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GAS-COOLED REACTOR COOLANT CHOICE

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GAS-COOLED REACTOR COOLANT CHOICE

Gas-Cooled Reactor Project

Reactor Projects Division

June 12, 1958

Classification cancelled (changed to)
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SUMMARY

This memorandum is a compilation of the technical information presently available which bears on the problem of selecting a coolant for the graphite moderated gas-cooled reactor.

Discussions are included on the question of helium containment and availability, the status of the technology on carbon dioxide-graphite reactions, the influence of coolant choice on power cost and the compatibility of carbon dioxide with various fuel cladding materials.

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1. HELIUM CONTAINMENT PROBLEM

1.1 Containment Experience

The GCR-2 helium-cooled graphite-moderated reactor proposed in ORNL-2500 includes an operating system consisting of the following equipment:

1	50-ft diameter sphere
4	20-ft diameter, 40-ft high cylindrical steam generators
4	axial-flow helium blowers with motors contained in the system
4	60-in. diameter butterfly control valves
4	30-in. diameter butterfly control valves
800 ft	60-in. diameter pipe
120 ft	30-in. diameter pipe
24	60-in. diameter expansion joints
8	30-in. diameter expansion joints

Total system volume = $105,000 \text{ ft}^3$; 940,000 scf helium

The economic evaluation of the system is based on containing helium in the system at 300 psia and 450°F with 1000°F gas temperature in the vessel discharge piping. The system is to be completely welded. The maximum allowable leakage is 1% of the system volume/day which will cost 0.05 mill/kwh at a price of \$22.70/1000 scf.

The question of whether the system leakage can be limited to the specified losses has been open to much positive and negative speculation. The designers' reasons for optimism largely evolve from U. S. gaseous diffusion plant experience where a leak-tightness which is 1/10 of the GCR-2 design value has been successfully demonstrated. The essential differences between the gaseous diffusion plant type installations and the present system are:

1. The differential pressure is 19 times that of a vacuum system (based on the same mechanical integrity).
2. The operating temperature is substantially higher.
3. Gaseous diffusion plant compressors are sealed by a special dynamic shaft seal whose design is presently classified. The leak tests are carried out with the dynamic seal replaced by a static seal.

This survey has been carried out to determine whether experience in other helium installations will support the optimism of the GCR-2 designers.

The possible sources of leakage in the GCR-2 helium systems are:

1. Diffusion of helium through the metal wall.
2. Leakage through welded connections.
3. Leakage past valve stems through valve packing or bellows seals.
4. Leakage through electrical connections.
5. Losses at the fuel element sampling connections or other instrument leads.
6. Losses through compressor seals if used.

The diffusion of helium through metals has been a common fear among helium users. There is a marked difference in the performance of cast and wrought metal products with respect to helium leakage. It is an accepted fact throughout the industry that castings are generally unsatisfactory. Although it is not universally true that castings will not contain helium, there are cases (e.g., some cast valves) in which castings which would satisfactorily hold air have been found to leak helium. Techniques for producing helium-tight castings would require special investigation and perhaps development work. On the other hand, leakage of helium through wrought steel materials such as rolled plate, extruded or drawn piping and tubing, and forged shapes has been established to be nil when the material has been previously inspected and found free of defects. Commercially available material without special manufacturing requirements has proved acceptable in this respect. Mr. Ken Premo¹ of the Robertshaw-Fulton Company states that his company has thoroughly investigated the matter and has fully established this premise. The conclusion is further borne out by the use of large quantities of plate and pipe in the helium storage facilities of the Naval Air Station at Bainbridge, Maryland, and Lakehurst, New Jersey, and the Bureau of Mines at Amarillo, Texas. All of these

¹ 1957 Helium Symposium, Anaheim, California, "Helium Components and Systems Development Problems", Ken Premo, Robertshaw-Fulton Company, Anaheim, California.

installations report (see Appendix I) no difficulty with this problem when storing helium at pressures up to 2000 psig. The Edwards Rocket Base at Edwards, California (see Appendix I-H), reports similar experience in stainless steel containers at pressures up to 6000 psig. The Bureau of Mines also reports that it uses forged steel tanks for helium shipment by tank truck throughout the U. S. and experiences no measurable leakage at pressures up to 2000 psig.

Very little experience is available in high-temperature systems for helium storage. Reactor studies by the Sanderson and Porter Company are predicated on the use of helium at pressures from 50 to 100 atmospheres and temperatures up to 1500°F. Mr. S. C. Robinson of Sanderson and Porter states that he has found no evidence to indicate that the helium diffusion through the metal walls will increase significantly because of high temperatures if the metal is metallurgically sound (see Appendix I-B). Many high- and low-alloy steels have been leak-tested at high temperatures for various experimental purposes in connection with nuclear reactor work at Oak Ridge National Laboratory and no evidence of change in leakage characteristics has been reported. It would appear, then, that the high operating temperature of the gas-cooled reactor will not materially affect the helium leakage problem. This observation must be qualified by the fact that no test data have been collected which are intended primarily for the measurement of helium leakage under pressure at elevated temperatures. The possibility exists that there may be some phenomena heretofore not encountered in test work which might accelerate the helium-leakage rates. It is suggested that several autoclave tests with helium at elevated temperatures be carried out to confirm the effect of temperature.

A 24-ft diameter and 30-ft high vessel capable of containing 200 psig of gas pressure is also available at the Oak Ridge National Laboratory and could be assembled in a manner which permits a test of its helium containment capabilities at elevated temperatures (up to 500°F). The cost of such a test would be around \$15,000 (see Appendix II). It would show whether significant leakage occurs through welds and metal walls. At additional expense electrical penetrations, valves, gasketed joints, or other areas of suspect could also be tested. This

test is only recommended if information developed elsewhere than in this report shows the need for such a demonstration.

Leakage through weld joints was also discussed with personnel at the abovementioned sources. The response in all cases reveals that if the welds were made by fully qualified welders and thereafter inspected in accordance with ASME Unfired Pressure Vessel Code practice, no leakage difficulties were experienced. At the Edwards Rocket Base the helium is tested by mass spectrometry and constantly monitored. Thus, leakage rate below that detectable with their test instruments is maintained.

The loss of helium through valves was included in the discussions. All sources report difficulty with packed valves; however, the rate of leakage is not substantial. The Bureau of Mines at Amarillo, which has a storage capacity of 7.5 millions scf, reports (see Appendix I-A) that losses in that system are probably in the neighborhood of 0.1%/day from all sources. At Edwards Rocket Base, where bellows-sealed valves are used, no detectable leakage past the valve stems has been observed.

There is no experience with helium leakage past electrical connections. A comparable problem has been encountered in the PWR reactors which are required to contain radioactive vapors. The Shippingport Reactor designers report that the seal developed for this application has been adequate. The problem was also discussed² with Mr. Lynn Donnelan of the Allis Chalmers Nuclear Products Division who stated that a "mass spectrometer-tight" seal which would hold 2000 psi had been developed and that the adaptation of a seal of this type to contain helium would be a straightforward engineering problem.

The potential of instrument connections as sources of leakage can be considered to fall into two categories. The first includes sampling tubes and pressure taps which are essentially welded connections and fall within the scope of previous discussions concerning the tightness of welded connections. The second includes mainly thermocouple leads and may be treated in the same manner as electrical connections.

² Discussion between R. D. Stulting and M. Bender of ORNL and Lynn Donnelan of Allis Chalmers.

While not included in the design described by ORNL-2500, the utilization of a dynamic compressor shaft seal is potentially desirable since it would permit the motor to be exterior to the coolant system. The design of a satisfactory seal has been discussed with the Allis Chalmers Company.³ Allis Chalmers has successfully developed a water- or oil-buffered journal type seal which is used as a positive sealant for their hydrogen compressors which operate at pressures up to 750 psi with capacities up to 2500 cfm. These seals do permit leakage of some sealant into the hydrogen system. The leakage can be and is sometimes restricted by the use of labyrinth seal in series with the journal seal. The sealant-leakage can be completely eliminated by bleeding off some of the pumped gas through the labyrinth to prevent diffusion of sealant into the system. The bleed stream must then be removed from the system. Allis Chalmers has estimated that not more than 0.1% of the compressor throughput need be purged through the labyrinth to avoid contamination of the gas system.

Allis Chalmers is currently developing this design in connection with the Aerojet-General nitrogen-cooled reactor. Journal seals can be designed for use with either oil or water. The liquid, according to A-C engineers, poses no problem as an effective sealant so long as it has a low viscosity. The purge stream of 0.1% through the labyrinth while insignificant with respect to compressor performance could not be discarded in a helium system since it would amount to about 3 times the system volume of GCR-2 each day. The helium would have to be recovered, cleaned, and returned to the system. The flow rate of 125 cfm might be accomplished by the use of an auxiliary compressor or by feeding the bleed stream from the compressor discharge to the labyrinth and returning it through a clean-up system to the compressor suction.

The temperature of the gas in the bleed stream must, of course, be the same as that in the compressor. The Allis Chalmers engineers do not feel that the 450-500°F temperatures in the gas stream of the GCR-2 cause any major complications. The sealant fluid (whether oil

³ Conversations between M. Bender and W. T. Furgerson of ORNL, C. F. Codrington, D. D. Miller, H. T. Welch of Allis Chalmers Company, and representatives of Kaiser-ACF (see Appendix III).

or water) maintains the entire seal assembly as well as the compressor shaft in the seal area at the sealant temperature. Thus a heat sink is available to extract heat from the gas bleed as it passes through the labyrinth seal so that little danger exists that the bleed gas would vaporize the sealant.

An activated alumina or carbon absorber trap would probably be adequate to remove oil vapors from the helium. (Note: this system is frequently used for removing oil from dry air systems where the need for clean air is important. The Bureau of Mines uses activated carbon in the final purification step of its helium recovery system.) Allis Chalmers stated that they have been using their journal seal in hydrogen systems for two years without failure. Eight such compressors have been shipped from the A-C plant.

The Allis Chalmers Company also reported that work was being carried out on a rotary disc face type seal but the reliability of this unit was open to question. They could not at present recommend it for the proposed gas-cooled reactor installation.

The above discussion leads to the conclusion that an all-welded system which can meet the helium leakage requirements can be constructed without the development of new technology. The prospects appear hopeful for obtaining a compressor shaft seal which could meet the helium-cooled reactor requirements and while this is unnecessary to the successful execution of the design, it would be a desirable feature from the standpoint of improved maintenance access.

Acceptance of these conclusions, however, should not cause one to ignore the several design requirements which are implied. The GCR-2 design was based on fuel loading at zero power in an unpressurized system. Continuous fuel loading would be a difficult accomplishment for a helium-tight system. Undoubtedly, a substantial amount of development would be necessary to produce a seal which would permit a fuel-loading machine to be connected and disconnected remotely without significant loss of helium. The anticipated long fuel lifetime which is essential to the economic success of the system makes continuous fuel loading an unnecessary complication, but not an undesirable one. A CO₂ continuous-loading system would undoubtedly require a less complex sealing problem because of larger

leakage allowances and would then be easier to design.

In spite of the emphasis which has been placed on an all-welded system, the use of bolted and gasketed connections has not been shown to be impractical. Connections of this type are used in high-vacuum systems and in some helium systems.⁴ The reliability of this type connection must be established by some test experience.

The bellows-sealed butterfly valve is another problem which should not be lightly passed. The sealing problem is straightforward, but in order to obtain the desired flexibility, the bellows wall must be thin and a counterbalancing pressure may be required to limit pressure stresses. While the counterbalancing pressure could be applied with air, the valve design must be carefully tested to assure complete integrity of the bellows seal.

Finally, it should be noted that while the economic incentive to obtain a leakage specification of less than 1% of the system volume/day is not great for the design described by ORNL-2500, a smaller 40-Mw prototype reactor would have a higher helium cost factor/unit of energy output (about 0.25 mills/kwh) and would represent a significant part of the operating budget. Furthermore, the design of a system of greater tightness would reduce the drain on the domestic helium resources and remove a substantial part of the objection to the use of helium in the reactor export market. Since no evidence of the impracticability of obtaining a tighter system has been uncovered, it is suggested that a system designed to limit losses to 0.1% of the system volume/day be used as a goal for helium-cooled reactor installations rather than the 1% specified by ORNL-2500.

12. Helium Availability

The helium supply for use in gas-cooled reactors has been surveyed by L. G. Overholser of ORNL.⁵ Usage during the years 1957, 1958, 1959,

⁴ W. H. Lawrence of Edwards Rocket Base reported that ring-joint-flanges have been successfully used although they are not preferred in the high-pressure storage system at that installation. "Operational Helium" by W. H. Lawrence, Edwards Rocket Base, Edwards, California, Helium Symposium 1957, Anaheim, California.

⁵ ORNL-CF-58-4-112, Memo from L. G. Overholser to W. H. Jordan (see Appendix IV).

and 1960 are estimated by the Bureau of Mines as:

1957	281.5 scf x 10 ⁶
1958	387.1 scf x 10 ⁶
1959	432.7 scf x 10 ⁶
1960	464.6 scf x 10 ⁶

Of this amount, 12,000,000 scf or 2.6% is allocated to nuclear reactors for the year 1960.

The recent Bureau of Mines proposal⁵ for the conservation of helium proposes the construction of 13 recovery plants capable of increasing helium recovery to 2.4 billions scf/year by 1975. If this proposal is accepted, a 32-billion scf reserve of helium would be developed by 1975. The proposal would cause an increase in helium price from \$19.00/1000 scf to \$44.50/1000 scf. The GCR-2 helium costs would increase from 0.05 mills/kwh to 0.12 mills/kwh if this price increase becomes effective and would thus have very little influence on the overall power cost of a GCR-2 type reactor.

The proposed expansion of our helium recovery system by the Bureau of Mines should make available sufficient helium to support a substantial portion of the United States electrical power needs through the year 2005. This conclusion can be drawn from the fact that the Bureau of Mines proposal plans a helium recovery capacity of 2.4 billions scf/year by 1975 while the domestic usage in that year would be 1.2 billions scf. The difference between the capacity and the usage would indicate that 1.2 billions scf of excess capacity would be available for nuclear power activities. Some of the production resources included in the Bureau of Mines conservation plan may be nearly exhausted by the year 1975 while others with lower helium concentrations not presently included in the plan presumably could be developed if required.

A specific accounting of helium usage might clarify this position. The thirteen fields included in the recovery plan have 119 billions scf of recoverable helium of which 72.5 billions is to be recovered under the current plan. In 1975 the Bureau of Mines expects that 11.5 billions scf will have been exhausted. In the same year helium usage is estimated at 1.5 billions scf. The Bureau of Mines usage rate includes nuclear power application requirements so that it is probably fair to estimate that non-nuclear helium usage in 1975 will be somewhat lower, and that the yearly

average for the period 1975-2005 may approach this amount. The total non-nuclear consumption would thus be 45 billions scf from 1975 to 2005. Subtracting this quantity, plus the 11.5 billions scf utilized up to 1975, from the total of 72.5 billions scf to be recovered leaves 16 billions scf of helium available for nuclear power reactors. If the conservative helium usage values of ORNL-2500 are the basis, a 225-Mw reactor will use 3,440,000 scf/year. In 20 years, the anticipated power plant lifetime, it will use 69 millions scf of helium. The 16 billions scf of helium available for nuclear reactors would thus permit operation of 233 nuclear power plants of 225-Mw capacity, or 52,300 Mw. This represents 56.5% of the U. S. 1955 steam-electrical capacity of 92,000 Mw. By expanding the Bureau of Mines helium conservation plan to include recovery of the remaining 46.5 billions scf of helium which is potentially available but not now included, the amount of nuclear electrical power which could be supported amounts to 204,000 Mw.

Expansion of the Bureau of Mines conservation plan to the extent outlined above may not be practical. Probably, it would not be necessary. Experience with U. S. gaseous diffusion plants has shown that a leakage of 0.1% of the system volume per day is feasible. The fact is further supported by the experience of the Bureau of Mines at Amarillo (see Appendix IA). The possibility that higher capacity plants than the ORNL GCR-2 can be built without increasing helium consumption rates also favors a reduction in helium consumption. If the 0.1%/day loss rate can be effected, the 16 billions scf of helium available (from 1975 to 2005) under the current Bureau of Mines conservation proposal would support 523,000 Mw of GCR-2 type plants. It is of interest to note that if the 10-year doubling time for electrical capacity in the U. S. is used as a criterion, the steam-electrical capacity in the U. S. would be:

1955	92,000 Mw
1965	184,000 Mw
1975	368,000 Mw
1985	736,000 Mw

The potential helium supply of 16 billions scf for nuclear power under the current Bureau of Mines helium conservation proposal might support virtually all of the steam-electrical capacity constructed between 1965 and 1985, the period when domestic nuclear electrical power capacity is

expected to gain real impetus, by a realization of the 0.1%/day helium loss rate.

The fact that the helium supply is limited should not lead to the conclusion that this type of gas-cooled reactor is dead-ended. Experience with gas containment and advancement in high-temperature materials technology will ultimately permit helium to be displaced as a coolant by some higher-performance fluid such as hydrogen in gas-cooled reactor systems.

The short-range picture is, perhaps, less attractive. If the recent announcement of the Joint Committee for Atomic Energy is taken as a criterion, the U. S. plans to be able to produce power competitively in foreign markets in five years and in domestic markets in ten years. This would mean competitive foreign reactors available in the year 1962 and competitive domestic reactors available in 1967. The principal blocks of foreign nuclear power are probably in the United Kingdom and the Euratom group. These are reported,⁶ respectively, as 6000 Mw in 1965 and 15,000 Mw in 1967. The United Kingdom program is predicated on the use of CO₂-cooled reactors and is probably not worth consideration as a helium user in short-range planning. The Euratom group, therefore, represents the major potential user. If the ORNL-2500 GCR-2 plant with 1%/day losses is again used as a reference, it can be seen that 67 such plants would be required to meet the Euratom demands through 1967. These plants would use 230 millions scf/year of helium or 1.64 times the quantity estimated for non-federal users⁵ in 1960. (If the inference from the Bureau of Mines data is correct, the non-federal usage is increasing at the rate of 20 millions scf/year so that in 1967 it would be 280 millions scf, a more favorable comparative value than the 1960 figure.) The need for immediate action to implement the Bureau of Mines conservation plan is indicated if domestic helium is to be utilized in the foreign reactor market. These expanded production facilities could meet the needs of a foreign reactor market up to 1967 and could continue for an extended period if the leakage rates could be cut to 0.1% of the system volume per day. (The 15,000 Mw required by the Euratom group would require only 23 millions scf/year in the latter case.)

⁶ "A Target for Euratom", Armand, Etzel and Giordani, 5-7-57.

1.3 Availability of Helium in Foreign Countries

As noted by Mr. Overholser in his April 29, 1958, memorandum,⁵ little is known about the availability of helium outside the North American continent. The helium deposits which were reported in the literature survey are certainly of such low concentrations as to be prohibitively expensive to separate using current technology, but it is also apparent that the meager information is not sufficient to justify a conclusion that there are no substantial helium deposits outside the U. S. Mr. Paul V. Mullins of the Bureau of Mines was consulted concerning this matter and he, too, concurred that there was insufficient evidence on which to base a conclusion. The Bureau of Mines has tested only a few samples from foreign sources, according to Mr. Mullins (see Appendix IA), and these have not shown the presence of significant quantities of helium.

A more extensive search for foreign helium resources would be desirable if helium is to be considered a nuclear reactor coolant in export reactors.

It is not impractical to consider the export of helium to foreign users. As noted previously, the anticipated requirements through 1957 would not severely tax the U. S. supply if the Bureau of Mines conservation plan is implemented. Helium is now exported in small quantities under special export licenses by the Interior Department and the State Department. (Mr. Mullins guessed that the quantity might be several hundred thousands scf/year.) In addition, the Defense Department must be shipping some helium in connection with overseas missile bases.

Dr. F. C. Balcor of Air Reduction Company was contacted (see Appendix ID) concerning overseas shipments of helium gas. Dr. Balcor, after consulting with his associates in the gas distribution department of Air Reduction Company, stated that either gaseous or liquid helium could be shipped overseas. He suggested that the tank cars presently used for transportation could hold about 250,000 scf of helium in gaseous form. These cars cost about \$100,000 each. Two such units could probably maintain a supply of helium for a GCR-2 type plant. Dr. Balcor could not estimate what the shipping cost might be.

Mr. R. E. Brown of the Bureau of Aeronautics in Washington was consulted (see Appendix IE) about the transportation of liquid helium. The Bureau of Aeronautics is planning a 100,000- to 300,000-scf/d liquifier for installation at Amarillo, Texas, in 1960. The installation is intended to permit shipments of liquid helium by air to overseas installations. Mr. Brown does not feel that this is cheaper than water transport, but it is much faster. Mr. Brown stated that liquid helium could be shipped by rail from Amarillo to Lakehurst, New Jersey, without difficulty. The lapsed time is about 2 weeks. The time cannot be substantially longer than 2 weeks because of a gradual increase in pressure caused by helium vaporization. The time period could be extended by designing shipping containers for higher pressures or for additional storage capacity.

The economics of water shipment would suggest that overseas transport should not be substantially greater than rail on a mileage basis. Since only about \$0.006/scf is applied to transportation costs for U. S. shipments from Amarillo to Oak Ridge, Tennessee, it is anticipated that ocean shipment to northern Europe would not be prohibitive. (A check with one shipper in the New York area indicates that a 35,000-scf helium trailer could be shipped from New York to Calais, France, for \$0.03/scf. If this were added to a probably maximum cost of \$0.015/scf for shipment to New York, the total cost would be \$0.045/scf. Using the projected price for helium of \$0.044/scf based on the Bureau of Mines conservation plan price, the total cost would be \$0.089/ft³. If ORNL-GCR-2 consumption rates are used as a basis, the helium contribution to power cost would be approximately 0.2 mills/kwh at Calais, France. Rates do vary to other countries because of shipping regulations and negotiation would be necessary to obtain competitive rates to other European countries.)

2. STATUS OF TECHNOLOGY OF CARBON DIOXIDE-GRAPHITE REACTIONS

2.1 Reaction of Carbon Dioxide With Graphite in the Absence of Radiation

Although considerable data are available in the literature for the CO_2 -graphite system in the absence of radiation part of it is of no interest since the graphites used have different characteristics and in general are much more reactive than the reactor grades of graphite. Studies on reactor grade graphites have been to a large degree performed at Hanford or by the British. The data presented in the following sections have been drawn mainly from these sources. It will be observed that no information on the behavior of the CO_2 -graphite system in closed flow systems is available and that studies of such factors as the partial pressures of CO_2 and CO and flow rates are very incomplete.

Reaction Rate Data

Rate data for the thermally induced reaction as reported by Davidge⁷, Burton⁸ and Walker⁹ are given in Table 2.1 and Fig. 2.1. Davidge employed a 1/4 in. x 2 in. cylinder of AGXP graphite in the temperature range 685-785°C at a flow rate of 1-2 liters/min. Burton used a 0.426 in. x 1 in. cylinder of CSF graphite having a surface area of 0.30 to 0.40 m²/g, a temperature range of 850-1000°C and a flow rate of 100 cm³/min. A 1/2 in. x 2 in. cylinder of Speers Carbon Co. graphite was used by Walker at 900-1200°C with a flow rate of 2 liters/min. Measurements were made at essentially one atmosphere of CO_2 pressure and the gas was not recycled. The agreement between Davidge's and Burton's results is satisfactory in view of the fact that different graphites were used and it appears reasonable to assume

⁷ P. C. Davidge, "The Reaction of CO_2 With Graphite", Dec. 2, 1949, AERE-C/R-465.

⁸ H. H. Burton, "Technical Activities Report-Graphite Studies Group- Dec. 1951, HW-23169; April 1952, HW-24489; May 1952, HW-24676.

⁹ P. L. Walker, Jr., R. J. Forrester, Jr., and C. C. Wright, Ind. Eng. Chem. 45, 1703 (1953).

Table 2.1 Reaction Rates for the Thermally Induced Carbon Dioxide-Graphite Reaction

Temperature		Percent Weight Loss per 1000 Days		
°C	°F	Davidge	Burton	Walker
300	572	1.3×10^{-8}	1.4×10^{-9}	8.2×10^{-6}
400	752	3.0×10^{-5}	4.4×10^{-6}	4.3×10^{-3}
500	932	8.5×10^{-3}	1.5×10^{-3}	4.6×10^{-1}
550	1022	8.8×10^{-2}	1.8×10^{-2}	3.1
600	1112	7.0×10^{-1}	1.5×10^{-1}	16
650	1202	4.4	1.1	72
700	1292	22	6.0	2.7×10^2
750	1382	1.0×10^2	30	9.8×10^2
800	1472	3.9×10^2	1.2×10^2	2.9×10^3
850	1562	1.3×10^3	4.1×10^2	7.7×10^3
900	1652	4.6×10^3	1.4×10^3	2.1×10^4
Activation Energy, kcal/mole		58.7	61.5	48

that rates of this order would be applicable to the reactor grade graphites used in this country. The higher rates reported by Walker very likely resulted from the greater surface area of the graphite used.

Factors Affecting the Reaction Rate

a. Surface Area. The reaction rate for the graphite-CO₂ system is markedly affected by the available graphite surface. A very effective method of increasing the surface area is through oxidation as shown by the studies of Spalaris¹⁰ and of Walker⁹. The surface area and pore size distribution were determined as a function of the degree of oxidation by CO₂ using nitrogen adsorption-desorption isotherms. The surface area was found to increase with increasing degree of oxidation with the most rapid increase occurring during the initial slight oxidation at which time the reaction rate also increases rapidly. Two of the principal distribution peaks in the micropore structure of graphite occur at 20-35 and 90-150 Å radii. The peak intensity of the former increases rapidly between zero and about two percent oxidation, decreases between two and six percent and remains essentially constant from six to thirty percent. The peak at 90-150 Å increases in intensity up to about six percent oxidation and changes very little thereafter. It is postulated that the 20-35 Å pores are unblocked during the early stages of oxidation resulting in the rapid increase in surface area and reaction rate. Subsequently, as they are enlarged the rate of increase diminishes and above six percent oxidation a generalized attack of graphite crystal probably occurs characterized by a constant reaction rate, the result of a dynamic balance between internal diffusion and chemical reaction. This is the reaction rate which is commonly reported.

Another method of increasing the surface area is to merely cut up the graphite. Spalaris¹¹ studied the reaction rates of CSF graphite using a

¹⁰C. N. Spalaris, "Surface Studies of Irradiated Graphite", July 14, 1953, HW-29082.

¹¹C. N. Spalaris, "Role of Surface Area in the Kinetics of Oxidation of Graphite", May 24, 1954, HW-31928.

1 in. x 1 in. cylinder with a surface area of $0.32 \text{ m}^2/\text{g}$ and discs cut from another cylinder having a surface area of $0.56 \text{ m}^2/\text{g}$. Oxidation in CO_2 at 1000°C showed that by increasing the initial area by a factor of about two the reaction rate was increased by a factor of about three. Thus, it is evident that part of the inaccessible pore space can be opened by reducing the size of the graphite specimens as well as by oxidizing the binding material and/or the crystallinities blocking the entrance to these pores. This suggests that data obtained for small specimens cannot safely be extrapolated to graphite bars.

b. Prior Irradiation. Hanford work¹² has shown that the reaction rates with CO_2 for a sample of CSF graphite subjected to a pile exposure of 3.1×10^{20} nvt are about four times as large as the rates found for virgin CSF graphite at $950\text{-}1000^\circ\text{C}$. Activation energies of 61.5 and 51.5 kcal/mole were found for the virgin and irradiated graphite samples, respectively. Similar effects on the reaction rate for the O_2 -graphite system were observed. Spalaris¹⁰ noted that the irradiated samples had larger surface areas than the virgin samples at comparable percentages of weight losses indicating an increase in surface area was produced by irradiation.

c. Type of Graphite. The reactivity of various graphites with CO_2 is mainly determined by the available surface area. Various types of reactor grade graphites produced in this country probably do not show any large differences in reactivity. Small variations in density may be unimportant. For example, Hanford has used KC graphite with a density of $1.70\text{-}1.80 \text{ g/cm}^3$ and CSF with a density range of $1.60\text{-}1.65 \text{ g/cm}^3$ and found no important differences¹². Although it is possible to produce special graphites which while having the same density may vary widely in surface area the use of such graphites in reactors is not likely. The type of binder could have some effect since the reaction with CO_2 involves removal of blockages from the pores and the blockages may be binding material and/or poorly crystallized material. Any departure from the use of standard binders also seems improbable. Likewise the use of low density graphites which would very likely be more reactive does not appear probable.

¹²R. E. Woodley, AEC Technical Cooperation Program, U.K.-U.S.A., "Discussions on Graphite Oxidation - Hanford Contribution", April 16, 1956, Wash-690.

d. Pressure and Flow Rate of Gas. For a heterogeneous reaction of the type CO_2 -graphite the reactive sites on the graphite surface are of prime importance. It would appear that at moderate pressures and flow rates of CO_2 that the availability of the sites would be the rate controlling factor. It is generally agreed that a rapid reaction of CO_2 occurs at the reactive sites giving rise to CO which passes into the gas phase and a surface oxide or oxygenated surface.^{13,14,15} The oxygenated surface undergoes a slow decomposition giving rise to CO which may either leave the surface or remain adsorbed on the surface. An adsorption-desorption equilibrium is established for CO in which part of the reaction sites on the carbon surface is occupied by CO and hence not available for the initial CO_2 reaction. An increase in the partial pressure of CO would be expected to retard the overall reaction rate by blocking more reaction sites.

There is very little information available on the effect of CO_2 and CO pressures and flow rates. Gadsby et al.¹³, studied the reaction of CO_2 with charcoal and gave an expression for the overall reaction rate for CO_2 pressures from 10-760 mm of Hg taking into account the three stages:

1. initial CO_2 reaction with C
2. evaporation of adsorbed O with atom of carbon from the surface
3. adsorption-desorption of CO on carbon surface.

It is doubtful, however, that the expression has any valid application to graphites.

Studies¹⁶ at Hanford have shown that the reaction for the CO_2 -graphite system becomes constant at some definite flow rate and an increase in the flow rate above this value does not increase the reaction rate as measured by the concentration of CO in the effluent gases. A decrease in the

¹³ J. Gadsby, F. J. Long, P. Sleightholm and K. W. Sykes, Proc. Roy. Soc. A193, 357 (1948).

¹⁴ W. E. J. Broom and M. W. Travers, Ibid 135A, 512 (1932).

¹⁵ F. Bonner and J. Turkevich, Journal American Chemical Society 73, 561, (1951).

¹⁶ R. E. Woodley, "Graphite Burnout Interim Report on PT-105-532-E", December 15, 1954, HW-34154.

surface to volume ratio of the graphite (radius of a cylindrical sample of given length is increased) requires a higher flow rate to reach the constant rate. Other studies¹⁷ have shown that if the partial pressure of CO_2 is reduced by the addition of helium the reaction rate is decreased showing that the reaction rate is dependent on the partial pressure of CO_2 . The addition of small amounts (3.4 percent) of CO to the CO_2 has been shown¹⁸ to reduce the reaction rates in the temperature range 850-1025°C. The data show, however, that with increasing temperature the logarithm of the rate for the mixture approaches that for pure CO_2 asymptotically indicating that the inhibiting effect of the CO is less at the higher temperatures due to increased desorption of CO from the surface at the higher temperatures. None of this or any of the other work was done in a closed flow system and consequently no information on the steady-state conditions is available.

Thermodynamic Equilibrium Data

The equilibrium concentrations of CO for the reaction $\text{CO}_{2(g)} + \text{C}_{(c)} \rightleftharpoons 2\text{CO}_{(g)}$ at various temperatures and pressures are given in Table 2.2. These values are of academic interest only since it is realized that for the systems of interest the steady-state conditions differ significantly from the thermodynamic equilibrium state.

¹⁷ H. H. Burton, Technical Activities Report - December 1952 - "Graphite Development-Pile Graphite", January 12, 1953, HW-26618.

¹⁸ H. H. Burton, Technical Activities Report-Graphite Studies Group, May 1952, June 5, 1952, HW-24676.

Table 2.2 Thermodynamic Equilibrium Concentrations of Carbon Monoxide at Various Temperatures and Pressures for Reaction:

Temperature		Mole percent CO					
°C	°F	1 atm	5 atm	10 atm	15 atm	20 atm	25 atm
350	662	0.28	0.13	0.07	0.06	0.05	0.04
400	752	0.94	0.42	0.30	0.24	0.20	0.18
450	842	2.8	1.2	0.88	0.72	0.62	0.56
500	932	7.0	3.2	2.2	1.8	1.6	1.4
550	1022	15	6.6	4.6	3.8	3.3	3.0
600	1112	32	14	10	8.2	7.2	6.4

2.2 Radiation Induced Carbon Dioxide-Graphite Reaction

The intense radiation present in a reactor may lead to chemical reactions which otherwise do not occur except at much higher temperatures. The reaction of graphite with CO_2 is radiation induced through the ionized and excited gas molecules, active species or short-lived intermediates. Some of the information available related to the radiation behavior of this system is presented in the following sections, but it will be apparent that the overall picture is quite complex.

Decomposition of CO_2

The decomposition of CO_2 by pile irradiation in the absence of graphite occurs to a limited extent. Woodley¹⁹ reports that approximately one percent of the CO_2 is decomposed while studies by Harteck and Dondes²⁰ in the Brookhaven reactor gave about 0.1 percent decomposition. Davidge and Marsh²¹ reported a maximum final CO concentration of 8×10^{-3} percent at 325°C using a CO_2 pressure of 75 cm of Hg and an exposure of 5.25×10^{17} n/cm². The gas phase processes have been represented as follows:²²

- (1) $\text{CO}_2 = \text{CO} + \text{O}$
- (2) $\text{O} + \text{O} (\text{wall}) = \text{O}_2$
- (3) $\text{O} + \text{O}_2 = \text{O}_3$
- (4) $\text{O}_3 + \text{CO} = \text{CO}_2 + \text{O}_2$

In the presence of a graphite surface the atomic oxygen or ozone will react with the formation of adsorbed CO or a surface oxide layer. Davidge and Marsh²¹ give the following scheme for the reaction with graphite utilizing the oxygen resulting from the CO_2 decomposition.

¹⁹R. E. Woodley, "The Promotion of Chemical Reaction by Pile Radiation", November 22, 1954, HW-40142.

²⁰P. Harteck and S. Dondes, J. Chem. Phys. 23, p. 902 (1955).

²¹P. C. Davidge and W. R. Marsh, "The Effect of Pile Radiation on the Carbon Dioxide-Graphite Reaction", 1955, AERE-C/R-1374.

²²J. O. Hirschfelder and H. S. Taylor, J. Chem. Phys. 6, p. 783 (1938)

or

A combination of (5), (6) and (7) or of (5), (6) and (8) yield $\text{CO}_2 + \text{C} = 2\text{CO}$ the first combination giving one molecule of CO per molecule of CO_2 dissociated, the second twice as much CO. The British admit this is speculative and conclude only that the forward reaction

proceeds through an intermediary having a mean life comparable to or shorter than one minute, the time of circulation in their experimental system.

The thermal reaction is inhibited by adsorbed CO on the graphite surface and a similar inhibiting effect would be expected for radiation induced reaction unless the adsorbed layer is removed by some means. Woodley²³ found that the gamma-induced reaction for graphite exposed to CO_2 in the MTR canal gamma facility was suppressed by adsorbed CO whereas exposure in the Hanford pile¹⁹ gave results which indicate that the CO was not an effective inhibitor. The graphite temperature, in either case, was not sufficiently high to desorb the CO and the difference in behavior is thought to be due to the desorption of CO in-pile by the energetic neutrons. While this implies a continuing reaction with the graphite and resulting build up of CO, the CO according to reaction (4) as well as the suboxide polymers will become competitors for the ozone molecules giving rise to an equilibrium state in which the rate of reduction of CO_2 will be equal to its rate of formation.

Decomposition of CO

The decomposition of CO under irradiation was observed by Lind and Bardwell²⁴ and more recently investigated by Woodley^{19,23}. The latter exposed samples of CO in both the MTR canal gamma facility and a Hanford pile and

²³R. E. Woodley, "The Promotion of Chemical Reaction in Gas-Graphite Systems by Gamma Radiation", May 24, 1954, HW-31929.

²⁴S. C. Lind and D. C. Bardwell, J. Am. Chem. Soc. 47, p. 2675 (1925).

observed the formation of carbon suboxide polymers in both facilities. Systems containing graphite and CO_2 gave the polymers in-pile but not in the gamma facility. Woodley believes that the following mechanism may be used to explain the decomposition of CO in-pile:

One molecule of CO is primarily dissociated per ion pair as in (1), but four additional molecules of CO are subsequently consumed in the processes represented by (2) and (3), plus either (4), (5b) and (4) or (5a), (3) and (4). This leads to the overall reaction

which is believed to occur at least during the initial period of irradiation. For the studies²³ in the gamma facility the ratio of moles of CO decomposed to moles of CO_2 formed remained constant at 5. In the case of pile radiation the ratio is not constant and in the fringe areas of the pile is approximately 4, corresponding to

Woodley believes that the lower value for the ratio in-pile is due to a degradation of the suboxide polymers by the energetic neutrons as follows:

The CO formed can subsequently decompose as before, but the reverse of either of the two processes given does not readily occur. As a result, carbon suboxide polymers with increased percentages of carbon are formed with increasing in-pile exposure. It is believed that the formation of C_4O_3 predominates initially, reaches a maximum and decreases as its rate of degradation exceeds its rate of formation. A similar behavior was postulated for C_3O_2 , but with the maximum occurring at a higher total dosage. The formation of elemental carbon would continually increase and might result in a solid composed largely of carbon after a long exposure. The color of the solid changes from reddish-brown for short exposures to dark brown for longer exposures. It is believed that the polymerization occurs in part in the gas phase, but there also is some indication that the surface may be instrumental in initiating polymerization.

Reaction Rate Data

The Hanford people have reported a large quantity of data bearing on the graphite burnout gathered from experimental studies conducted in the production piles. It is generally recognized that in many cases the test specimens have been subjected to numerous pile shutdowns and startups and that the pile atmospheres due to high leak rates are not always as carefully controlled as would be desired. Some of Hanford information related to reaction rates in-pile is given in the following sections. Finally, some British data on this subject are presented.

Hanford Studies

a. Effect of Temperature. Studies in the MTR canal gamma facility²³ using a static CO_2 -graphite system at $\sim 30^\circ C$ gave an average burnout rate of 0.06 percent/1000 days. Similar experiments using CO_2 diluted with 15 percent He gave an average value of 0.03 percent/1000 days. These experiments were run in a temperature range where the thermal reaction rate is not measurable and hence the burnout must be due entirely to the radiation-induced reaction. Experiments run in a water-cooled facility of a pile gave an average burnout rate of 0.15 to 0.25 percent/1000 days²⁵

²⁵J. C. Fox, R. E. Woodley and J. M. Wright, Production Test 105-504E, "Study of Gas-Graphite Reactions", February 29, 1952, HW-24676.

at a comparable temperature, demonstrating the effect of the neutron flux. A burnout rate of 0.044 percent/1000 days²⁶ was calculated from the CO content of a pile atmosphere operating at temperatures in the range 284-353°C. This represents the burnout from the entire stack of graphite and it is thought that the rate would be lower were it not for a leak rate of about 13 percent/day for this particular pile.

The results of numerous additional studies made in pile atmospheres below 450°C could be cited, but it may be stated that in general the values for the burnout rates fall in the range 0.1-0.8 percent/1000 days. The point of interest is the determination of the temperature at which the thermal reaction overshadows the radiation-induced reaction. Studies on the rate of the radiation-induced reaction as a function of temperature gave results that indicate that the activation energy for the radiation-induced reaction is from one to three kcal/mole²⁷. It is believed that the radiation-induced reaction is essentially independent of temperature below the temperature where the thermal reaction does not overshadow the radiation-induced reaction.¹⁶ In more recent studies²⁸ graphite cylinders 0.426 in. x 3-1/2 in. of 4 in. and tubes 3/4 in. OD, 1/4 in. ID x 3-1/2 in. were exposed in process channels, one channel at 550-600°C and the other at 650-750°C. A follower furnace, controlled by the 650-750°C channel, located outside of the pile but with pile gas flowing through it, was used for out-of-pile exposure of similar specimens. A rigorous analysis of the data is not possible due to fluctuations in the temperature and composition of the pile gas (50-85 percent CO₂, small but varying amounts of O₂ and H₂O). Weight losses of graphite at 650-750°C ranged from 10 to 20 percent/1000 operating days with no significant difference found for specimens exposed in-pile or out-of-pile. Exposure in-pile at 550-600°C resulted in losses of one to two percent/1000 operating days. From a comparison of these data with that for the thermal reaction it was concluded that at about 600-650°C the rate of graphite oxidation in the pile becomes strongly temperature

²⁶G. J. Alkire, R. D. Tillson, H. J. Anderson and J. L. Carroll, "Graphite Burnout Resulting from Carbon Monoxide Formation in C-Pile", February 1, 1954, HW-30627.

²⁷R. E. Woodley, "Final Report on Production Tests PT-105-391-P, PT-105-435-P and PT-105-435-P Supplement A", July 1, 1952, HW-26320.

²⁸J. C. Fox, "C-Pile Graphite Burnout Experiment Final Report, PT-105-548-E", August 31, 1956, HW-45942.

dependent due to the thermal reaction and that the radiation-induced reactions which are of extreme importance at lower temperatures are probably of negligible importance above 650°C.

In this same study, graphite bars 4 in. x 4 in. x 12 in. were exposed to pile radiation at temperatures ranging from 500-540°C. Losses ranging from 0.23-0.29 percent/1000 operating days were found which are 1/8 to 1/3 as high as the rate measured for 0.425 in. x 4 in. specimens. The latter oxidized about three times as rapidly as the 1.125 in. x 4 in. specimens. It was concluded that no quantitative relationship exists between the oxidation rate and sample size.

b. Effect of Pile Atmosphere. Lower burnout rates of graphite result when the CO₂ is diluted with helium. For example, graphite samples exposed to a pile atmosphere containing about 30 percent helium at a temperature of about 450°C gave an average burnout rate of 0.8 percent/1000 days compared to 1.6 percent/1000 days for a similar exposure in a pile gas containing mainly CO₂.¹⁶

In a study²⁹ of the effect of CO additions graphite samples were exposed to pile conditions in an uncooled process tube and the weight changes of the specimens observed along the length of this tube. With an atmosphere consisting of 95 percent CO₂ and five percent CO a maximum burnout rate of about three percent/1000 days was observed for virgin graphite compared to 4.5 percent/1000 days for previously irradiated graphite. These values were observed at 95 in. upstream from the moderator centerline at a temperature of about 245°C. At the centerline (415°C) the rates were down to around one percent/1000 days and remained nearly constant to 100 in. downstream (240°C) where they decreased rather sharply to less than 0.2 percent/1000 days at 120 in. downstream (80°C). The rates increased rapidly from 160 in. upstream (less than 100°C) to the maximum at 95 in. upstream from the centerline. While the flux as well as the temperature attain maximum values at the centerline, the maximum burnout occurred well upstream from this point. It is believed that two types of molecular species are formed. One from CO₂ which oxidizes the graphite (ozone and/or atomic oxygen), the other, probably carbon suboxides and/or carbon, from

²⁹R. E. Woodley, "Alternate Pile Atmosphere Study, Final Report PT-105-548-E", August 31, 1956, HW-45942.

the decomposition of CO which either removes the active species of oxygen or is eventually deposited as carbon. Ozone and atomic oxygen are probably produced more rapidly than are carbon suboxide and carbon since weight losses occur in the cooler upstream part of the facility. The burnout rate increases with increasing temperature and flux until the concentration of carbon suboxide or atomic carbon builds up to the point where a rapid gas phase reaction occurs or the deposition rate starts compensating for the rate of gasification. The burnout rate starts decreasing but does not become negligible until the cooler downstream fringe is reached. In the extreme downstream region solid deposition increases to the point where positive weight changes may occur. In a similar study using 75 percent CO₂ and 25 percent CO, a small gain was noted at the extreme upstream facility and also beyond about 100 in. downstream from the centerline. A burnout rate of 0.2-0.4 percent/1000 days was observed over the rest of the length. It was concluded that if the CO concentration were increased to the point where no burnout might occur, that considerable deposition of carbon would take place in the fringe areas.

British Data

The British have made studies of the CO₂-graphite reaction in closed flow systems in connection with their gas-cooled reactor program.

Davidge and Marsh²¹ have reported the results obtained using a glass loop at temperatures up to 350°C in the BEPO. Experiments carried out with AGXP graphite using flowing CO₂ with the pile at zero power gave a reaction rate of less than 2.5×10^{-4} mg CO/hr at 350°C. The lowest reaction rate found under similar conditions with the pile at power was 3.6×10^{-3} mg CO/hr (0.03 wt percent/1000 days) demonstrating the increase of reaction rate that occurs under pile irradiation. The rate of reaction was found to be proportional to pile power and therefore to neutron flux and dose rate over the range studied. The reaction rate of the radiation induced reaction is only doubled upon increasing the temperature from 30° to 300°C. It was anticipated that the thermal reaction would mask the effect of radiation at temperatures of 500°C and above due to the small temperature coefficient. A marked effect was noted upon reversing the gas flow. The geometry was such that under normal flow the gas passed through a small

volume, over the graphite and then through a large volume. The reaction rate was doubled if the CO_2 passed through the large volume before passing over the graphite. It was concluded from this large effect that the reaction is controlled more by energy deposition in the gas than by energy deposition in the graphite. This is effected by decomposition of CO_2 by pile radiation to oxidizing intermediates which may either react with graphite or reform CO_2 . It was assumed also that the mean life of the intermediates must be less than or comparable with the time of circulation of the gas (one min). Otherwise, the position of the large volume with respect to the graphite would not have produced the marked effect on the reaction rate. An increase in pressure should increase the reaction rate since the mass of gas irradiated would be increased. Experimentally, it was found that a seven fold increase in pressure (28 to 220 cm of Hg) increased the reaction rate by a factor of less than three. The rate of reaction would be expected to be low at a very low flow rate since most of the intermediates will have recombined before they reach the graphite, even though nearly every intermediate reaching the graphite should react. The time of contact over the graphite will be short at high flow rates and therefore a low rate of reaction should also occur under these conditions. At intermediate flow rates an appreciable fraction of the intermediates should reach the graphite as well as react with it producing a maximum reaction rate at some optimum flow rate. For the experimental conditions used the rate of reaction decreased with increasing flow rates above one g/min indicating that at flow rates greater than one g/min the optimum flow rate had been exceeded. A single experiment was run in which 0.87 percent of water vapor was added to the CO_2 . This addition had no effect on the rate of reaction. The exact steady-state concentration of CO at one atmosphere pressure of CO_2 probably lies between 0.034 and 0.6 percent at 350°C . The equilibrium CO concentration in the absence of radiation is 0.27 percent at this temperature. One cannot say whether radiation has shifted the steady-state CO concentration or not, but it is believed that the radiation equilibrium will depend on the geometry of the system. One of the conclusions reached by the British workers follows: "The observed variations in rate caused by altering the geometrical conditions of experiment and the possibility that graphite was not being removed uniformly from the samples in these experiments suggest that it might be quite misleading to attempt any

calculation of burning rates of graphite in a gas-cooled reactor from these data. A model-channel experiment is being conducted in B.E.P.O. under conditions simulating those in P.I.P.P.A. type reactors more closely."

A CO₂-cooled pile loop designed to simulate a model channel in a Calder Hall Reactor has been run in BEPO with another to follow in DIDO. It consists of a closed loop containing graphite sections with fuel elements inserted in the graphite tube. Removable portions of the tube serve as graphite samples. The information recorded here is due to Dr. Anderson of Harwell.³⁰ The flux for the loop was 10^{12} compared to 5×10^{12} for Calder Hall, the inlet temperature of 140°C and outlet temperature of 350°C were comparable and the flow rate for the model channel was 1/2 that for the reactor. The gas to graphite volume ratio were equal as was the in-pile gas volume to out-of-pile gas volume ratio. A steady-state value of about one percent CO was reported. The weight loss distribution compared to the temperature and flux distribution is given in Fig. 2.2.

The maximum weight loss occurred on the inlet or lower temperature side rather than on the high temperature side of the centerline. This indicates that more back reaction occurs on the high temperature side probably because the back reaction is more temperature sensitive than the forward reaction. The addition of ten percent CO resulted in small weight gains for all samples and formation of dark deposits on some surface including some areas in the outer circuit after three months exposure.

J. S. Nairn of Windscale reported some information on Calder Hall operations at the same conference.³⁰ The production of CO has been found to be proportional to the power density with the steady-state conditions limited mainly by loss of gas through leakage. At 196 Mw power and 100 psi, the CO leveled off at about 0.35 mole percent in both Calder Hall reactors after initial startup. The loss of graphite was found to be linear with CO₂ pressure and was about 0.13 tons/yr at 100 psi pressure. No positive evidence for the deposition of carbon from CO has been observed.

³⁰ I. F. Zartman, U.S./U.K. Graphite Conference held at St. Giles Court, London, December 16-18, 1957 (Section 1-6).

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FIGURE 2.2 WEIGHT LOSS DISTRIBUTION AS A FUNCTION OF CHANNEL POSITION.

2.3 Effectiveness of Graphite Coatings

Coatings on graphite have been under study for many years and at many laboratories around the country. The coatings, which are designed to improve erosion resistance, corrosion resistance, or both, have generally been restricted to the carbides, or to carbide-metal mixtures. The techniques for applying them are many, but in general they will fall into the following categories: vapor deposition, flame spray, slurry coatings, and liquid metal immersion.

In general, the coatings designed to protect graphite from erosion have been more successful than those designed to prevent corrosion or reaction with an atmosphere. Until very recently there have been no coatings which gave any sort of a permanent resistance to oxidation. The popular application for graphite coatings, at present, is in the missile field. The requirements for this application are high as regards refractoriness, resistance to spalling and thermal fracturing, and erosion and corrosion by high velocity gases, but the life requirements are very short so that low permeability coatings can be attractive if they can meet the other requirements.

Applications to gas-cooled power reactors require coatings which give protection over very long periods of time. Requirements for refractoriness and resistance to thermal shock will be low compared to missile standards. In the particular case of the CO_2 -cooled reactor, however, the coatings must be essentially impermeable to effectively inhibit the reaction of CO_2 with graphite. It can be seen that a permeable coating will subject the graphite base material to the oxidizing action of CO_2 , with gradual removal of graphite and serious effects on the mechanical properties. This in turn may result in spalling of the coating and exposure of more of the graphite.

The coating of graphite has been studied by Minnesota Mining and Mfg. Co., Horizons, Inc., Battelle Memorial Institute, ORNL and several other laboratories. The ZrC coating developed by Horizons for Project Rover³¹ is sufficiently refractory and resistant to spalling and corrosion, but is permeable. A similar coating of zirconium carbide-zirconium, as received

³¹E. C. Hirakis, "The Continuation of Investigation of Coatings of Graphite Tubes for High Temperature Corrosion and Erosion Resistance", (undocumented) Progress Report from Horizons, Inc. to Director, University of California Radiation Laboratory.

from the Stanford Research Institute, was somewhat less permeable but not completely protective. Further development of these two coatings may be successful, however. Various other boride-, oxide-, and carbide-base coatings of a protective nature have been prepared, but are still experimental or under development.

The most promising coating for graphite at the present time is the silicon-silicon carbide coating developed by Minnesota Mining and Mfg. Co. It is the only coating which offers a good possibility of being non-permeable as well as fulfilling the other requirements of low cross section, good thermal conductivity, refractoriness, and resistance to thermal fracture and spalling. This coating has been applied to graphite only on a laboratory scale, and has been satisfactory only on a special grade of isotropic graphite produced by the Great Lakes Carbon Corporation. Additional samples of this coating are being obtained for evaluation. However, there are always the questions as to whether the process could be scaled up to produce the large quantities and sizes required, and whether sufficient special graphite could be obtained.

While the various available coatings for graphite should certainly be tested, there is a good possibility of using pure silicon carbide sleeves for the prototype reactor. This material has the inherent advantage of complete resistance to corrosion by CO_2 , as well as being refractory and having a low cross section. There exists also a well established fabrication technology, and production facilities are available for fabricating sleeves of the required sizes.

2.4 Influence on Reactor Performance

a. The Relationship of Available Data for the Carbon Dioxide-Graphite System to GCR. An examination of the available data for the CO_2 -graphite reaction suggests that it is impossible to formulate any reliable estimate of the losses of graphite or the deposition of carbon either for the reactor as a whole or for any particular region within the reactor. The Hanford data have very limited application and although the British data obtained for the model channel experiment are the type required, only very limited information is accessible from these studies. It is believed that some consideration of the various factors affecting the reaction may be of benefit in planning some of the projected experimental programs designed to obtain the needed information.

The evidence at hand suggests that somewhere in the temperature region 550-600°C the thermal reaction of CO_2 with graphite controls the rate of oxidation of the graphite by overshadowing the radiation-induced reaction. If it is assumed that the maximum graphite temperature in GCR does not exceed 550°C then the radiation-induced reaction is the one to be considered. This reaction apparently has a very small temperature coefficient so far as the forward part of the reaction is concerned. If one uses the value of 0.13 tons/yr for the graphite burnout in the Calder Hall reactor, the higher temperature of the GCR probably would not increase this amount by more than 50 percent.

The distribution of the graphite burnout is also of interest. Hanford experience and results from the British model channel experiment suggest that the maximum oxidation rates occur between the gas inlet and the center of the reactor. In the case of the model channel experiment the maximum flux occurred at the center of the channel and the maximum temperature at the outlet area. Both the temperature and flux peaked near the center line in the Hanford facility. Thus, the maximum reaction rates were observed in relatively cool areas outside the maximum flux-area. This behavior may be explained by a mechanism involving the decomposition of CO_2 by radiation with the formation of oxidizing intermediates, probably ozone and/or atomic oxygen, which have a limited life. These intermediates may either react with the graphite or be destroyed by reaction with CO or its decomposition products. The model channel results suggest that between the inlet and center the oxidizing intermediates are most effective in reacting with

graphite. In other words their destruction by CO and its decomposition products is not as great in this area as farther downstream where the concentrations of CO and its decomposition products are larger because more CO has been formed and after formation has been exposed to increased dosages of radiation. The important point, however, is that the reaction is controlled more by energy deposition in the gas than in the graphite. One may guess that the weight loss distribution in the channels of GCR will be similar to that observed in the model channel experiment.

An increase in pressure of the CO_2 would be expected to increase the oxidation rate of the graphite due simply to the fact that more oxidizing intermediates would be produced per unit volume. It is not clear, however, just what relationship exists between the rate of oxidation and pressure in the high pressure regions. Calder Hall experience suggests that the loss of graphite is linear with respect to the CO_2 pressure, but data demonstrating this are not accessible. The glass loop experiments in BEPO showed that an increase in pressure from 28 to 220 cm of Hg increased the reaction rate threefold. The geometry of the system probably influences the pressure effect to a considerable extent. It appears likely that an increase in the CO_2 pressure from 100 to 300 or 400 psi would cause a significant increase in the reaction rate. Whether or not the pressure effect is linear in this pressure region is not known, but it is believed that a three- or four-fold increase in the reaction rate is the maximum likely to occur upon increasing the pressure from 100 to 300 or 400 psi and the effect may be considerably less.

The effect of flow rate on the oxidation rates of graphite also is obscure particularly at high flow rates. It would be expected that the mean life of the oxidizing intermediates and the contact time required for the reaction of the intermediate with the graphite would determine the particular flow rate (constant pressure) required to produce the maximum oxidation rate. At low flow rates the intermediates will have ample time to react with the graphite, but many of them will have reacted in the gas phase before reaching the surface. At the other extreme the contact time is short, but many intermediates survive to reach the surface. The maximum reaction rate would be expected to occur between these extremes. This behavior was found experimentally in the glass loop exposed in BEPO. No information is accessible on the effect of flow rate on the oxidation

of graphite in either the model channel or Calder Hall reactor. One might assume that the flow rate in Calder Hall is sufficiently high that the oxidation rate is less than the maximum attainable at some lower flow rate. If this were the case then the projected flow rate for GCR might influence the reaction rate only to a small degree. This is probably oversimplifying the case, however, since the pressure and geometry are tied in with the flow rate.

Although one might make some estimate of the initial rate of oxidation this rate would decrease with increasing CO concentrations and eventually some radiation-induced equilibrium or steady-state conditions would prevail. In actual reactor operations the leakage would influence the steady-state conditions. The large leakage of the Calder Hall reactor mainly determines the steady-state conditions, namely a CO concentration of 0.35 percent at 196 Mw and 100 psi. At a state in which the CO concentration is constant its rate of formation is balanced by its decomposition plus that lost through leakage. The decomposition of CO under irradiation proceeds through polymerized suboxides and the mechanism is believed to be quite complex. The effect of temperature on this decomposition is not understood and consequently the regions in which the decomposition attains a maximum is open to speculation. This decomposition could be of considerable importance if it is promoted in the cooler regions of the reactor resulting in deposition of solids on the heat exchanger surfaces. (The catalytic decomposition of CO by the metal surfaces could conceivably occur although it does not appear likely in view of the inactive state of the metal surfaces present.) Calder Hall experience sheds no light on this question since no positive evidence for the deposition of carbon has been observed despite an estimated burnout of 0.13 tons of graphite per year. It is true that a good share of the carbon has been lost from the system as CO due to the high leak rates. The model channel experiment indicates that some burnout of graphite occurs along the entire length of the channel and if this is true it suggests that carbon is deposited outside of the channel.

As mentioned previously, the steady-state value for CO in the Calder Hall reactors is about 0.35 percent and is limited mainly by the leak rate. A steady-state value of about one percent CO was found in the model channel at a flux $1/5$ and a flow rate $1/2$ that prevailing in the Calder Hall reactors.

This difference in CO levels is to a large degree due to the high leak rate present in the Calder Hall reactors. The glass experiment in BEPO gave a steady-state concentration of CO somewhere between 0.3 and 0.64 percent. Calder Hall operations indicate that in reactor operation the leak rate may play a dominant role in establishing the steady-state conditions and the same case may prevail for GCR. If the burnout of graphite is of prime concern the leak rate should be kept as low as practical since loss of CO favors the forward reaction. On the other hand if the formation of solids from the decomposition of CO is troublesome, leak tightness of the reactor is not so important.

In addition to the leak rate such factors as maximum temperature, temperature gradients, radiation dosage, pressure, flow rate, resonance time in relation to transit time, gas to graphite volume ratio and in-pile gas volume to out-of-pile gas volume ratio must be considered in arriving at the steady state conditions.

b. Influence of Exit Gas Temperature on Performance. The mean temperature of the gas leaving the reactor is likely to be set as a result of a compromise between reactor performance and steam system performance. Since the ultimate limiting temperature in the reactor is generally the fuel surface temperature, good reactor performance does not favor very high gas outlet temperature. This is so because high heat transfer coefficients with gas coolants tend to require high gas velocity, and hence high pumping power, and therefore, within limits, it proves to be more advantageous to secure good heat transfer performance by means of a large temperature difference between fuel and coolant. Good steam system performance on the other hand is promoted by high gas outlet temperature, which not only permits improvement in the thermal efficiency of the power recovery cycle, but also may lead to substantially smaller, and hence less costly, power recovery equipment. This improvement results from higher temperature difference in the steam generator, from reduced mass flow of the coolant with consequent reduction in pressure drop, from higher steam temperature and pressure with lower flow rate, or from some combination of these effects. Offsetting these advantages, in part, is the possibility that higher temperatures will require the use of more costly materials, both in the steam system and in the gas piping.

To ascertain what combination of the very numerous variables of the entire system will produce lowest cost power, and to determine how sensitive the power cost is to such variables as the gas outlet temperature, is a difficult and tedious task, requiring the evaluation of very many individual cases. The problem is seriously complicated by the fact that most of the variables commonly used to describe the reactor and power recovery system are not independent, so that the results that are obtained and the conclusions that are drawn from a parametric survey depend critically on the assumptions made and on which variables are held constant.

Several studies have been made from which one may attempt to determine the effect of gas outlet temperature on power cost. One of these is the survey made by the General Electric Company, Hanford³², in which the system is characterized by the gas outlet temperature, the reactor ΔT , the total thermal power, and the tube specific power in kw/ft of fuel element. In this study, the cooling channel diameter was held fixed at one-half of the equivalent lattice cell diameter for all cases studied, and the number and size of fuel rods per fuel element cluster was determined by the allowable wall temperature and the specified moderator to uranium ratio. The fuel surface temperature was fixed at 1500°F. Subject to these restrictions, it was concluded that the power costs are not very sensitive to gas outlet temperature, in the range of 1000 to 1200°F when other variables are optimized, and indeed for some cases the net power costs appear to be lower with 1000°F than with 1200°F outlet temperature.

A parametric survey of reactor performance has been initiated at ORNL, significant independently chosen variables being gas pressure, inlet and outlet gas temperature, fuel surface temperature, number of fuel rods per cluster, and the gas channel physical diameter. Among the preliminary results obtained so far are the contribution of the reactor and gas system cost to the net power cost, as a function of gas outlet temperature for various values of the other variables. Some of the results are presented in Table 3.1, which permits a comparison of performance at 1000° and 900°F gas outlet temperature, with a 1200°F maximum fuel element surface temperature. Over a range of nearly a factor of two in heat output per channel,

³²"A Parametric Study of the Gas-Cooled Reactor Concept", March 1, 1958, HW-54727.

it can be seen that the lower gas temperature results in slightly better reactor performance. The effect is more pronounced, the more heat is extracted per channel, and is slightly greater for CO₂ than for helium.

On the other hand, a study of the effect of gas outlet temperature on steam system performance was made in connection with the GCR-2 study,³³ which indicated a cost incentive of about 0.25 mills/kwhr per 100°F, to increase the gas temperature. Thus, the opposite effects of gas temperature on reactor and steam system performance will tend to produce a broad minimum in power cost with respect to gas temperature. The location of this minimum will certainly depend on other design variables, and has not been determined exactly in the case of GCR-2. It would appear, however, that the power cost is not very sensitive to gas temperature within 100°F of the optimum.

³³"The ORNL Gas-Cooled Reactor", Part 3, p. 9.24, Fig. 9.4, ORNL-2500.

3. INFLUENCE OF COOLANT CHOICE ON REACTOR PERFORMANCE

3.1 Basis for Comparison of Coolants

One of the criteria of suitability of a given fluid as a nuclear reactor coolant is its ability to provide high rates of heat transfer from the fuel to the coolant stream and high rates of transport of heat from the reactor to the thermodynamic cycle. A distinction must be made between these two processes since they are functions of different properties of the gas. The former is dependent on the conductivity and the Reynolds and Prandtl numbers; the latter is a function of the molar heat capacity. Carbon-dioxide, for instance, has excellent heat transport properties, but is relatively poor from a standpoint of heat transfer, while helium is just the reverse. The amount of pumping power required to remove a given quantity of heat from a reactor under a given system pressure level and temperature conditions is, therefore, an index of its quality as a coolant.

The expression $mPr^{2/3}/(mC_p)^3$, where m is the molecular weight of the gas, Pr the Prandtl number and mC_p the molar heat capacity, has often been presented as an index of heat transfer efficiency of gases.^{34,35} If one examines the derivation of this expression, however, it will be found that it involves the assumption that the effective diameter of the heat transfer channel is an independent variable. This cannot be the case for a given fuel element configuration if the heat flux is fixed together with the temperatures of the fuel element surface and the coolant stream. It has been shown³⁶ that under the above constraints the channel equivalent diameter has a strong influence on the pumping power requirements.

The best comparison of different coolants is on the basis of unit net power costs for two systems separately optimized for the two gases with care being taken that no parameter be allowed to vary except as required by specific coolant properties. A fair comparison cannot be made by, for

³⁴"Army Mobile Power Plant", Sanderson and Porter, July 15, 1955, Report No. 1959-1.2.

³⁵"Journal of the British Nuclear Energy Conference", 1956, 1, p. 227.

³⁶G. Samuels, "Comparison of Gases for Use as the Coolant in a Gas-Cooled Reactor", ORNL CF 58-4-108 (see Appendix V).

instance, allowing a higher fuel element surface temperature in one case than in the other. Since this method is too involved to permit a wide range of comparison, the present cost estimates are based on that portion of the plant which are chargeable directly to the primary coolant circuit. This includes costs of the pressure vessel, graphite, ducting, blower, and blower power. Costs of fuel, steam generators, and turbine — generator machinery are not closely related to the properties of the coolant and are excluded. When the range of independent parameters has been sufficiently narrowed on this simplified basis the more important cases can be examined on the basis of total unit power cost.

Table 3.1 gives the partial power cost as described above together with channel equivalent diameter and percent gross electric power used for pumping for carbon-dioxide and helium for different coolant outlet and fuel element surface temperatures. The cost information is plotted vs. power per channel on Fig. 3.1.

It can be seen that for any given set of heat flux and temperature conditions over the ranges covered that the use of helium results in consistently lower power costs than does carbon dioxide. The difference is small at heat rates of about 0.8 MwT*/channel, but rapid divergence occurs at the higher rates. Note that the GCR-2 system at the design output of 700 MwT if adjusted to 485 psia would correspond to an average heat rate of 0.7 MwT/channel. However, the GCR-2 study showed that reduced power costs would result if the output were increased to approximately 1100 MwT which corresponds to 1.1 MwT/channel. At this point the partial power cost of a carbon dioxide system is approximately 40 percent higher than that of a helium system.

Aside from the question of chemical compatibility of carbon dioxide and graphite it appears that the advantage of using helium is not great below heat rates corresponding to approximately 0.7 MwT/channel. Current interest, however, is in heat rates up to twice as great. There is some interest in reactors having low heat rates per channel for prototype or pilot plant purposes, but to be of any value a prototype must duplicate conditions which will exist in the later, full size, plant. It would therefore seem desirable for a prototype to use the same coolant as the projected reactor.

*MwT denotes megawatts of thermal power.

Table 3.1 Some Comparisons of Partial Power Costs* for Helium
and CO₂-Cooled Power Reactors

Gas Outlet Temperature (°F)	Capsule Operating Temperature (°F)	Power Per Channel (Mw)	Equivalent Diameter (in.)		Pumping Power (percent of total thermal output)		Cost (mills/kwhr)	
			He	CO ₂	He	CO ₂	He	CO ₂
1000	1300	0.80	3.74	3.35	2.40	3.72	1.04	1.07
		0.97	3.69	3.30	3.75	5.93	0.935	1.01
		1.40	3.60	3.23	8.69	14.00	1.09	1.68
1000	1200	0.82	3.41	3.10	4.17	6.94	1.10	1.20
		1.00	3.38	3.05	6.65	11.10	1.07	1.37
		1.44	3.30	3.00	15.64	26.50	2.05	4.69
900	1200	0.82	3.85	3.42	3.72	5.76	1.09	1.15
		1.00	3.80	3.38	5.78	9.09	1.04	1.21
		1.44	3.71	3.31	13.4	19.5	1.52	3.15

*Costs are calculated on the basis of 700 channels 20 ft long with seven 0.8 in. dia fuel rods. Coolant gas pressure taken as 485 psi and inlet temperature as $T_1 = 450^\circ\text{F}$. Cost includes reactor and gas system.

Based On:
700 Channels 20 ft. long
7-0.8 in dia Fuel Rods
465 psia Mean System Press.

FIGURE 3.1 POWER COST ATTRIBUTABLE TO
COOLANT CIRCUIT

3.2 Interchangeability of Coolants

The physical properties of the most attractive coolants (hydrogen, helium, water vapor, or carbon-dioxide) are such that they do not permit interchangeability in a given reactor system at the same heat flux and temperature conditions. Hydrogen and helium are close to being interchangeable, as are carbon dioxide and water vapor, but interchanging the light and heavy gases would necessitate major changes in the operating conditions or the system components.

The volumetric capacity requirements of the coolant circulating blowers is directly related to the volumetric heat capacity of the coolant, and the blower head is affected by these properties of the coolant which influence the heat transfer coefficients. Interchanges between the light and heavy gases mentioned above call for changes in head and flow which cannot be accomplished in a turbo-dynamic blower by speed change alone. It would be necessary to change the blower design and replace the drive motors with units of different rating if helium and carbon dioxide, for instance, were interchanged.

For small reactor systems wherein the required blower capacity is not greater than approximately 10,000 cfm (per unit) it might be advisable to use positive displacement blowers if coolant interchangeability is contemplated. In these machines the head is independent of rpm, and volumetric capacity can be changed simply by changing speed.

4. COMPATIBILITY OF FUEL-CLADDING MATERIALS WITH CARBON DIOXIDE

The compatibility of a cladding material with a coolant gas must be evaluated from two standpoints: (1) the maximum temperature at which long-time operation will be feasible, and (2) the hazards associated with a short-time excursion to much higher temperatures than the operating temperature. An ideal system from the compatibility standpoint is one in which the temperature limits are set by considerations other than the reaction between the cladding material and the coolant and in which no hazards exist. This section is a discussion of the degree to which these conditions are met with magnesium alloys, beryllium, and the austenitic stainless steels in contact with CO_2 in a reactor environment. The compatibility of impervious or coated graphite cladding materials is the same as that for the previously discussed sleeve material and therefore no further discussion on these materials is required.

The discussion which follows is based almost entirely on out-of-pile oxidation information. There is an unfortunate lack of information on the effects of radiation on the oxidation characteristics of metals. However, the general experience has been that radiation does not produce major increases in the oxidation rates of structural metals. Thus the effects of radiation are assumed to be negligible.

4.1 Magnesium Alloys

Discussions have previously been given of the existing information on the reaction between Mg and CO_2 in a reactor environment.^{37,38} The following conclusions may be derived from these reports:

- (a) Magnox C (one percent Al, 0.05 percent Be, remainder Mg) is the most promising magnesium alloy for use as a cladding material. In bulk forms it is compatible with CO_2 to temperatures above its melting point of 645°C .
- (b) A possible hazard exists with Magnox C in a CO_2 -cooled reactor, if the temperature of the cladding exceeds the melting point.

³⁷ H. Inouye, "The Reactions of Magnesium and Magnesium Alloys with Gases at High Temperatures", ORNL CF 58-1-93, January 20, 1958.

³⁸ J. L. Scott, "An Analysis of the Mg- CO_2 Reaction in Terms of a Reactor Environment", ORNL CF 58-5-100, May 30, 1958.

- (c) The low strength of magnesium alloys at elevated temperatures limits their usefulness as cladding materials.

4.2 Beryllium

Huddle³⁹ has reported that the oxidation resistance of beryllium is excellent below 750°C. This temperature is above the temperature at which swelling has been observed to occur (700°C),⁴⁰ so that the compatibility of beryllium with CO₂ may be considered to be good. Swelling is caused by helium produced by a neutron-alpha particle reaction. Since beryllium does not ignite in air, there is no hazard associated with its use in contact with CO₂ in a gas-cooled reactor.

4.3 Austenitic Stainless Steels

The austenitic stainless steels are particularly noted for their oxidation resistance and high elevated-temperature strength so that they come to the foreground as fuel cladding materials for operating temperatures in the range 600-1100°C, despite their relatively high neutron absorption cross sections. The improved properties of the austenitic stainless steels relative to ordinary steels are derived from beneficial additions of nickel and chromium. Two basic types of stainless steels are in common use: the 18 Cr - 8 Ni types and the more expensive 25 Cr - 20 Ni types. In air the limiting temperature for 18 Cr - 8 Ni steels is about 870°C and for 25 Cr - 20 Ni steels it is about 1150°C.⁴¹ In addition the oxide film, which is responsible for the excellent oxidation resistance, does not spall off when the material is thermally cycled.

The behavior of the austenitic stainless steels in a reactor cooled with CO₂ is difficult to predict. Pure CO₂ is oxidizing to steels and stainless steels at all temperatures⁴² and the rate of scaling attack in

³⁹R. A. U. Huddle, "Oxidation Behavior of Reactor Materials", Problems in Nuclear Engineering, 1, Pergamon 1957, p. 79.

⁴⁰H. M. Finniston, Private Communication to J. L. Scott, November 1, 1957.

⁴¹H. H. Uhlig, Corrosion Handbook, Wiley, 1948, p. 644.

⁴²J. B. Austin and M. J. Day, "Chemical Equilibrium as a Guide to the Control of Furnace Atmospheres - A Review of Equilibrium Data", Controlled Atmospheres, ASM, 1952, p. 20.

CO_2 is less than in air⁴³. However, the reaction between CO_2 and the graphite moderator will produce some CO, as discussed in Sect. 2. Depending on the CO_2/CO ratio, the temperature, and the composition of the stainless steel, the resulting atmosphere could be oxidizing or reducing. Since the temperature would be expected to fluctuate widely during the course of operation of a reactor, and the fuel elements may be periodically moved to increase the fuel burnup, the stainless steel might be exposed to cyclic oxidizing and reducing conditions.

Such conditions could result in a variety of harmful effects to the austenitic stainless steel. If the CO content were sufficiently high in a localized region of the reactor the stainless steel might be heavily carburized. Although carburization increases the creep strength of the stainless steel, it also results in an embrittlement. Further, since chromium carbide is more stable than iron carbide, the chromium reacts preferentially with carbon when carburization occurs. In this way the amount of chromium in solid solution is reduced. Since 12 percent of chromium in solid solution is required for good oxidation resistance, a heavily carburized stainless steel exposed to an oxidizing atmosphere would be expected to be rapidly attacked.

Another harmful effect could result from the reducing action of CO- CO_2 mixtures. Since the protective oxide film on stainless steels contains some Fe_2O_3 as well as the stable spinel $\text{FeO}\cdot\text{Cr}_2\text{O}_3$, the reduction of Fe_2O_3 by CO could loosen the protective film and result in selective internal oxidation of chromium in the alloy. Such attack, which would seriously impair the mechanical properties, would be similar to the phenomenon called "green rot" which has been observed in nickel-chromium alloys exposed to a CO atmosphere.⁴⁴

A third type of attack which might occur when stainless steels are exposed to alternating oxidizing and reducing conditions is "metal dusting". Metal dusting is the mechanical removal of particles of metal or oxide from the surface due to formation of solid carbon beneath the particles in regions

⁴³Uhlig, Op Cit p. 636.

⁴⁴H. R. Copson and F. S. Lang, "Some Experiments on Internal Green Rot Oxidation of Nickel-Chromium Alloys", The International Nickel Company, Inc., Research Laboratory, TP: No. 277, Submitted for publication in Corrosion, January 1958.

where the decomposition of carbon monoxide is possible ($400-700^{\circ}\text{C}$). The process has been observed to result in the rapid deterioration of metal rails leading into annealing furnaces in regions where the opening and closing of doors leads to cycles oxidizing and reducing conditions. The mechanism for this type of attack is similar to that described by Baukloh for the rapid attack of cast iron in CO .⁴⁵ In the present case on the oxidizing cycle, a film would be formed which would contain both $\text{FeO}\cdot\text{Cr}_2\text{O}_3$ and Fe_2O_3 .⁴⁶ On the reducing cycle the Fe_2O_3 would be reduced to metallic iron. The $\text{FeO}\cdot\text{Cr}_2\text{O}_3$ is sufficiently stable that it would not be reduced even in pure CO . If the mixture of CO and CO_2 happened to be sufficiently reducing, some of the CO might be reduced to solid carbon at the metal surface in such a way that the resulting stresses would mechanically fracture the remaining oxide film. A second oxidation cycle would result in the formation of a new film with the consumption of more metal. A sequence of such cycles could lead to the destruction of the capsule wall.

A consideration of the above effects indicates that from the metallurgical standpoint the CO concentration in a CO_2 -cooled reactor should be limited to the order of one percent. The difficulties associated with maintaining these conditions are connected with the development of a completely impervious coating on the graphite, as previously discussed. If the atmosphere consists only of CO_2 , the life of the stainless steel in the reactor environment should exceed its life in air. Thus, type 304 stainless steel would be expected to perform satisfactorily up to 870°C and no hazard exists up to the melting point of the alloy (1400°C).

If an alloy with higher elevated-temperature strength than type 304 is required, type 310 stainless steel (25 Cr - 20 Ni) is recommended since its oxidation resistance is superior to that of type 304. Type 316 stainless steel (18 Cr - 8 Ni - 2 Mo) is not recommended for use in CO_2 , because of the possible accelerated oxidation due to the highly corrosive MoO_3 ,

⁴⁵W. Baukloh, "Concerning the Destructive Effects of CO and Gases Containing CO ", Die Chemische Fabrik, 13, 101 Marz 1940.

⁴⁶H. M. McCullough, M. G. Fontana, F. H. Beck, "Formation of Oxides on Some Stainless Steels at High Temperatures", Trans. ASM, 41, 1951 p. 404.

which may be formed. This type of corrosion, which has been observed in all molybdenum-containing steels, is most likely to occur in those regions of the reactor where the localized gas flow rate is low.⁴⁷ For the same reason, alloys containing vanadium as a strengthener are to be avoided.

If type 310 stainless steel were selected as the cladding material, extensive studies would have to be carried out to determine the susceptibility of the material to sigma phase embrittlement under conditions of the reactor operation.

⁴⁷A. deS. Brasunas and N. J. Grant, "Accelerated Oxidation of Metals at High Temperatures", Trans. ASM, 44, p. 1117, 1952.

APPENDIX I

A. Telephone Transcript, 5-28-58

To: Mr. Paul V. Mullins of the Bureau of Mines, Helium Production Station at Amarillo, Texas, Phone No: DRake 4-0376

From: M. Bender

Subject: Helium Storage

The Bureau of Mines has storage capacity for 7.5×10^6 scf of helium gas. The gas is stored in steel pipe with welded caps $10\frac{3}{4}$ in. diameter by 90 to 180 ft long at 1500-1800 psi. The tanks are interconnected with welded pipe using packed valves. Mr. Mullins believes that their losses are in the neighborhood of 0.1% of the system volume/day. He stated that a loss of 1.0%/day could be held within any system designed for high-pressure gas storage using packed valves, even without special testing.

The Bureau of Mines ships helium in tank trucks made up of forged steel vessels with packless valves. They experience essentially no leakage in their systems.

Mr. Mullins was asked about the availability of helium overseas. He stated that the Bureau of Mines does not know of any significant helium deposits outside of the U. S. They have investigated only a few sources, however, and Mr. Mullins expressed the opinion that insufficient investigation has been carried out to establish the availability of the gas. He would be interested in testing additional samples if they were made available.

He was asked about the export problems of shipping helium overseas. Mr. Mullins reported that some helium is currently shipped overseas by commercial gas suppliers under special export licenses which could be issued with the approval of the Interior and State Departments. He estimated the quantity to be several hundred thousands scf/year. He thought it was shipped in small cylinders and he did not know the cost of the gas.

Mr. Mullins was also asked about the feasibility of helium separation by diffusion using the new glass ceramic material recently developed

by Bell Laboratories. He and Dr. Siebel of the Bureau of Mines had visited Dr. McAfee of Bell Laboratories recently to investigate the potential of the idea. The separation is carried out at high pressures (15,000 psi to 30,000 psi). It appeared to require a large amount of separating surface. He also felt that a means for recovering the energy of compression might be necessary if the process were to be economically practical. He thought it might have definite merit for obtaining high-purity gas.

B. Telephone Transcript, 6-2-58

To: Mr. S. T. Robinson of Sanderson and Porter Company
From: G. C. Robinson
Subject: Integrity of Helium-Handling Equipment

Mr. S. T. Robinson indicated that he has had no personal experience on the integrity of helium-handling equipment, but drawing upon the experience of others, he does not believe any difficulty exists. He is now correlating data of over 100 manufacturers of helium-handling equipment in the missile and wind tunnel business and expects to have the report completed in approximately one month. He would be willing at that time to exchange data that he has accumulated with such data as we have available from plant experience at Y-12 and K-25. He also cited a GE report which indicates leakage in some vessels to be more severe for CO₂ and N₂ than helium and indicated that he would write us soon concerning the number of the GE report.

C. Telephone Transcript, 5-28-58

To: Charles Coffman, Lakehurst Naval Air Station, Lakehurst,
New Jersey
From: G. C. Robinson
Subject: Helium Storage

Mr. Charles Coffman of the Lakehurst Naval Air Station provided information on the type, capacity and integrity of vessels for storing

helium at the base as follows:

1. 1,000,000 scf low pressure, 6-1/2, 8-1/2, 10 in. H₂O gasometer built by Cruse-Kemper Company in 1921. Transfer records indicate no leakage.
2. 40 4-ft dia x 40-ft long forged cylinders made by Bethlehem Steel installed in 1920. Leakage does not exceed 1 to 2%/year. No leakage observed for cylinders. Only leakage observed has been at packed valves.
3. In 1927, 23 banks of standard cylinders, 927 cylinders/bank, were installed. The only leakage observed was at screwed and brazed connections. There are 10 banks in use at present, storing 1,250,000 scf at 1800 psig. Leakage according to transfer records does not exceed 1 to 2%/year.
4. 163 2-ft dia x 80-ft long cylinders with a storage pressure of 750 psi, manufactured by A. O. Smith, are in use. Leakage has not exceeded 1 to 2%/year.
5. Also, recently 348 2-ft dia x 80-ft long cylinders manufactured by National Tube Company for storage at 1680 psi have been installed. Leakage has not exceeded 1 to 2%/year.

No leakage has been experienced through any of the cylinders.

Packed valves, screwed or brazed joints have provided all the leakage difficulty experienced. The Nordstrom hyperseal valve has been the most satisfactory valve used; however, some leakage is experienced through it.

D. Telephone Transcript, 5-29-58

To: Dr. F. C. Balcors, Air Reduction Company, New York, New York,
Phone No: Murray Hill 2-6700

From: M. Bender

Subject: Helium Transportation

Air Reduction Company has no specific experience with large shipments of helium overseas. Dr. Balcors stated that the conventional railway tank cars could be adapted to overseas shipments. These can hold 250,000 scf and cost about \$100,000 each.

Liquification is also practical and its cost is moderate. Mr. R. E. Brown of the Bureau of Aeronautics, Washington, D. C., and Mr. B. W. Birmingham, Assistant Manager, Cryogenic Engineering Laboratory, Bureau of Standards at Boulder, Colorado, are suggested as authoritative sources of information on helium shipments and handling.

E. Telephone Transcript, 6-3-58

To: Mr. R. E. Brown, Helium Distribution Department, Bureau of Aeronautics

From: M. Bender

Subject: Helium Distribution

Mr. Brown has been connected with helium shipments for the Navy Department and currently with the Bureau of Aeronautics. He is not familiar with any shipments which approach the magnitude of the gas-cooled reactor requirements. He is currently active in the development of helium liquification equipment. A 35,000-scf/d unit now is being planned for installation at the Lakehurst Naval Air Station. A liquification unit to produce 100,000-300,000 scfd of helium is planned for Amarillo in 1960. Part of this capacity could be available for overseas shipments if desired.

Lapsed time is important in liquid helium systems because helium pressure builds up in the shipping container. The 2-week shipping time from Amarillo to Lakehurst is not too long for liquid helium shipments. Longer periods of time could be accomplished by designing for higher pressures or providing more gas containment volume.

Mr. Brown did not know the cost of shipping overseas. Liquid helium transport by air is not cheaper than gaseous helium transport by water. It is much quicker. Transportation of liquid helium by water would probably be cheaper than transportation of helium gas.

F. Telephone Transcript, 5-28-58

To: Mr. E. F. Love, Hypersonic Wind Tunnel Station, NACA Laboratories, Langley Field, Virginia

From: M. Bender

Subject: Helium Storage

A helium-supplied hypersonic wind tunnel is being installed at Langley Field. Mr. Love does not have details of the volume, pressure, or the effectiveness of the recovery system. The system was studied by NACA engineers and has been shown to be economically feasible.

pressure- and vacuum-tested before accepting for operation. Ring flanges are taped and "sniffed" for leaks periodically during operation.

Stainless steel is used to maintain the high degree of purity of the gas and also eliminates pressure regulator trouble from flakes of rust, etc., from black iron, or steel pipe.

Losses are essentially zero from the standpoint of system leakage. Leaks may occur in handling; i.e., pumping, making and breaking usage connections, but no leakage results from an all-welded, well-inspected storage and distribution system.

The 4000-psi system is apparently the most economical because of pumping costs. Edwards uses the Haskell pumps. Annin packless valves are used throughout.

The pressure vessels have been isolated for weeks at a time without any apparent leakage.

I. Telephone Transcript, 5-22-58

To: Mr. S. H. Yee, National Aeronautic Association, Canoga Park Laboratory, Canoga Park, California

From: R. B. Clarke

Subject: Helium Storage System at Canoga Park Laboratory

The helium system for the Laboratory is not of the latest design. It is a system of gas bottles of at least two sizes, 54 and 200 ft³, manifolded together to the distribution system. This system utilizes black iron pipe, packed gate valves, and numerous screwed connections.

The storage manifold is about 1000 ft from the most remote joint of consumption. Main gas lines are a maximum of 2 in. dia pipe at 5000 psi. Total storage capacity is 500,000 scf.

The system is given an air-soap test only. A hydro test would lead to rust and corrosion problems. No vacuum test appeared practical.

Clark compressors are used to boost the pressure chiefly because of space requirement. A silica gel-drying system is used for moisture only. High purity is not so critical as at the rocket base.

G. Telephone Transcript, 5-29-58

To: Dr. S. Bogbonoff, Forestal Scientific Laboratory, Princeton,
New Jersey

From: M. Bender

Subject: Helium Storage

A small helium-supplied wind tunnel is in operation. No recovery system is included. Helium is obtained in tank trucks from Lakehurst Naval Air Station.

Helium-supplied wind tunnels are planned for Ames and Langley Field NACA Laboratories. Both installations have a recovery system which Dr. Bogbonoff thinks is a vacuum tank with a helium purification system included. The tunnels are under low pressure and the problem is mainly one of air contamination. Neither is yet in operation.

Dr. Bogbonoff suggested contacting Lt. Palmer at Lakehurst who should be able to furnish information about experience at that storage depot.

H. Telephone Transcript, 5-27-58

To: W. H. Lawrence, Edwards Rocket Base, Edwards, California

From: R. B. Clarke

Subject: Helium Storage

Helium is stored at Edwards Rocket Base at pressures of 4000, 6000, 10,000, and 12,000 psi in A. O. Smith layer vessels. The 4000-psi vessel is 47 ft long x 30 in. deep and has a volume of 300 ft³. The 10,000-psi vessel is 100-ft³ capacity.

The inner layer of all vessels is made from 304 or 347 stainless steel. The heads are formed in hemispherical shape. Each weld is x-rayed as fabricated in making the multilayer vessel.

The entire storage and distribution system at Edwards Rocket Base is of welded or ring-flange construction, using 304 or 347 stainless steel. All joints are made by heliarc welding using expert welders. Joints are

Total leakage	5%/month
Total usage	1,500,000 scf/month
Total loss	75,000 scf/month

J. Telephone Transcript, 5-22-58

To: Mr. Ken Premo, Robertshaw-Fulton, Anaheim, California
From: R. B. Clarke
Subject: Helium Storage

Usage of helium at Robertshaw-Fulton is for test purposes on valves, regulators, etc. Some losses are recovered by collecting in plastic bags. Leakage through threaded joints and pumps accounts for most of the leakage.

The operation at this facility is not comparable to a GCR, nor is the type of system fabrication.

K. Telephone Transcript, 6-5-58

To: Dr. K. B. McAfee, Bell Telephone Laboratories, Murray Hill, New York, Phone No.: CRestview 3600, Ext. 3574
From: M. Bender
Subject: Helium Containment

Dr. McAfee has been carrying out work on the diffusion of helium through various materials. He states that he has only incidental experience with helium diffusion through steel but thinks there are very good theoretical reasons why helium diffusion would not be expected to occur until metals are near their melting points. He has had excellent success with kovar seals made up from nickel alloys.

Electrical penetrations should be readily prepared. Bell has a number of glass seals (about 60) installed in transatlantic cables. These hold a pressure of 6000 psi and withstand 3000 volts. Current flows are small, however. He does not visualize any difficulties in designing such a connection for higher current flows in the order of several hundred amperes.

Ceramic electrical penetrations have been developed which can be bonded to metals. Dr. McAfee thinks the conical arrangement of Bridgeman would be effective. Ceramic seals should be satisfactory at temperatures up to 400-500°F.

Epoxy resins are excellent for a quick and dirty solution to the problem. A mixture of epoxy and silica with a long leakage path would be effective in holding 300-400 psia helium at temperatures up to 100°C. Polyethylene is adequate up to 35°C. Within the specified temperature limits a leakage rate of 100 scf/day could be held without difficulty. Dr. McAfee reported that a ceramic seal had been developed to hold 6000 psi of helium with a leakage of 1 cm³ in 30 years.

With regard to the helium recovery system recently announced by Bell Laboratories, a symposium is scheduled to be held on this subject at Bell Laboratories on July 1, 1958. Most information on the subject is being withheld until that time. Dr. McAfee did say that the separation process is comparable to that used in recovery of uranium isotopes by gaseous diffusion. The helium recovery is very effective, extracting 99.99% helium from natural gas. He suggested that it could be an effective means for purifying helium in nuclear reactors.

Invitations to the symposium on helium extraction may be obtained by contacting C. I. Baker, Western Electric Company, 195 Broadway, New York, New York.

I. Helium Shipping Information

Tube trailers for highway transportation weigh 34,000 lb and at the nominal 1750-psi loading contain 31,600 scf. (Liquid volume, 265 ft³.)

Tube rail cars have a gross weight of 232,000 lb and at their nominal 2600-psi rating contain approximately 270,000 scf. The pressure rating will be increased to 3600 psi beginning in September 1958. (Liquid volume, 1400 ft³.)

The Union Carbide Nuclear Company Traffic Department was requested to find the cost of shipping helium from Oak Ridge to New York to 1) Calais, France; 2) Naples, Italy; 3) Bombay, India; 4) Buenos Aires, Argentina, for a 100-ft³, 10,000-lb vessel containing 68,000 ft³ helium, and also the trailer.

<u>Destination</u>	<u>Tank</u>	<u>\$/ft³</u>	<u>Total \$/ft³</u>	<u>Trailer</u>	<u>\$/ft³</u>	<u>Total \$/ft³</u>
Oak Ridge- New York	1034.17	0.015		428.28	0.014	
Calais	1129.22	0.016	0.031	936.99	0.030	0.044
Naples	1584.58	0.023	0.038	4015.70	0.127	0.140
Bombay	1924.10	0.028	0.043	4393.44	0.139	0.143
*Buenos Aires	2436.86	0.036	0.051	5195.77	0.164	0.178

* A \$525.00-counselor fee may not be required if the material is consigned to the Argentine Government.

APPENDIX II

Use of 7503 Cell to Demonstrate Helium Leak-Tightness for GCR Application

A. Proposal

As a result of questions raised regarding the difficulty in retaining helium in a gas-cooled reactor, the proposal has been made to demonstrate the ability to retain helium in a welded system at 500°F and high pressures. In the 7503 Building at X-10 there is a 24-ft dia cylindrical tank designed for 200-psig pressure. This vessel, which has some plate as thick as 4 in. and is fabricated by techniques equivalent to those required for the GCR-2 vessel, can be assembled and a test applied for demonstration purposes.

Demonstration of the ability to provide leak-tightness will involve the following tests of the 7503 pressure vessel:

1. Pneumatic testing of the 7503 cell at 255 psia.
2. Pressure-testing with air at 200 psig using soap bubble technique at a number of pressure levels up to 200 psig. Probe with a halogen leak-detector with the pressure vessel having air and freon vapor at 200 psig.
3. Purge pressure vessel and fill with the desired helium atmosphere to 200 psig with vessel being maintained at temperature of 400 to 500°F. Observe continuous recording of pressure and temperature over a period of 15 days to determine leakage rate.
4. Sample gas for evidence of preferential helium diffusion losses.

B. Preparation Required

At the present time the head of the 7503 pressure vessel is stored across the road from the building. Also, all of the eight 24-in. dia junction panel nozzles, the twelve 8-in. dia, and six 6-in. dia coolant nozzles, the two 2-in. dia off-gas nozzles, the five 2-1/2-in. dia spectrometer nozzles, and the 5-ft dia manhole are open.

The following items of preparation are required for the tests listed above:

1. Transfer and weld head to cylindrical section, magnaflux and x-ray weld.

2. Bolt manhole cover to nozzle after hydro test seal weld closure.
3. Weld closure plates to all other nozzles listed above.
4. Free pressure vessel anchor bolts, transfer and tack-weld water tank head in place. Insulate interior of water tank. Install heating equipment, either oil burner or electric resistance heaters, in annular space between the pressure vessel and water tank. Provide closure plates for five 30-in. water tank junction panel openings. Attach thermocouples to pressure vessel as per ASME code recommendations and install continuous temperature point recorder. Install continuous pressure recorder. Install 315-psia rupture disc and relief valve.

The vessel has been stress-relieved, hydro-tested, vacuum-tested, sandblasted, and painted. The welded head and nozzle cover have been removed preparatory to installation of equipment. Stress relief will not be required after replacement of the head but pneumatic testing and leak testing will be necessary. Helium vacuum-testing would be desirable but the painted surface will cause a serious out-gassing problem. The halogen leak-detector test will probably be ample to locate significant ~~leaks in the system if carried out carefully.~~

Since there are no qualified welders available in the X-10 organization for ASME code welding of SA-201 Gr B in heavy sections, and since probably a month or two at best would elapse in qualifying X-10 welders for the work, it appears more expedient to release this work to Chicago Bridge and Iron Company, the original fabricator. Also, on a cost basis, this latter arrangement should be more economical because of the lack of experience of X-10 craftsmen ~~with~~ this type of work.

All of the construction outlined above, including the instrumentation, could be installed by Chicago Bridge and Iron Company. One or two technicians for each shift would be required for the duration of the hot pressure-testing of the vessel.

Construction expense is estimated to be \$10,000.

Expense of operation of hot pressure test is estimated to run from \$300 to \$500/day. This expense will cover the technicians and craft required and fuel oil requirements.

Helium requirements of the system are dependent upon the concentration in the tank whose volume is 12,000 ft³. If the test for

leakage is carried out at 200 psig (the design pressure), the volume required will be 95,000 scf for 100% helium at 500°F, costing about \$2,300.

APPENDIX III

(Letter from W. T. Furgerson to W. H. Jordan, May 15, 1958, "Report of Discussion of GCR Blowers and Drives with Allis Chalmers Mfg. Co.")

To: W. H. Jordan Date: May 15, 1958
Copies: A. P. Fraas Subject: Report of Discussion of GCR Blowers
R. V. Meghreblian and Drives with Allis-Chalmers Mfg. Co.

Present:

For Oak Ridge National Laboratory,
M. Bender
W. T. Furgerson
R. D. Stulting

For ACF Industries,
W. Banks
Earl F. Palmer
K. Trichelt

For Kaiser Engineers,
J. P. Ernst
J. T. Jackman, Jr.
Harold D. Young

For Allis-Chalmers,
C. F. Codrington
D. O. Millar
F. F. Szczesny
H. J. Welch

The subject meeting was the result of a suggestion made by the Allis-Chalmers representative in Knoxville. In view of certain differences between the blower and drive concepts set forth in the ORNL and Kaiser-ACF gas-cooled reactor studies this was a welcome opportunity to get the viewpoint of a third party which was experienced in the field.

Since it was known that the Allis-Chalmers personnel who were to be contacted had no first hand information on the gas-cooled reactor project and were not aware of what specific information was desired, a list of questions and topics for discussion was prepared and delivered to them in advance. An attempt was made to include topics of general interest to all parties even though our own answers to many of the questions had been developed during the course of the GCR-2 study. The items which were included are listed below together with answers and information developed.

(1) Is aerodynamic technology sufficiently established that blowers could be designed for any of the coolant gases contemplated without aerodynamic development? This would include hydrogen and gases of greater molecular weight up to carbon-dioxide.

The answer here was an unqualified yes.

(2) What development would you consider necessary if you were asked now to produce a blower for a gas cooled reactor?

It was the consensus of opinion that the principal development would be mechanical with aerodynamic development being limited to possible adjustment of flow rate to meet specifications. The latter, incidentally, would depend on the specified limits of tolerance on flow rate. It has been the writer's personal experience that the flow rate of a newly designed turbo blower or compressor can be expected to fall within $\pm 5\%$ of the design value for the first test. One adjustment can be expected to reduce this to less than $\pm 1\%$. A certain amount of model testing may be called for in order to determine the best shape for the inlet or discharge volutes. However, this can be done simultaneously with the design work.

(3) Review the design conditions best suited for centrifugal blowers and which are best suited for axial flow machines.

This topic was discussed at length. The results of the discussion might be summarized by saying that for a given design head there is a certain design flow rate above which axial machines are superior from an efficiency standpoint and below which centrifugals are superior. This is shown in Fig. I.

Fig. I

The crossover point for air may be approximately 50,000 cfm. This depends on several things, however, such as gas composition, head, and pressure level.

Note that the efficiencies of both machines level off at some maximum value as flow rate increases. The writer has a minor disagreement with the Allis-Chalmers engineers as to the difference to be expected between

the ultimate efficiencies of the two types. It is their feeling that the difference is approximately 5% while the writer believes it to be perhaps half as much provided both designs represent the same technological level. The writer certainly feels that the expected difference in efficiency is sufficiently small that it may be overshadowed by other considerations such as the mechanical design, adaptability to certain system layouts, or some specially desired shape of the characteristic curve.

In the latter regard it was brought out that in an experimental reactor where fuel elements having a range of pressure losses might be tested, the steeper characteristic which is generally typical of axial machines would have the advantage of permitting changes in operating head with relatively small changes in flow rate. It should be pointed out, however, that the blower designer has a high degree of control over the shape of the characteristic curve for either axials or centrifugals. If the reactor system designer requires a certain shape characteristic curve he should specify the required shape rather than specify the type of blower in hope of getting the desired shape.

(4) Would you consider performance tests adequate if run with air at room temperature and corrected to reactor coolant composition, pressure, and temperature by calculation?

The answer was yes provided incompressible flow existed at both test and reactor conditions.

(5) Review the effect of gas composition, suction volume, and head on blower design.

Blowers for low molecular weight gases will be smaller in diameter and run at higher rpm than those designed for high molecular weights at the same volumetric flow. For a given gas higher volumes call for larger diameters at lower rpm with tip velocity remaining constant. Higher heads require either higher tip velocities or more stages.

(6) How is the drive mechanism affected by blower requirements?

As developed in the previous item the blower rpm is related to the gas composition and flow rate. It is of interest that all of the blower design points under consideration, i.e., helium at 125,000 cfm, carbon-dioxide at 50,000 cfm or 15,000 cfm would require 3600 rpm or higher. The first two cases could be direct-driven by a two-pole induction motor; the last might require a gear speed increaser if 60 cycle a-c motors are used.

(7) Review experience in designing machinery to handle light gases, hydrogen or helium, particularly at high pressure and temperature. The interest here is in containment problems such as the effect of light gases on materials, etc. rather than aerodynamic problems.

Allis-Chalmers is building centrifugal hydrogen compressors for the petro-chemical industry, eight having been delivered in the past two years. These operate at pressures up to 1000 psi and temperatures up to approximately 400°F at the discharge. Some materials problems did arise, but these appear to have been satisfactorily solved. The gases handled by these machines contain some corrosive compounds, and it is difficult to separate the effects of these from those of the hydrogen. It was mentioned that hydrogen did cause blistering of porous castings or forgings having imperfections. This effect was controlled by careful inspection of materials by radiography, Zy-glo, etc. Generally, best results were obtained with steels such as 4140, 403, 404. Difficulty has been experienced with 18-8 stainless and monel, but the exact nature of these difficulties was not discussed.

It is of interest that these machines were tested by Allis-Chalmers using helium as the working fluid.

(8) Do you have shaft seal experience applicable to gas cooled reactor requirements?

The shaft seal used for hydrogen compressors should provide a basis for development of a seal for gas cooled reactor blowers. Allis-Chalmers is currently designing a seal of this type for a reactor project of Aerojet-General.

FIG. II

The seal is a sleeve type using a liquid sealant as shown in Fig. II. This sealant is usually a light oil, but will be water in the Aerojet-General unit. The amount of sealant vapor which can be allowed to back-diffuse from the inner sealant catch basin into the coolant system has a great effect on the complexity of the seal and auxilliary equipment. If stringent requirements are placed on this rate of back diffusion it will be necessary to use an inner seal of complex design (and probably shorter life) than the simple labyrinth shown in Fig. II; or, alternatively, it will be necessary to increase the outward flow of gas through the inner seal which means that either a high out-leakage of coolant must be tolerated or a large coolant recovery system must be installed. An outflow rate of 0.1% of the blower flow rate per seal was mentioned, but this figure is highly tentative since it depends on information which was not available at the time of the meeting.

Allis-Chalmers agreed to determine the daily leakage and yearly maintainance for the following conditions:

1. "No" sealant inleakage
2. Charge downtime at the rate of \$50,000 per day
3. Pressure and temperature 400 psi and 450°F, respectively
4. Helium and carbon-dioxide
5. Blower capacities 15,000 and 50,000 cfm.

It is apparent that the reactor system designers must specify an allowable sealant diffusion rate. The rate specified must be a bona-fide requirement. To arbitrarily say "no" inleakage when a few ppm may be allowable could be expensive.

Allis-Chalmers would be interested in undertaking a seal development program. It is believed that if the Laboratory does not plan to undertake such a project, itself, it would be well to open negotiations with Allis-Chalmers in this regard.

(9) What is your experience with "canned" or hermetically sealed motors?

"Canned" motors have been built in sizes smaller than gas cooled reactor requirements, but no difficulty is anticipated in going to larger sizes.

(10) Certain reactor system layouts could be simplified if vertical shaft motors could be used. Could you supply vertical shaft motors at 3600 rpm and in the range from 5,000 to 10,000 hp?

Such motors are currently not available but should be possible to develop in approximately 1 year.

(11) What is available in gas turbine equipment?

Allis-Chalmers is currently working on a maritime gas turbine power plant of 22,000 shp at a turbine inlet temperature of 1300°F. It seems, incidentally, that 20,000 to 25,000 shp is about the highest power currently available in gas turbine power plants, either in this country or in Europe.

The general impression was gained that coolant circulating blowers and drive motors for well designed gas cooled reactor systems will be available when needed.

It should be possible, according to Allis-Chalmers personnel to develop a suitable shaft seal within a year. This was a pleasant surprise which is due in part to previous work which has been done relative to hydrogen compressors.

The information on the hydrogen compressors is considered important. Hydrogen is the ultimate cooling medium for gas cooled reactors. If the material and psychological problems connected with its use can be solved reduction of power costs of approximately 10% of the figures quoted in the GCR-2 report can be effected. The fact that problems in at least the former category are being solved is encouraging.

W. T. Furgerson

W. T. Furgerson

WTF:bhf

APPENDIX IV
(ORNL-CF-58-4-112)

I. HELIUM RESOURCES OF THE UNITED STATES

Information available in recent Bureau of Mines Minerals Yearbooks show a helium production of 190 million cubic feet for 1954 and a price of \$14.65 per thousand cubic feet. Helium is being produced at four plants (1) Amarillo, (2) Exell, Texas (north of Amarillo), (3) Navajo, New Mexico (Shiprock) and (4) Otis, Kansas. The Amarillo plant is supplied by the Cliffside field, the Navajo plant by the Rattlesnake field. These fields are owned or directly controlled by the Government as are the Woodside Structure and Harley Dome properties located in Utah. The Exell and Otis plants utilize natural gases produced for fuel markets and whose helium content would otherwise be wasted. Reserves of 3.4 billion cubic feet of helium were present in the Government controlled fields in 1953 with some 7 billion cubic feet available for the Otis and Exell plants, most of this being in the Channing area of the Texas Panhandle. The helium concentrations present in gases from these fields are listed below.

<u>Field</u>	<u>Vol % of Helium</u>
Cliffside	1.8
Rattlesnake	7.5
Woodside	1.3
Harley Dome	7
Channing	0.9 (31 wells supplying Exell plant)
Otis	1.4 (87 wells in Kansas)

Natural gases from other fields in the same general area are produced for the fuel market and their relatively large helium content is being wasted. The Bureau attempts to operate the Otis and Exell plants at capacity with additional production necessary to meet demands coming from the Amarillo and Navajo plants. During one year excess production from the Exell plant was piped to and stored in the Cliffside field. Increased demands for helium have necessitated the operation of all four plants during the last several years and resulted in extensive use of gases from the Government preserves.

The Bureau of Mines expressed concern over the loss of helium from fields being utilized for fuel markets in 1953. The fields were in production at that time but the helium demand did not warrant the construction of plants to recover the helium and furthermore it appeared that the helium loss would be distressingly large before the demand developed.

Some positive steps have been taken during the past year to implement a helium conservation program. Mr. Gordon Gray,

Director, Office of Defense Mobilization, requested that the Hon. O. Hatfield Chilson, Under Secretary of the Interior, set up a group to determine how a national policy to conserve helium best be accomplished and the cost of such a program. The study was extended to include an estimate of the value of helium to defense and atomic energy programs. Representatives from the Department of Defense, Department of Commerce, Atomic Energy Commission, Bureau of Budget, Office of Defense Mobilization, and Department of Interior considered the conservation program and released on January 1, 1958 a report entitled, "The Cost and Implementation of a National Helium Conservation Policy designed to prevent the loss into the atmosphere of any appreciable quantity of helium which has not first served a useful purpose (including a discussion of the value of helium)"--Helium Policy Working Group, Hon. O. Hatfield Chilson, Chairman. Some of the information contained in this report is given on the following pages.

Current Production and Future Requirements

Actual production of helium was 281 million cubic feet in FY-1957. Anticipated production is 387 million cubic feet for FY-1958. All helium is produced currently at the four plants previously mentioned. The Bureau is requesting funds in the FY-1959 budget for construction of another plant to be located at Keyes, Oklahoma to meet requirements in the immediate future. It is anticipated that the annual helium requirements will continue to grow reaching 500 million cubic feet by or shortly after 1960 and expanding to at least 1.2 billion cubic feet by 1975.

Reserves

Ninety-nine percent of the known helium-bearing natural gas resources in the U.S. occurs in four fields: (1) the Hugoton field of Texas, Oklahoma and Kansas; (2) the Panhandle field of Texas; (3) the Keyes field of Oklahoma; and (4) the Greenwood field of Kansas. These fields comprise an area including the Texas Panhandle, extreme western Oklahoma and southwestern Kansas. These fields are in production for fuel with the helium being wasted. Some data on these fields are compiled below.

	<u>Keyes Cimarron County Oklahoma</u>	<u>Greenwood Morton County Kansas</u>	<u>Texas Panhandle Texas</u>	<u>Hugoton Kansas Oklahoma and Texas</u>	<u>Totals</u>
Gas produced in 1956 (million cubic feet)	10,730	35,430	632,450	739,260	1,417,870
Est. reserves as of 1-1-57 (billion cubic feet)	620	1,800	14,100	19,900	36,420
Helium content (average percent)	2.0	0.6	0.48	0.46	-
Est. recoverable helium (million cubic feet)	7,900	7,060	44,000	60,000	118,960

It is estimated that about 60 percent of the approximately 119 billion cubic feet of helium contained in these fields is recoverable by 13 plants located at strategic points on pipelines leading from the fields.

Conservation of Helium

The plan proposed by the Bureau of Mines for the Conservation of helium contained in the four fields entails the construction of 13 plants as listed below.

<u>Plant Number</u>	<u>Plant Location</u>	<u>Annual Helium Production (million cubic feet)</u>
1	Keyes, Oklahoma	290
2	Meade, Kansas	550
3	Sublette, Kansas	250
4	Sunray, Texas	120
5	Greenwood, Kansas	85
6	Fritch, Texas	195
7	Lakin, Kansas	100
8	Stinnet, Texas	180
9	Stinnet, Texas	160
10	Liberal, Kansas	130
11	Dumas, Texas	120
12	Straight, Oklahoma	125
13	Holcomb, Oklahoma	90
		<u>2,395</u>

It is estimated that these plants, plus the four now in operation, could recover 43.5 billion cubic feet of helium by 1975. Allowing for sales of 11.5 billion cubic feet during this period, 32 billion cubic feet of helium would be available for conservation in underground storage. A pipe-line system would be built to transport the helium to the Cliffside field for storage. This field has produced more than 32 billion cubic feet of gas and, therefore, should have sufficient capacity for the helium without exceeding the original field pressure. Purification of the helium after withdrawal from storage would cost less than half of the initial cost of production.

Recovery of more than about 60 percent of the helium from these fields would necessitate construction of additional plants, many of which would be small and marginal. Further study would be required before any attempt is made to recover additional helium.

Cost of Conservation

The cost of the 13 plants and connecting pipeline is estimated at 238 million dollars. Operation and maintenance costs would run to 382 million dollars making a total of 620 million dollars for the period 1960-1975. About 85% of the helium produced would be conserved or 525 million dollars should be charged to the program. If the cost of financing the program is included about 300 million dollars additional would be involved. Using present day costs and expenses and the best available estimates of future consumption it is estimated that the operation and maintenance costs of producing 32 billion feet of helium for storage could be recovered by charging \$44.50 per thousand cubic feet for helium sold during the 1960-1975 period. Continuation of this price through 1985 would provide sufficient revenue to retire the capital costs and deferred interest.

Private Participation

The Helium Policy Working Group believes that private industry should be invited to participate in the program although it recognizes that some serious problems would have to be resolved. It is proposed that the Government contract for helium extraction rights and these be made available to private companies interested in building and operating plants for helium extraction from the natural gases. Also, it is recommended that the Government purchase all helium so produced thus insuring control of production, distribution and use of helium. In case private industry is not interested in such a program the Government should proceed with it.

Such legislative changes that would be required through amendment of the Helium Act or otherwise should be sought through Congress.

Helium Utilization

The present utilization and future helium requirements are presented in Table 1. It is evident that the Federal Government is and will continue to be the consumer of a large fraction of the helium utilized. The Bureau of Mines in a study of private helium utilization presented data which show that as of December 1955 about 50 percent of the helium supplied to non-Federal users was being used for work done on Government contracts. If this percentage held through 1957, the Government used either directly or indirectly 85 percent of the helium consumed during this period.

Essentiality of Uses

Airships. No acceptable substitute for helium.

Guided Missiles. Helium is required for the presently designed missiles. Future designs might use some other gas such as nitrogen or air.

Welding. This accounted for about 65 percent of helium supplied non-Federal users in 1955. Except for certain AEC welding requirements it is believed that argon may be used in place of helium at some sacrifice in quality and output.

Aerology. Helium will be used as long as it is available.

Nuclear Reactors. Some substitution of argon for helium has been accomplished but it appears improbable that any significant replacement will occur as long as helium is available. The development of gas-cooled reactors using helium is anticipated.

Aeronautical Research. Helium is essential and its use expanding rapidly. Efforts will be made to purify and reuse the helium in wind tunnels whenever practical.

Low Temperature Research. There is no suitable substitute for helium available.

Leak Detection. There is no suitable substitute for helium in detecting extremely small leaks. Industry also prefers helium for less rigid leak-testing.

Titanium and Zirconium Production. Argon will be used in future production of these metals except where heat conductivity requires the use of helium.

Table 1

Present and Estimated Future Helium Requirements
(Millions of Cubic Feet - Fiscal Years)

	Actual 1957	Estimated		
		1958	1959	1960
Department of Defense	136.8	198.6	208.6	215.6
Airships		21.9	17.4	17.0
Guided missiles		75.0	75.0	75.0
Welding		6.8	7.0	6.0
Aerology		46.6	52.6	54.4
Nuclear reactors		0.1	0.1	0.1
Breathing atmospheres		0.5	0.5	0.5
Aeronautical research		30.7	33.0	37.0
General Stores		17.0	23.0	25.6
Atomic Energy Commission	51.8	75.5	90.1	94.0
Military application		17.1	19.3	19.8
Production		48.5	60.0	62.2
Reactor development		9.9	10.8	12.0
Weather Bureau	7.9	11.0	12.0	13.0
Other Federal	2.0	2.0	2.0	2.0
Total Federal	198.5	287.1	312.7	324.6
Non-Federal Users	83.0	100.0	120.0	140.0
Total	281.5	387.1	432.7	464.6

Breathing Atmospheres. There is no suitable substitute for helium available.

The Helium Policy Working Group is convinced that the uses of helium are highly essential to the welfare of the United States--particularly with respect to its military strength--and that the proposed conservation program is justified. If such action is not taken a tremendous loss of helium to the atmosphere will occur as the fuel gases from these fields are burned. The only alternate source of helium is the atmosphere which contains one part in 200,000. The estimated cost of recovery from this source is \$10-\$20 per cubic foot. Only about 2 million cubic feet per year could be recovered if all of the air now processed for oxygen recovery were also processed for the recovery of helium.

II. HELIUM RESOURCES OUTSIDE OF THE UNITED STATES

Helium resources of North America outside of the United States are of importance only in the Canadian Provinces. The helium content of gases from 12 New Brunswick wells (Stony Creek Field near Monoton) ranged from nil in 2 wells to 0.69 cu. ft./1000 cu. ft. (0.069%) with an average of 0.024%.¹ Production from this field during the period 1915-1925 ranged from 430 to 796 million cu. ft. Many Canadian natural gases contain 0.1% of helium or more with a range from 0.003 to 0.34%.² Another study³ found an average of 0.36% helium in selected natural gases. It was estimated in 1926 that over 5 million cu. ft. of helium was recoverable annually from Alberta and Ontario gases.⁴ Later⁵ an estimate of 4 million cu. ft. of helium from 117 wells was made at a cost of \$100 per thousand cu. ft. So far as is known no recovery of helium from Canadian gases has been carried out. It appears likely that the situation at present may be more favorable in view of the numerous fields brought in within the past 25 years.

Numerous small gas fields in Europe contain more than 0.01% of helium. The helium contents of a number of German fields have been reported.^{6,7,8} For instance, gases from the Rhine area were found to contain 0.088 to 1.09%, gases from a Hamburg field 0.016%. Deposits at Liege, Mons and Charlerai in Belgium are reported⁹ to contain 0.057, 0.050, and 0.33%, respectively. French sources range from 0.055% at Crespin to 0.12% at Anzin.⁹ Italian gases have been reported to contain 0.0005 to 0.12%.¹⁰ The Carpathian fields in Poland contain helium but generally less than 0.05% with the helium content increasing in an easterly direction.¹¹ The helium contents of 8 wells examined in Roumania varied from a trace to 0.002%.¹² One well of 15 examined in Hungary was found to contain 1.54%, the others ranged

from 0.0023 to 0.026%.¹³ In general the quantities of gases available are too small to justify the outlay required for helium recovery.

The Russians have a helium industry but no information on its capacity is available.^{14,15} Gas from the S.E. Caucasus contain 0.004% helium, from the Taman Peninsula 0.002% and from the Kerch Peninsula (Crimea) 0.005%.¹⁶ Natural gases from the western Urals contain about 0.05% helium.¹⁷ The Ishimbaev gases (southern Urals) are similar to the United States helium containing gases but the helium contents are slightly lower.¹⁷ No data related to the production or reserves of these fields are available.

Elsewhere in the world, samples from 13 Japanese wells showed a maximum helium content of 0.2% with most samples falling in the range 0.002 to 0.007%.¹⁸ Gas samples from Java and Sumatra contained up to 0.0033%.¹⁹ All but three of 81 samples from New Zealand contained helium, the contents ranging from 0.002 to 0.12%.²⁰ Only one reference concerning the Middle East fields was found. A gas sample from Hurghada, Egypt was found to contain 0.005%.²¹ It is not known if the large oil producing fields contain significant gas deposits or not. The same is true of the South American fields.

It appears that the United States possesses the important helium resources of the world. Canada may have gas fields from which commercial quantities of helium could be obtained but the cost would probably be considerably greater than for production in the United States. No idea of the Russian production is available nor of that country's potential. Elsewhere the gas deposits appear to be very limited and although small amounts of helium are obtainable from various fields the costs would undoubtedly be high.

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APPENDIX V

(ORNL CF-58-4-108)

COMPARISON OF GASES FOR USE AS THE COOLANT IN A GAS-COOLED REACTOR

There is no simple and accurate method of comparing the merits of different gases as a coolant in a gas-cooled reactor. One very common method is shown below. The basic equations for the comparison are as follows:

$$h = \frac{0.023}{Re^{.2}} \frac{V \rho C_p}{(Pr)^{2/3}} \quad (1)$$

$$Q = h A_s \Delta t_w \quad (2)$$

$$Q = w C_p \Delta t_g = V \rho A_c C_p \Delta t_g \quad (3)$$

$$\Delta H_f = 4 f \frac{V^2}{2g} \frac{L}{D_e} \quad (4)$$

$$W = w \Delta H_f = V \rho A_c \Delta H_f \quad (5)$$

The object then is to determine which gas requires the smallest amount of work to deliver a given amount of heat from the reactor core.

Combining equations (1) and (2)

$$Q = \frac{0.023}{Re^{.2}} \frac{V \rho C_p}{(Pr)^{2/3}} A_s \Delta t_w \quad (6)$$

Combining equations (4) and (5)

$$W = V \rho A_c 4 f \frac{V^2}{2g} \frac{L}{D_e} \quad (7)$$

Dividing (7) by (6)

$$\frac{W}{Q} = \frac{V \rho A_c 4 f \frac{V^2}{2g} \frac{L}{D_e} (Pr)^{2/3}}{\left(\frac{0.023}{Re^{.2}} \right) V \rho C_p \frac{V^2}{2g} \frac{L}{D_e} A_s \Delta t_w} \quad (8)$$

Substituting (3) in (8)

$$\frac{W}{Q} = \frac{4 f Q^2 L (Pr)^{2/3}}{\left(\frac{0.023}{Re^{.2}} \right) 2g D_e A_s \Delta t_w \Delta T_g^2 A_c \rho^2 C_p^3} \quad (9)$$

Also $\rho = \frac{PM}{RT}$ and $f = \frac{.046}{Re^{.2}}$ therefore

$$\frac{W}{Q} = \frac{4 Q^2 L R^2 T^2 (Pr)^{2/3}}{g A_s A_c \Delta t_w \Delta T_g^2 D_e P^2 M^2 C_p^3} \quad (10)$$

or

$$\frac{W}{Q} = \frac{4 Q^2 L R^2 T^2}{g A_s A_c \Delta t_w \Delta t_g^2 D_e P^2} \frac{M (Pr)^{2/3}}{(M C_p)^3} \quad (11)$$

Although the last term in equation (11) is the only term involving the properties of the gas, it does not follow that the gas which has the smallest value of

$$\frac{M (Pr)^{2/3}}{(M C_p)^3}^*$$

will have the smallest required pumping power. In a reactor the variation of heat flux along a fuel channel is independent of the gas, and the temperature profile of the gas is a function only of the gas temperature in and out of the channel. In calculating the performance of a reactor the heat transfer coefficient may be determined by using the properties of the gas at the average temperature and then treated as a constant along the length of the channel. This approximation is usually good to within 5% of the true integrated value of the coefficient and is probably within the accuracy of the equations and data used to determine the coefficient. With the gas temperatures as a function only of its location in the reactor and the heat transfer coefficient a constant, the temperature of the fuel element and its cladding is a function only of its location in the reactor. For all gas-cooled reactors proposed or built to date either the temperature of the fuel element or the clad has limited the reactor output. If it is assumed that the maximum allowable fuel element or clad temperature is independent of the gas, and with the temperature of both the fuel element and the gas independent of the gas, then it follows that the heat transfer coefficient must be the same regardless of the gas used as a coolant. The heat transfer coefficient for gases of different properties will not be the same unless the geometry of the system is altered when a different gas is used. The fallacy then of using only the last term of equation (11) is that the terms involving the geometry of the coolant channel cannot be treated as constants.

If the inlet and exit gas temperatures and the maximum fuel element temperature are fixed, then equations (1) and (3) give a relation between the gas properties and the geometry of the system.

* The expression

$$\frac{M (Pr)^{2/3}}{(M C_p)^3} \text{ is usually simplified to } \frac{M}{(M C_p)^3} \text{ with the variation of}$$

Pr assumed negligible. This expression has appeared repeatedly in the literature (1,2,3) concerned with gas-cooled reactors. In one case (1) the expression is presented as an absolute criteria of the heat transfer merits of a gas while another (3) states "the differences are not so marked as expressions like $M^2 C_p^3$ suggest".

$$h = \frac{0.023}{Re^{.2}} \frac{V \rho C_p}{(Pr)^{2/3}} \quad (1)$$

$$Q = V \rho A_c C_p \Delta t_g \quad (3)$$

$$\text{also } Re = \frac{D V}{\mu} \quad (12)$$

Combining (1), (3) and (12)

$$\frac{h \Delta t_g^{.8}}{0.023} = \frac{C_p^{.2} Q^{.8} \mu^2}{D_e^{.2} A_c^{.8} (Pr)^{2/3}} = \text{constant} \quad (13)$$

$$\text{also } D_e = \frac{4 A_c}{W_p} \quad (14)$$

From equations (13) and (14) it becomes obvious that the relative merits of different coolant gases cannot be determined until the configuration of the fuel elements and the channel have been fixed. If the channel is assumed to be circular and the fuel elements made up of one or more circular rods then

$$A_c = \frac{\pi}{4} [D_c^2 - N(D_R)^2] \quad (15)$$

$$\text{and } W_p = \pi [D_c + N(D_R)]$$

$$\text{and } D_e = \frac{4 A_c}{W_p} = \frac{D_c^2 - N(D_R)^2}{D_c + N(D_R)} \quad (16)$$

From equations (13), (15) and (16) it can be seen that several dimensions must be fixed. If the number of rods and their diameter are fixed then the channel diameter can be found for any heat output per channel. If the gas properties are changed then a new channel diameter must be found to satisfy the equations. Once the channel diameter and therefore the free flow area are known it is then a simple matter to find the velocity and the work required to overcome the friction losses. It should be noted that equation (4) and therefore equation (11) are for the core friction loss only. There are also expansion and contraction losses in the core. These will depend only on the gas velocity and may be of the same order as the friction loss. There are also losses in the system external to the core, but these are usually relatively small.

Given below is a comparison of the pumping power required for a system using both CO₂ and He. The reactor chosen for the comparison is a design⁽⁴⁾ by Kaiser Engineers and Nuclear Products-ERCO in which CO₂ is the proposed coolant. The following design data were taken from Tables 3.1.1 and 3.1.4 of reference 4.

Type of Fuel	7 slug UO_2 cluster
Fuel Slug Diameter	0.75 in.
I.D. of Graphite Sleeve	3.0 in.
Core Height	25.75 ft
Reactor Thermal Output	600 Mw
Pumping Power	28 Mw (Electrical)
Inlet Gas Temperature	473°F
Mean Exit Gas Temperature	1000°F
Reactor Coolant Flow	14×10^6 lb/hr
Reactor Inlet Pressure	400 psia
Maximum Cladding Temperature	1300°F
Coolant Mass Velocity in Center Channel	7.4×10^5 lb/hr ft

In comparing the performance of the two gases the properties and also the heat transfer coefficient are determined at the average temperature and are treated as being constant. It is also assumed that the mixed mean exit temperature of the gas from the reactor is identical to the temperature leaving the center reactor channel. This can be done by orificing the the channels so that the flow through any channel is proportional to the heat generated in that channel. Also the maximum fuel element temperature in any channel will be less than that of the center channel. This then allows one to calculate the overall performance of a reactor on the basis of the center channel only.

The gas properties were taken at 740°F and are as follows:

	CO_2	He
Specific heat	0.27	1.248 Btu/lb °F
Viscosity	1.95×10^{-5}	2.28×10^{-5} lb/sec ft
Prandtl No.	0.72	0.734
Density	1.373	0.1247 lb/ft ³

The heat transfer coefficient is calculated from equation (1) and for CO_2 with a mass velocity of 7.4×10^5 lb/hr ft²

$$h = \frac{0.023 \times 7.45 \times 10^5 \times 0.27}{\left(\frac{D_e \times 7.45 \times 10^5}{0.0702} \right)^{0.2} 0.72^{2/3} 3600}$$

For a channel diameter of 3.0 in. and seven 0.75 in. rods

$$\begin{aligned} D_e &= 0.0512 \text{ ft} \\ A_c &= 0.0276 \text{ ft}^2 \\ h &= 0.114 \text{ Btu/sec ft}^2 \text{ } ^\circ\text{F} \\ V &= 149 \text{ ft/sec} \\ Re &= 5.4 \times 10^5 \end{aligned}$$

For a Re of 5.4×10^5 and for commercial tubing the fanning friction factor

$$\begin{aligned} f &= 0.004 \\ \Delta H_f &= 4 f \frac{V^2}{2g} \frac{L}{D_e} \\ &= 2780 \text{ ft lb/lb.} \end{aligned}$$

The expansion and contraction losses in the core are assumed to be as follows:

$$\begin{aligned} \text{inlet loss to core} &= \frac{1}{2} \frac{V^2}{2g} \quad (V \text{ at inlet temperature}) \\ \text{fuel element expansion} & \\ \text{and contraction} & \\ \text{loss} &= 1 \frac{V^2}{2g} \quad (V \text{ at average temperature}) \\ \text{exit loss from core} &= 1 \frac{V^2}{2g} \quad (V \text{ at exit temperature}) \end{aligned}$$

or

$$\begin{aligned} \Delta H (\text{expansion and contraction}) &= \frac{V_{avg}^2}{2g} \left[.5 \left(\frac{473}{736} \right) + 1 + 1 \left(\frac{1000}{736} \right)^2 \right] \\ &= 0.0475 V_{avg}^2 \\ &= 1050 \text{ ft lb/lb.} \end{aligned}$$

In the ORNL gas-cooled reactor study⁽⁵⁾ the head loss in the external circuit could be represented by the equation:

$$\Delta H_{\text{external}} = 2.78 \times 10^{-12} \left(\frac{W_T}{\rho} \right)^2$$

Assuming that the external circuit in the Kaiser Engineers-Nuclear Products study is similar to that of the ORNL study the external head loss is found to be

$$\Delta H_{\text{external}} = 2.78 \times 10^{-12} \left(\frac{14 \times 10^6}{1.373} \right)^2 = 290 \text{ ft lb/lb}$$

and the total head loss is

$$\Delta H_T = 2780 + 1050 + 290 = 4120 \text{ ft lb/lb.}$$

When He is substituted for CO_2 it is assumed that the only change to be made to the system is an enlargement of the diameter of the coolant channel. As stated previously the coefficient must be the same as for CO_2 .

$$h = 0.114 = \frac{0.23}{Re^{.2}} \frac{\rho V C_p}{(Pr)^{2/3}} \text{ Btu/sec ft}^2 \text{ } ^\circ F$$

and

$$0.114 = \frac{0.023 \times 0.1247 \times V \times 1.248}{\left(\frac{D_e V \times 0.1247}{2.28 \times 10^{-5}} \right)^{0.2} (0.734)^{2/3}}$$

Also the temperature rise through the reactor must be the same as for CO_2 . For this to be true

$$(V \rho A_c C_p)_{CO_2} = (V \rho A_c C_p)_{He}$$

Both of the above equations are satisfied for a channel diameter of 3.3 in. with

$$\begin{aligned} D_e &= 0.0716 \text{ ft} \\ A_c &= 0.0386 \text{ ft}^2 \\ V &= 254 \text{ ft/sec} \\ Re &= 9.95 \times 10^4 \end{aligned}$$

The friction factor is then 0.0055 and

$$\Delta H_f = 4 \times 0.0055 \frac{(254)^2}{64.4} \times \frac{25.75}{0.0716} = 7920 \text{ ft lb/lb}$$

$$\begin{aligned} \Delta H(\text{expansion and contraction}) &= 0.0475 (254)^2 \\ &= 3060 \text{ ft lb/lb} \end{aligned}$$

$$\Delta H_{\text{external}} = 2.78 \times 10^{-12} \left(\frac{3.03 \times 10^6}{0.1247} \right)^2 = 1640 \text{ ft lb/lb}$$

$$\Delta H_T = 7920 + 3060 + 1640 = 12,620 \text{ ft lb/lb}$$

The ratio of the pumping power of CO_2 to that of He is

$$\frac{\text{Power to } CO_2}{\text{Power to He}} = \frac{14 \times 10^6 \times 4120}{3.03 \times 10^6 \times 12,620} = 1.51$$

It is interesting to note that if the last term in equation (11) were used to evaluate the gases the ratio would be

$$\frac{\text{Power to CO}_2}{\text{Power to He}} = \frac{\left[\frac{M(\text{Pr})^{2/3}}{(M C_p)^3} \right]_{\text{CO}_2}}{\left[\frac{M(\text{Pr})^{2/3}}{(M C_p)^3} \right]_{\text{He}}}$$

$$= \frac{\frac{44(0.72)^{2/3}}{(44 \times 0.27)^3}}{\frac{4(0.734)^{2/3}}{(4 \times 1.248)^3}} = 0.81$$

The above example shows that the expression $\frac{M(\text{Pr})^{2/3}}{(M C_p)^3}$ can be very misleading in evaluating the merits of different gases. In the above example the difference amounts to a factor of approximately two. From equation (11) it can be seen that the required pumping power is proportional to the cube of the reactor output for a fixed reactor design and coolant. Thus, an increase by a factor of two in pumping power would increase the output of the system by about 26%. The actual reduction in pumping power of He compared to CO₂ is 1.51. By increasing the power input to the He by a factor of 1.51 the output of the system would be increased by about 15%.

NOMENCLATURE

A_c	Free flow cross-sectional area	ft^2
A_s	Heat transfer surface area	ft^2
C_p	Specific heat	Btu/lb $^{\circ}F$
D_e	Equivalent diameter	ft
D_c	Channel diameter	ft
D_R	Fuel rod diameter	ft
f	Friction factor	
g	Conversion factor	$ft\ lb/sec^2\ lb$
h	Heat transfer coefficient	Btu/sec $ft^2\ ^{\circ}F$
ΔH	Heat loss	$ft\ lb/lb$
L	Length	ft
M	Molecular weight	$lb/lb\ mol$
N	Number of fuel elements in cluster	
P	Pressure	$lb/ft^2\ abs.$
Pr	Prandtl Number	
Q	Heat transferred	Btu/sec
R	Universal gas constant	$ft\ lb/^{\circ}R\ lb\ mol$
Re	Reynolds Number	
T	Average temperature	$^{\circ}R\ abs$
V	Velocity	ft/sec
Δt_g	Gas temperature rise	$^{\circ}F$
Δt_w	Temperature difference from wall to gas	$^{\circ}F$
w	Weight flow	lb/sec
W_p	Wetted perimeter	ft

NOMENCLATURE (Continued)

W	Flow work	ft lb/sec
W_T	Total system flow	lb/hr
μ	Viscosity	lb/ft hr
ρ	Density	lb/ft ³

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