basis for the conclusions reached. Consideration should be given to the construction of a small Borax-type reactor experiment to test the most favorable concept found in the study.

B. Subdivided Fuel Concepts

Fluidized-bed reactors utilizing organic coolants have been proposed by both the Martin Company and Westinghouse. The unique feature of this reactor concept is that the fuel, instead of being in the form of rigidly supported rods, is in the form of free-moving pellets which are fluidized by upward flowing liquid coolant. The fuel proposed for the Liquid Fluidized Bed Reactor Experiment (Martin) consists of uranium dioxide pellets, 230 mils in diameter clad with 10 mils of aluminum, although metallic fuels could also be considered. The use of organic terphenyl coolant is incidental to the main feature of the concept, which is the fluidized fuel. However, because of the choice of coolant the design of the LFBRE is strongly influenced by the design of the OMRE.

The program which has been proposed to study the fluidized-bed reactor is not intended specifically to further the technology of organic coolants. Consequently, it has been proposed that only those aspects of the coolant which pertain directly to the requirements of the fluidized-bed reactor should be studied. For example, changes in physical properties (such as density and viscosity) of the coolant resulting from irradiation would affect fluidization of the bed, and therefore would require study. Certain aspects of organic-cooled reactors, such as fouling of heat transfer surfaces and rate of buildup of high boilers, might be affected by the different type of fuel, and would also be investigated

The contamination of coolant by (1) fission products released through defects in the cladding of the fuel pellets, and (2) debris resulting from fuel pellet abrasion, would be another effect specific to the fluidized-bed concept, and would require study. The extension of this program to include contamination resulting from the use of unclad fuel pellets has also been proposed.

Because of the present unknown effects of surface boiling on fluidized-bed stability, the use of local boiling has not been recommended. A bulk coolant temperature of 400°C and a maximum temperature of 455°C for the fuel surface have been proposed for a 100-Mw(e) reactor. The suggested fluidized-bed reactors appear to have a power density limitation lower than that of fixed-fuel, organic-moderated designs. This limitation would probably offset advantages which might be gained through improved neutron economy, low cost of fuel fabrication, and more uniform burnup. Moreover, the economic benefits of a fuel featuring partial fission product containment are questionable. Costs of fuel fabrication include a cladding item, while at the same time the reactor system design must provide for fission product release.

It appears that an approach which might hold significantly greater promise is a natural circulation boiling slurry concept similar to SLURREX,⁽²⁰⁾ but adapted to use of an organic coolant. Here the potential for high power density and low-cost fuel cycle are outstanding. Moreover, the slurry fuel concept has a particular advantage when coupled with organic coolants, in that the large heat transfer surface offered by the slurry minimizes the effect of the poor heat transfer characteristics of the organic. In addition, with organic fluids, consideration can be given to the use of metallic fuel powders, which may offer further advantages over the ceramic powders to which aqueous slurry concepts are limited.

C. Scope of Organic Reactor Program

There are indications that the relatively narrow scope of organic reactor investigation to date may have restricted development of concepts to lines previously employed for pressurized water reactors. Novel features particularly suited to the properties of organic coolants may thereby have been neglected. Both present and advanced organic reactor concepts should be reviewed more broadly, with the aim of determining areas where departures from present principles and utilization of specific coolant properties might result in significant economic improvements.

IX. APPARENT ECONOMIC POSITION OF ORGANIC REACTORS

The determination of the true economic position of organic reactors is not possible at the present time because of the limited experience with the concept and because of the many areas which require research and development. The results of the research and development efforts will have a pronounced influence on the economic analysis and obviously no absolute statements can be made of the current and future economic positions at this time. In lieu of absolute determinations, estimates have been made of current and future development efforts and the expected power cost for current and future organic reactor power plants.

As an aid in assessing the economic status of organic reactors, published power costs from various sources are listed in Table 2. This table presents the published estimates of organic reactor and coal-fired power plants in the range from 10 to 300 Mw(e). Current and future cost are shown for the organic reactor power plants, but only current coal-fired power costs are shown. It should be recognized that the power costs shown for the organic reactor depend upon the successful outcome of the research and development program.

| Item | Status of Lechnology | Power Mw(e) | Fixed(a) Charges | Fuel-cycle Cost | Operation and Maintenance | Nuclear Insurance | Total | Reference |
|--|--|--|---|--|--|---|---|--|
| 1 | Piqua | 11.5 | 13.94 | 4.5 to 5.5 | 2.0 | í. | 20.44 | 21 |
| 2 3 4 5 (1 7 8 | Current | 12.65 44 50 75 200 300 300 300 300 300 | 19.2 8.6 7.05 6.98 4.81 4.39 4.00 | 5.3 4.2 3.16 6.33 5.90 5.72 2.81 | 6.1 2.3 1.60 1.88 1.28 1.09 1.15 | 2.4(b) 1.1 0.73(b) 0.64 0.32 0.25 2 | 21.44 33 16.2 13.2 15.83 12 31 11 45 8.02 | 22 22 21 23 23 24 21 |
| $ \begin{array}{c} $ | Current Coal-fired Plant 35 c 10 ⁶ B | 12.65 -44 -60 -200 -325 | 10.4 6.1 4.41 3.57 3 31 | 4.6 3.8 3.88 3.34 3.32 | 2.8 1.2 0.75 0.50 0.36 | 0.5(b) 0.3(b) - - | $ \begin{array}{r} 18.3 \\ 11.4 \\ 9.04 \\ 7.41 \\ 7.0 \\ \end{array} $ | 22 22 23 23 23 |
| $ \begin{array}{c c} 14 \\ 15 \\ 16 \\ 17 \end{array} $ | Potential | 12.65 44 300 300 | 17 7.4 353 3.66 | 3.5 2.3 1.82 1.51 | 4.1 1.5 1.09 0.74 | 2.4(b) 1.1 0.22 : | 27 12 8 6.67 5.91 | 22 22 24 21 |

| | | | | | | , |
|-----------|---------|-------|------------|--------|-------|-----|
| ESTIMATED | NUCLEAR | POWER | GENERATION | COSTS, | MILLS | KWH |

Table 2

(a)14° fixed charges

(b)Includes working capital and initial replacement

From Table 2, the usual trend of lower power costs with size and time can be noted both for the current and future organic reactor and coal-fired plants. There is a noticeable difference between items 1 and 2 for current designs. A possible explanation is that the Piqua plant is AEC owned and does not include the same capital cost items as item 2. Items 3 to 5 inclusive show a reasonable agreement, but item 7 differs markedly from item 8. The difference in the current 300-Mw(e) power costs occurs primarily because of estimated fuel-cycle costs. The fuel-cycle cost of 2.81 mills/kwh for item 8 reflects a higher fuel burnup than in item 7. Aside from these discrepancies, the current and potential organic reactor total power costs agree within about 30% for the plants producing greater than 50 Mw(e).

The apparent economic position of organic reactors can be estimated by comparing the nuclear power costs with the power costs from a coalfired plant. The latter costs are shown in items 9 through 13. The spread between the nuclear and coal-fired power costs decreases with increasing plant size. In spite of this, the organic reactor is shown to have higher power cost for current designs. The incentive in pursuing organic nuclear reactor power plants is clearly tied to the cost advantages that will accrue with time as shown by items 14 to 17 if the research and development programs succeed in substantiating the conditions specified in the potential organic nuclear power plants and if coal-fired plant costs increase or remain constant.

In the studies listed in Table 2, the cost of the organic is taken to be about 17 cents/lb. For the decomposition rates with organic-moderatedand-cooled reactors, a makeup cost of 0.6 mill/kwh is indicated and is considered a part of the operation and maintenance cost shown in Table 2. If research and development of low-cost organics (about 3 cents/lb) is successful, lower power cost will result. For the same makeup rate, the use of the low-cost organic could result in a major cost reduction of 0.5 mill/kwh. For this reason, support of research and development of low-cost organic fluids should be encouraged.

Other cost reductions may be possible by minimizing containment, by increasing performance, and by simplifying the reactor system. It is expected that cost reductions in these areas will materialize after sufficient experience is gained in operating organic reactor power plants.

X. RECOMMENDED ADDITIONAL AREAS OF DEVELOPMENT

The current U.S. program aimed at the development of organic nuclear reactors is of considerable breadth and intensity. This, together with the Canadian project and with European work and plans, constitutes a diversified effort along two major lines: organic-moderated, enriched uranium reactors, and heavy water-moderated, natural uranium reactors. Nevertheless, it appears that most of the programs have fairly short-range and narrow goals in keeping with the specific nature of the concepts they are aimed to demonstrate, and that empirical rather than basic solutions to problems may be considered acceptable for this purpose.

It is, however, conceived that the field of useful organic reactors may well be considerably broader than is at present generally recognized. Furthermore, a truly adequate development of the presently emphasized concepts, in part because of the complexity of the coolant system, will require basic knowledge of the mechanisms, interactions, and limitations of the systems involved. It is important to proceed concurrently with present efforts toward such ultimate goals in order that organic concepts remain with other reactor concepts. Other types of reactors, as well as conventional power sources, can also expect continued benefits from technological advances within their areas. Consequently, the following areas of research and development pertinent to organic reactors are recommended for new investigations, or for amplification where present efforts seem undesirably limited:

A. Coolant Radiation Chemistry

1. Determination of the fundamental mechanisms of radiolysis of a broad spectrum of organic compounds the transient intermediates formed, the composition distribution, and properties of the compounds produced.

2. Determination of the temperature effect and catalytic effects on radiation decomposition and polymerization in the range of reactor operating conditions.

3. Development of analytical methods and tools for these studies.

B. Coolant Technology

1. Basic studies with the objectives of leading to new coolants having one or more of the following characteristics.

- a Readily reformable to useful coolants
- b. Capable of forming balanced mixtures of polymerizing and depolymerizing compounds.
- c Consisting of deuterated compounds.
- d. Optimum for reactors using D_2O and other moderators.
- e Optimum for direct-cycle power systems.

2. Studies to determine the relationship between fouling rate, size and composition of particles, and coolant composition.

3 Development of coolant management techniques for initial formulation and subsequent maintenance of coolant having specific properties. Investigation of optimum composition, as well as concentration, of high boiler fraction (i e., processing to leave quaterphenyl and hexaphenyl in "good" coolant, and removing only material above this). The scope of AI plans in this field is not known, and it is included because of its importance

C. Coolant Processing and Reclamation

1. Determination of the feasibility of a single system for performing the multiple functions involved in coolant processing. 2. Determination of the rate of particle formation in the coolant under various conditions and the requirements for removal of insoluble particles.

3. Examination of alternative methods of waste-coolant disposal, such as burial in terrestrial pits and refinements of known techniques; determination of the feasibility of separating potentially useful fractions (for other applications) from waste coolant.

4. Development of improved methods for disposal of waste gases containing gaseous fission products released during possible fuel element ruptures.

5. Determination of the economic feasibility of simplifying coolant-processing operations by modifications in the reactor system design to avoid complicating factors, such as formation of insoluble particles and in-leakage of water and air.

6. Determination of the feasibility of treating chemically the whole coolant (as distinguished from separated high boiler fraction) to accomplish one or more of the following items:

- a. Counteract the effects of radiolytic decomposition
- b. Provide improved resistance to radiation
- c. Lower viscosity
- d. Decrease melting point.
- 7. Coordination of coolant reclamation studies with:
 - a. coolant management studies to aid in selecting the optimum reclamation approach;
 - b. preliminary cost estimates to screen out expensive processes.

8. Examination of the potential for simplified coolant reclamation of thermally stable aromatic compounds other than the polyphenyls, and of thermally stable nonaromatic compounds.

D. Heat Transfer and Fluid Flow

1. Careful measurements of significant physical properties of various coolants over composition ranges conceivably influenced by reactor irradiation, processing, and reclamation; this could permit much-improved correlations of heat transfer data and optimization of system operating conditions.

2. Determination of the heat transfer and fluid flow characteristics of present and potential organic reactor coolants during local boiling and bulk boiling as a function of the many parameters involved in a multicomponent, irradiated organic fluid.

3. Determination of the allowable operating limits of fuel element spacing, and of fluid and surface temperatures.

4. Evaluation of burnout heat transfer limitations of seriously considered coolants and of the factors significant in burnout.

E. Fuels Development and Materials Evaluation

l. Development of effective and inexpensive methods of compaction of UO_2 powder in aluminum powder metallurgy tubes.

2. Determination of burnup limitations of uranium-molybdenum alloys.

- 3. Development of fuel elements based on other materials, such as:
 - a. Uranium alloys with high-temperature strength, high burnup capability, and low parasitic absorption. The latter is particularly important for nonorganic-moderated reactors.
 - b. Uranium compounds, UC and U₃Si
 - c. Uranium-containing glasses.

4. Organization and implementation of a cooperative program for the commercial development of aluminum powder metallurgy fuel element components; development of aluminum powder metallurgy products with greater strengths at elevated temperatures than those now being made.

5. Development work with the objective of producing zirconium alloys or products with resistance to hydrogen absorption from exposure to organic reactor coolants.

6. Evaluation of the seriousness of the potential problem of hydrogen embrittlement of steels in organic reactors.

7. Investigation of techniques for preparing organic reactor fuels suitable for reprocessing in conventional facilities.

8. Investigation of naturally formed organic films as potential cladding materials for fuels, particularly for slurry or fluid-bed fuels.

F. Reactor System Design and Component Development

1. Critical evaluation of specifications which have been written for components of organic reactor systems, and comparison with observed reactor operating conditions and results, to determine areas where specifications are unnecessarily stringent and where relaxation could reduce costs without affecting utility.

2. Development of improved gasket, packing, and sealing materials, and methods for containment of organic fluids at high temperatures.

3. Investigation of existing compact heat exchanger designs, and development of new designs, for organic reactor applications.

4. Development of simplified and rapid fuel-handling techniques.

5. Determination of the effect of varying content of radiolysis products on flash, flame and auto-ignition temperatures in presently considered coolants, as well as those developed in the future.

6. Investigation of the toxicity of terphenyls and other proposed coolants, and of their radiolysis products, with particular emphasis on the formation of carcinogenic compounds.

G. Advanced Concept Development and Evaluation

1. Broad investigation of the potentialities and limitations of possible direct-cycle organic reactor systems, including experimental work where necessary; construction of a small reactor experiment to test the most promising concept and to verify predicted potential and limits.

2. Consideration of the potentialities of a natural-circulation boiling organic slurry reactor (SLURREX principle), including evaluation of metallic fuel powders.

3. Broad review of scope of organic reactor program, including reconsideration of present and advanced concepts, to determine if departure from present principles and incorporation of novel features peculiar to the use of organic coolants might result in more economical systems.

In recognition of the significance of work now planned and under way in foreign countries, it is recommended that specific informationexchange (and possibly personnel exchange) agreements be made with Danish, German and EURATOM organic reactor projects. These should provide for frequent free interchange of internal reports and maintenance of close personal liaison with these project groups (and with CNEN, Italy, with which an agreement already exists) to ensure that all new developments are recognized as quickly as possible.

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Appendix

COMPILATION OF REPORTS ON VARIOUS PHASES OF ORGANIC REACTOR TECHNOLOGY

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COMPILATION OF REPORTS ON VARIOUS PHASES OF ORGANIC REACTOR TECHNOLOGY

I. PERIODIC REPORTS

A. Atomics International

Annual Technical Reports on AEC Unclassified Programs:

| Period | Report No. | Date |
|------------------|----------------------------|------------|
| Fiscal year 1960 | NAA-SR-5350 | Aug 1960 |
| Fiscal year 1959 | NAA-SR-3850 | Aug 1959 |
| Jan-Dec 1957 | NAA-SR-2400 (Pt I & II) | March 1958 |

Bimonthly Technical Progress Reports:

| Period | Report No. | Date |
|-------------------|-------------|------------|
| Nov-Dec, 1960 | NAA-SR-6004 | March 1961 |
| Sept-Oct 1, 1960 | 5836 | Jan 1961 |
| March-April, 1960 | 5242 | July 1960 |
| July-Aug, 1960 | 5666 | Oct 1960 |
| Jan-Feb, 1960 | 5042 | May 1960 |
| Nov-Dec, 1959 | 4820 | March 1960 |
| Sept-Oct, 1959 | 4586 | Jan 1960 |
| July-Aug, 1959 | 4313 | Oct 1959 |
| March-April, 1959 | 3815 | June 1959 |
| Jan-Feb, 1959 | 3579 | April 1959 |
| Nov-Dec, 1958 | 3450 | Feb 1959 |
| Sept-Oct, 1958 | 3350 | Dec 1958 |
| July-Aug, 1958 | 3050 | Oct 1958 |
| May-June, 1958 | 2950 | Aug 1958 |
| March-April, 1958 | 2750 | June 1958 |
| Jan-Feb, 1958 | 2650 | April 1958 |
| Nov-Dec, 1957 | 2468 | Feb 1958 |
| Sept-Oct, 1957 | 2300 | Dec 1957 |
| July-Aug, 1957 | 2131 | Oct 1957 |

General Chemistry Quarterly Progress Reports:

| Period | Report No. | Date |
|------------------|-------------|------------------|
| July-Sept, 1955 | NAA-SR-1544 | M ay 1956 |
| April-June, 1955 | 1476 | March 1956 |
| Oct-Dec, 1954 | 1433 | March 1956 |
| July-Sept, 1954 | 1205 | June 1955 |
| April-June, 1954 | 1087 | Dec 1954 |
| Jan-March, 1954 | 1026 | Nov 1955 |

Organic Moderated Reactor Experiment Progress Reports: C. A. Trilling, ed.

| Period | Report No. | Date |
|------------------|-------------|--------------------|
| April-June, 1957 | NAA-SR-2399 | Dec 1958 |
| Jan-March, 1957 | 2150 | Feb 1958 |
| Oct-July, 1956 | 1700 | M arch 1957 |

Organic Moderated Reactor Experiment Quarterly Progress Reports:

| Period | Report No. | Date |
|------------------|-------------|------------|
| April-June, 1957 | NAA-SR-2057 | March 1958 |
| Nov-Dec, 1956 | 1850 | May 1958 |
| Aug-Oct, 1956 | 1800 | Nov 1957 |

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| June-Sept, 1959 | AECU-4447 | Oct 1959 |
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