[redacted]

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES
3 4456 0361291 8

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION

3

ORNL-2588, Pt. II
Second Issue, Se
Special

Classification Cancelled
Or Changed To
By Authority Of OEC 9/13/71
By Goldberg Date 11/12/71

TRIP REPORT – VISIT OF US GAS-COOLED REACTOR TEAM TO UK – JULY 7-18, 1958

PART II

MATERIALS DEVELOPMENT, INCLUDING URANIUM DIOXIDE, BERYLLIUM, AND GRAPHITE

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON
If you wish someone else to see this document, send in name with document and the library will arrange a loan.

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

CLASSIFICATION CANCELLED
Chris Pac ORNL, CO 9/23/14
ADD Signature Date
Single review of CCRP-declassified documents was authorized by DOE Office of Declassification memo of August 22, 1994.

[redacted]

RESTRICTED DATA
This document contains Restricted Data as defined in the Atomic Energy Act of 1954. Its transmittal or the disclosure of its contents in any manner to an unauthorized person is prohibited.

[redacted]

ORNL-2588
Special

Second Issue

Copy _____

Distribution Limited to USAEC Offices and Direct Contractors Engaged on the Relevant Project Only

CONTRACT NO. [illegible]26

TRIP REPORT – VISIT OF US GAS-COOLED REACTOR TEAM TO UK-
July 7–18, 1958 – Part II
(Part I prepared and issued in Washington with no identifying number.)

MATERIALS DEVELOPMENT, INCLUDING URANIUM DIOXIDE, BERYLLIUM, AND GRAPHITE

JAN 22 1959

[redacted]

[redacted]

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES

3 4456 0361291 8

[redacted]

Distribution Limited to USAEC
Offices and Direct Contractors
Engaged in the Relevant
Projects Only

Distribution
Second Issue

1-50. Laboratory Records

[redacted]

[redacted]

[redacted]

CONTENTS

Abstract..... 1
Uranium Dioxide..... 4
Uranium.....18
Beryllium.....20
Liquid Metals.....26
Slurries.....30
Graphite.....32

Appendix A
Appendix B
Appendix C

[redacted]

[redacted]

ABSTRACT

Uranium Dioxide - The UO_2 for the British program is prepared from ADU precipitated material. In order to obtain reproducible pellet production, a polyvinyl resin is added and a high oxygen-uranium ratio oxide is used to promote densification during sintering. The entire fabrication procedure for the AGR pellets is presented. Information on the plastic properties of UO_2 and the effect of oxygen-uranium ratio and environment on sinterability is discussed. It has been found that the high oxygen-uranium ratio oxide sinters more easily, and a nitrogen or argon atmosphere is preferred since reduction of the oxygen-uranium ratio occurs in the hydrogen sintering atmosphere. Experiments have been performed by the British on producing bodies by warm-pressing, hot extrusion, and swaging.

Uranium - The present status of the growth and swelling of uranium under pile radiation is presented and effects of various variables, such as temperature, burn-up, thermal cycling, and alloying additions are described.

Beryllium - Beryllium is the preferred tubing material for UO_2-canned fuel elements for future gas-cooled reactors. The method that the British have developed for producing beryllium tubing is presented and the approximate cost of the fabricated metal is now quoted at $100/lb with possible reductions to $30 – $60/lb. Studies on the n, α reaction in beryllium and subsequent migration of helium in the lattice has been studied by two methods: (1) ejecting high-energy alpha particles in coarse-grained, as-cast, and fine-grained wrought beryllium; and (2) observations on the behavior of MTR shim rod beryllium upon heating.

Liquid Metal and Slurries - The current status of mass transfer studies in lead, bismuth, and lead-bismuth solutions are described. Some discussion on the Harwell studies of ThO_2 and UO_2 slurries in lead, bismuth, and Pb-Bi eutectic is presented.

Graphite - There are two major problems associated with the use of graphite as a moderator for gas-cooled power reactors. These are radiation damage of the graphite at low temperatures and, in the case of CO_2-cooled reactors, the reaction of the coolant with the moderator at elevated temperatures. The AEA has recently set a minimum graphite temperature of 260°C as the point above which no significant damage will occur at the expected lifetime of the new power reactors. This temperature is well above the inlet gas temperature of the four plants now under construction by industrial concerns in the UK. Means of increasing graphite temperatures in these initial plants are being investigated, and the design work on the next generation of power stations makes allowances for higher inlet gas temperatures. Several of the consortia plan to use channel liners (of graphite or

[redacted]

[redacted]

Magnox) in the lower portion of the vertical channels to impede heat flow from the graphite to the coolant, and hence increase the graphite temperature.

The problem of the CO_2-graphite reaction is under investigation by the AEA in connection with the Advanced Gas Reactor (AGR). The practical temperature limit for operation with CO_2 in contact with bare graphite is believed to be in the range 500 – 525°C. Treatment or coating of the graphite to increase the operating temperature has not met with much success. A phosphate treatment which provided good protection in out-of-pile tests did not prove effective under irradiation. It appears that no decision has yet been made on coating of the AGR graphite.

[redacted]

[redacted]

[redacted]

HARWELL DISCUSSIONS, JULY 14

The agenda for the discussions at Harwell was as follows:

A. UO_2 Fabrication

1. Chemistry of UO_2 production - various processes for producing ceramic grades of UO_2.
2. Characteristics of UO_2 from various processes - i.e., particle size, stoichiometric ratio, etc.
3. Fabrication of UO_2 fuel pellets -
   a. Grinding and granulation procedures.
   b. Compacting and presintering.
   c. Sintering conditions - time, temperature, atmosphere.
   d. Dimensional and density tolerances.

B. UO_2 Technology

1. The influence of stoichiometric ratio on fission-gas retention and radiation damage.
2. The effect of surface area, particle size, particle shape, stoichiometric ratio, and other physical properties on the sinterability of UO_2.
3. The effect of radiation damage on such properties of UO_2 as the thermal conductivity, vapor pressure, and melting point.
4. The effect of high atomic burn-up > 10,000 Mwd/T on fission-gas retention.
5. Mechanism of diffusion and escape of fission gases from internal voids. Relative rates of fission-gas diffusion through the crystal lattice or by way of grain boundaries.

C. Other Fuel Materials

1. Preparation and fabrication of uranium carbides and properties of UC-base fuels.
2. Fabrication techniques for preparing UO_2-graphite and UC-graphite fuel compositions and physical and mechanical properties of such experimental fuels.

D. Beryllium

1. Techniques developed for fabrication of beryllium, including preparation of initial billet material, temperatures, and schedule for reduction to sheet or plate.

[redacted]

[redacted]

2. Status of techniques for fabrication of tubing, such as roll-forming and extrusion.
3. Typical mechanical properties of beryllium sheet fabricated by various processes. Mechanical properties of interest include creep-rate and stress-rupture data, ductility/elongation/hardness, tensile and yield strength, and modulus of elasticity.
4. Effect of oxide content and particle size on the mechanical properties of hot-pressed beryllium.
5. The effect of strain rate on the tensile strength and ductility of beryllium sheet.
6. Alloying additions which show promise of impacting significant increases in strength and/or ductility of sheet.
7. Joining of sheet by brazing, including braze composition, flux, temperatures, etc.
8. Details of developments on diffusion bonding of beryllium sheet and plugged tubes.
9. Techniques for joining of end plugs in capsules by diffusion bonding, including surface preparation, temperature, and pressure.
10. Equipment and techniques for fusion welding of end plugs and sheet.
11. Limiting conditions of temperature and integrated neutron flux for beryllium exposed to reactor operation. Extent of swelling produced and effect of swelling on mechanical properties of beryllium.
12. Annealing of damage to beryllium reflector in MTR.

E. Graphite

1. Techniques for production of low-permeability graphite tubes and capsules, including materials used, sizes produced, and permeability of typical product.
2. Composition and method of applying non-permeable coatings on graphite suitable for protection under oxidizing conditions.
3. Techniques for producing fueled graphite capsules, including thermal treatments and method of sealing end caps.
4. Mechanism and rate of reaction of CO_2 with reactor-grade graphite at temperatures of 700 to 1500°F, and the effect of reaction on mechanical properties of graphite.

[redacted]

[redacted]

Unfortunately, we did not have sufficient time to touch on all the topics in the detail they deserved.

The following people took part in the Be and UO_2 discussions:

UK Personnel	U. S. Team Members
M. Finniston	J. J. Cadwell
L. Grainger	J. B. Philipson
J. Williams	E. E. Hayes
S. Pugh	T. E. Stephens
R. Barnes	W. D. Manly
H. Lloyd	J. M. Atwood*
J. Lewis	U. M. Staebler*
and members of the	E. L. Brady*
Chemistry Division	J. K. Davidson*

*Part of the time

The graphite discussions at Harwell were attended by the following:

UK Personnel	U. S. Team Members
J. F. Jackson	W. J. Larkin
P. W. Mummery	E. L. Brady
M. T. Kavanagh	A. M. Perry
J. H. W. Simmons	R. E. Nightingale
T. M. Fry	
J. Wright	

The organization of the Harwell Metallurgy Division is presented in Appendix A.

Uranium Dioxide

Our first discussions were concerned with the type of uranium oxide used by the British in their experiments. The standard Springfields product, the ADU precipitated material, is used. It is first converted to UO_3 and then reduced to UO_2 with hydrogen.

The British are also interested in the preparation of oxide by the electrolysis of a crude concentrate mixture of sodium-potassium-uranium fluorides. This type of uranium oxide, which has an oxygen/uranium ratio of about 2.08, is not suitable for cold-pressing and sintering but is excellent for swaged-type fuel elements or for use in a process requiring a high-tapped density. In some capsule-filling experiments, a packing density of 9.0 g/cc was approached. The British are satisfied with warm-pressing this type of oxide, and it can be fabricated to a density of 10 g/cc by a hot-swaging technique. Reference was made to a report[1] which describes the process of making this type of UO_2. The small pilot plant at Harwell can process batches of approximately 2 kg.

[1] The Chemical Extraction of UO_2, M/[redacted]

[redacted]

[redacted]

The question was asked regarding the type of oxide which the British prefer for fission-gas retention. Les Grainger replied that they prefer to have pores distributed throughout the body rather than at the perimeter, or to have the large hole in the inside as in the ORNL GCR-2 design. They would like to have these voids the right distance apart to collect the fission gases and to have their size small enough so that the fission gases cannot exert pressure. Jokingly, Jack Williams of the Harwell Research Staff said, "Mr. Grainger would like a dispersion of holes in a single crystal of uranium oxide."

The type of particle size of uranium oxide that would be necessary to make a good body was discussed. It was agreed that the smaller the particle size the higher the sintered density would become. One of the reasons for this is that the finer the grain size the higher the oxygen ratio in the uranium oxide body. This would increase the sinterability of the oxide. Jack Williams described how they had been doing some surface-area measurements and found that oxide with a surface area of 2 m^2/g will sinter well. He stated that, for the most part, a high "green" density is obtained from the coarser particle size. In discussing the correlation between the O/U ratio and particle size, it was pointed out that a particle size that gives 2 m^2/g is a 2.04 O/U ratio, whereas an 8 m^2/g surface area is pyrophoric.

Jack Williams discussed their experiments with UO_2 in which it was found that nucleation of U_3O_8 on the surface of the UO_2 was dependent upon the temperature and the O/U ratio in the starting material. It was learned that with small particle sizes of uranium oxide, the UO_2 would first oxidize to U_4O_9 and form a solid solution of the U_4O_9 and UO_2. Finally, the body would convert to U_3O_8 or a solid solution of U_4O_9 and U_3O_8 in the body. This was also shown to be dependent solely upon the oxygen concentration of the surrounding environment and the surface area of the body.

The British feel that the O/U ratio should be 2.00 and not exceed 2.043 for good fission-product retention and over-all behavior in the pile. However, Grainger feels that the method of preparation of UO_2 for the Advanced Gas Reactor, i.e., by reducing a high-oxygen-ratio material with hydrogen to get the proper O/U ratio, is somewhat different from producing a body from material with a certain O/U ratio and maintaining this ratio throughout the life of the body.

[redacted]

[redacted]

Mr. Grainger's discussion was the result of their studies on the use of an increased oxygen ratio in the UO_2 to give better sinterability. However, under irradiation with the high-oxygen level a two-phase relationship is obtained in which the gases can come in and out of solution going through this phase boundary. The breaking up and healing of the particle due to this transfer of oxygen from one part of the body to another causes the fission gases to be released, similar to a zone-refining step.

Harry Lloyd described the various fabrication methods that have been tried which led to the accepted fabrication method for producing the AGR bodies. He mentioned that in making oxide bodies, studies have been made of cold-pressing and sintering, warm-compacting, hot- and cold-swaging, and isostatic pressing. They feel that the most economical method is the pelletizing or cold-pressing followed by sintering.

Warm-compacting of uranium oxide bodies has been studied and approximately one tone of such material for BEPO has been produced. In warm-compacting UO_2 with an O/U ratio of 2.04, a multiple-acting die with a 10-tsi pressure at 800°C is used. One of the things which required extreme care was the coefficient of expansion differences between the body and the die. Finally, a stellite punch and chromium carbide die body were used with some success. The bodies had densities of approximately 10 g/cc^2, but they were extremely brittle and had high residual stresses which caused them to fracture and break up on standing. It would be quite expensive to make a large quantity of material by this method; however, it does give them very close dimensional control. Uranium oxide bodies have been produced that are 1-3/4 in. ID by 2-1/4 in. OD and 1-1/2 in. high. Harwell is preparing a report on the process.

Harry Lloyd spoke about their cold- and hot-swaging operations in which UO_2 had been compacted in aluminum, zirconium, and stainless steel cans. A density of approximately 8.8 g/cc with a 60% reduction was obtained by cold-swaging in aluminum cans. Warm-swaging or hot-swaging at 800°C in stainless steel cans produced UO_2 with a density of approximately 10 g/cc.

They spoke about hydrostatic pressing in which a 6-in.-long body 3 in. in diameter was compacted at a pressure of 10 – 30 tsi. After sintering, a high-density body was obtained. In future work they will make shorter bodies to get parallel surfaces. The possibility of extruding U_3O_8 and reducing it to UO_2 has been considered. [redacted]

[redacted]

[redacted]

Their cold-pressing and sintering operation utilizes uranium oxide from the ADU process, granulated by a wet granulation procedure to enhance flowability. The granulated powder is compacted in a double-acting press at pressures of 10 – 30 tsi and sintered for a period of 1/2 to 2 hr at 1450°C in argon and densities of 10.2 to 10.4 g/cc are produced. The surface of the bodies is quite good but it is necessary to do some grinding to get the desired dimensions. When ground, the pellets are found to have laminations.

After studying all of these methods, the production of the AGR pellets (0.3-in. dia x 0.4 in. long) will be by cold-pressing and sintering with the aid of a polyvinyl resin with the trade name "Cranko." The preparation of the AGR UO_2 bodies is as follows:

1. The uranium oxide is prepared from the ADU precipitate.
2. The Cranko binder is mixed with the oxide.
3. The material is granulated to the proper particle size.
4. It is pressed at 30 tsi in a double-action press, and when removed from the press, has extremely high green strength.
5. The Cranko binder is burned off at a temperature of 200 – 230°C in air, and during this treatment, the O/U ratio increases to 2.3.
6. The body is then sintered at 1450°C for one hour in argon.

If the body is then cooled to room temperature in nitrogen or argon, the O/U ratio will be 2.1. Since a lower oxygen ratio is desired in the material, it is cooled to 1200°C in argon or nitrogen, and hydrogen is then admitted for one-half hour reducing the oxide to a final O/U ratio of 2.03. If necessary, this can be lowered quite closely to O/U ratio of 2.00.

During the sintering operation, the body undergoes approximately 15% linear shrinkage; the total volume per cent shrinkage is roughly three times this. Oxide bodies produced by this process have a density of 10.4 – 10.8 g/cc. The British feel that this is the cheapest over-all uranium oxide body, since at the temperature of 1450°C it can be sintered in a molybdenum-wound furnace. However, they are interested in other sintering techniques such as using induction sintering with a graphite receiver and protecting the body with tantalum, tungsten, or some other highly refractory metal, since they have found that considerable contamination results from the carbon. They found that at 2000°C the carbon will reduce the uranium oxide to metal as well as form carbides.

[redacted]

[redacted]

Jack Williams then discussed the effect of various gaseous environments on sintering. This is a follow-up on the work that Peter Murray did several years ago. In Murray's old data, it was found that with an O/U ratio of 2.16 a better density could be obtained in the finished product by sintering in argon than by sintering in hydrogen. Williams took uranium oxide that had been carefully prepared in a dry box and oxidized it to variable levels. He found that there was quite a difference in the sinterability and densification of the body with small changes in the O/U ratio, since an oxide with a ratio of just 2.00 would sinter to a density of 9 g/cc; whereas, material with a ratio of 2.02 would sinter to a density of 10 g/cc. He varied the O/U ratio from the 2.02 material up to 2.18 using powder with a particle size which would yield a surface area of 2 m^2/g. This information is seen in Fig. 1.

Williams has also studied the effect of atmosphere on the sinterability of UO_2 by using argon, CO_2, vacuum, nitrogen, and hydrogen as the sintering environment and by varying the sintering time and temperature so that a body with a density of 10 g/cc would consistently be produced. The bodies were sintered after compacting at 10 tsi. The body would sinter to a density of 10 g/cc in one-half hour at 1100°C when the cover gas was nitrogen or argon, whereas in hydrogen it was necessary to sinter at 1600°C or higher to obtain this density, as seen in Fig. 2. This is due to the reduction of the O/U ratio by the hydrogen, since the higher the O/U ratio the easier it is to sinter, as seen in Fig. 3.

Figure 4 indicates the sinterability of $UO_{2.18}$ material in argon and nitrogen as a function of sintering temperature for a sintering time of two hours.

Williams is also doing some dilatometric studies to determine the shrinkage effect and how the O/U ratio and atmosphere affects sintering. The effect of cover-gas atmosphere on sinterability of $UO_{2.18}$ is seen in Fig. 5.

Lloyd Hall discussed the objections that the British have to the manufacture of fuel elements by the swaging technique. They are as follows: (1) the filling of the container with uranium oxide is not a simple operation, (2) the swaging could cause bursting of the tube, (3) the cap used to seal the can must be carefully designed so that no uranium oxide reaches the sealed edges, (4) the difficulty of maintaining straightness, (5) the difficulty of maintaining the temperature during swaging, (6) the excessive handling during the over-all swaging operation, (7) the inability to inspect the can after the swaging

[redacted]

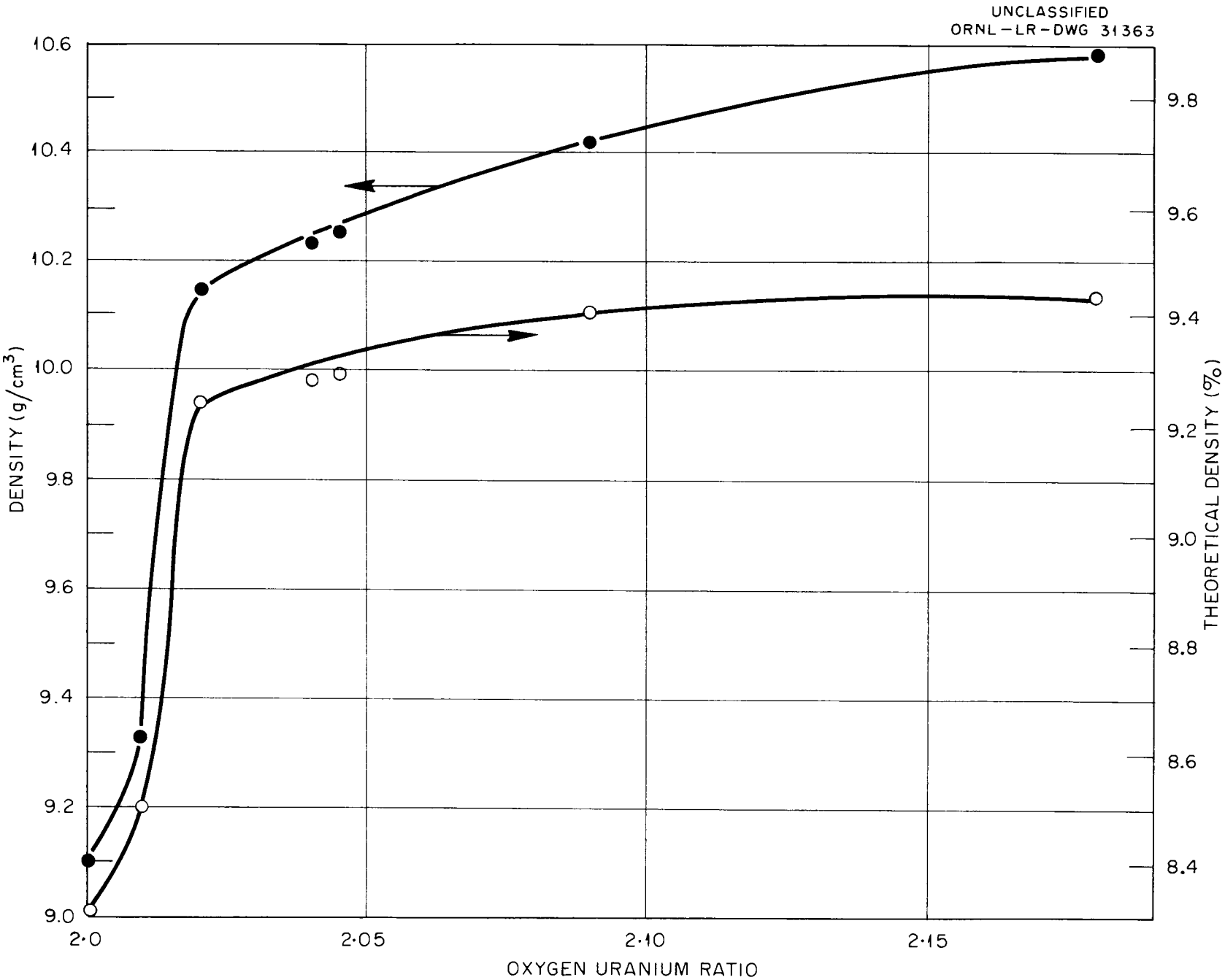


Fig. 1. Density-Composition Relationship.

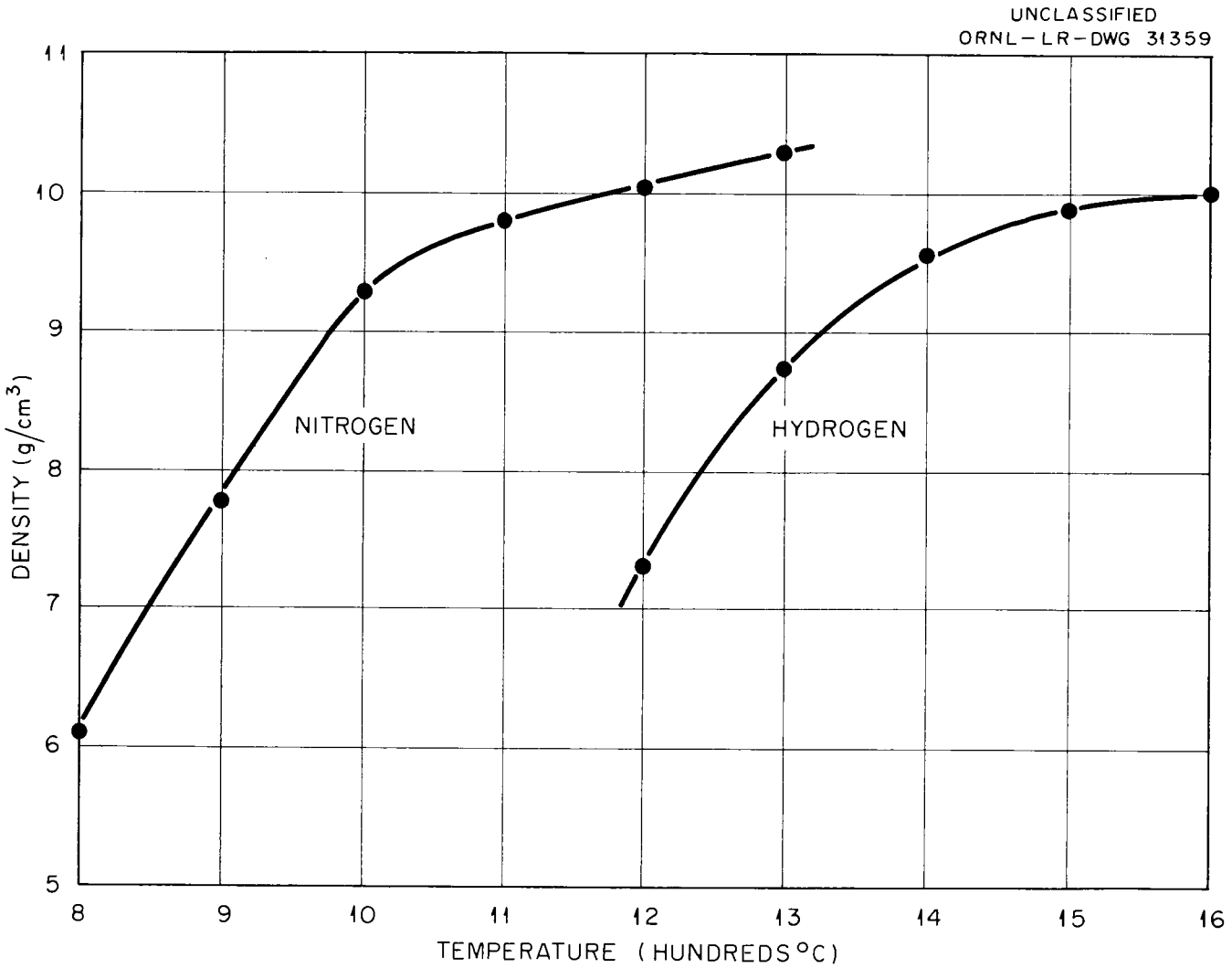


Fig. 2. Sintering of $UO_{2.18}$ in Nitrogen and Hydrogen (After 2 hours at Temperature).

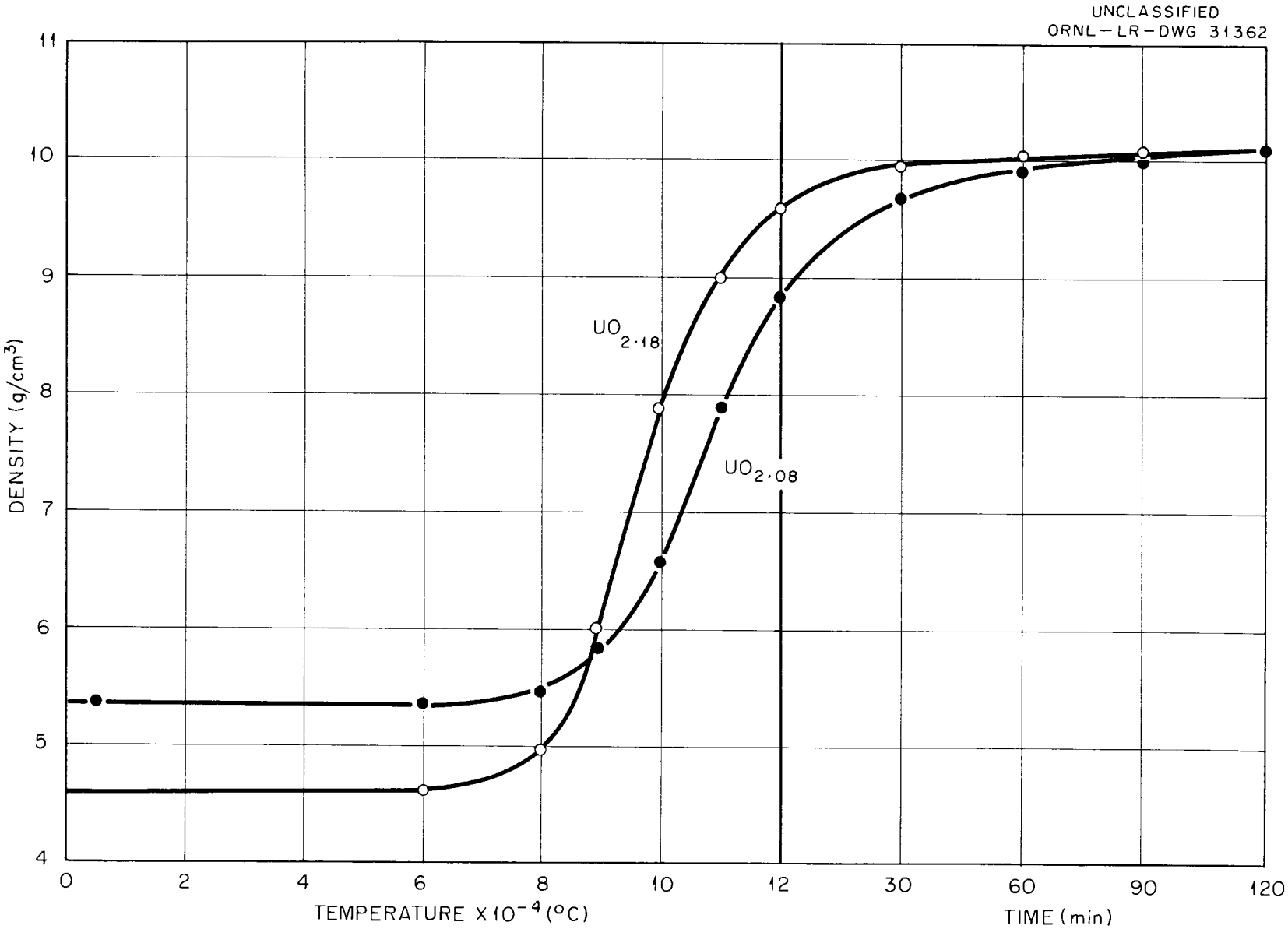


Fig. 3. Sintering $UO_{2.18}$ and $UO_{2.08}$ in Argon or Nitrogen (Dilatometer).

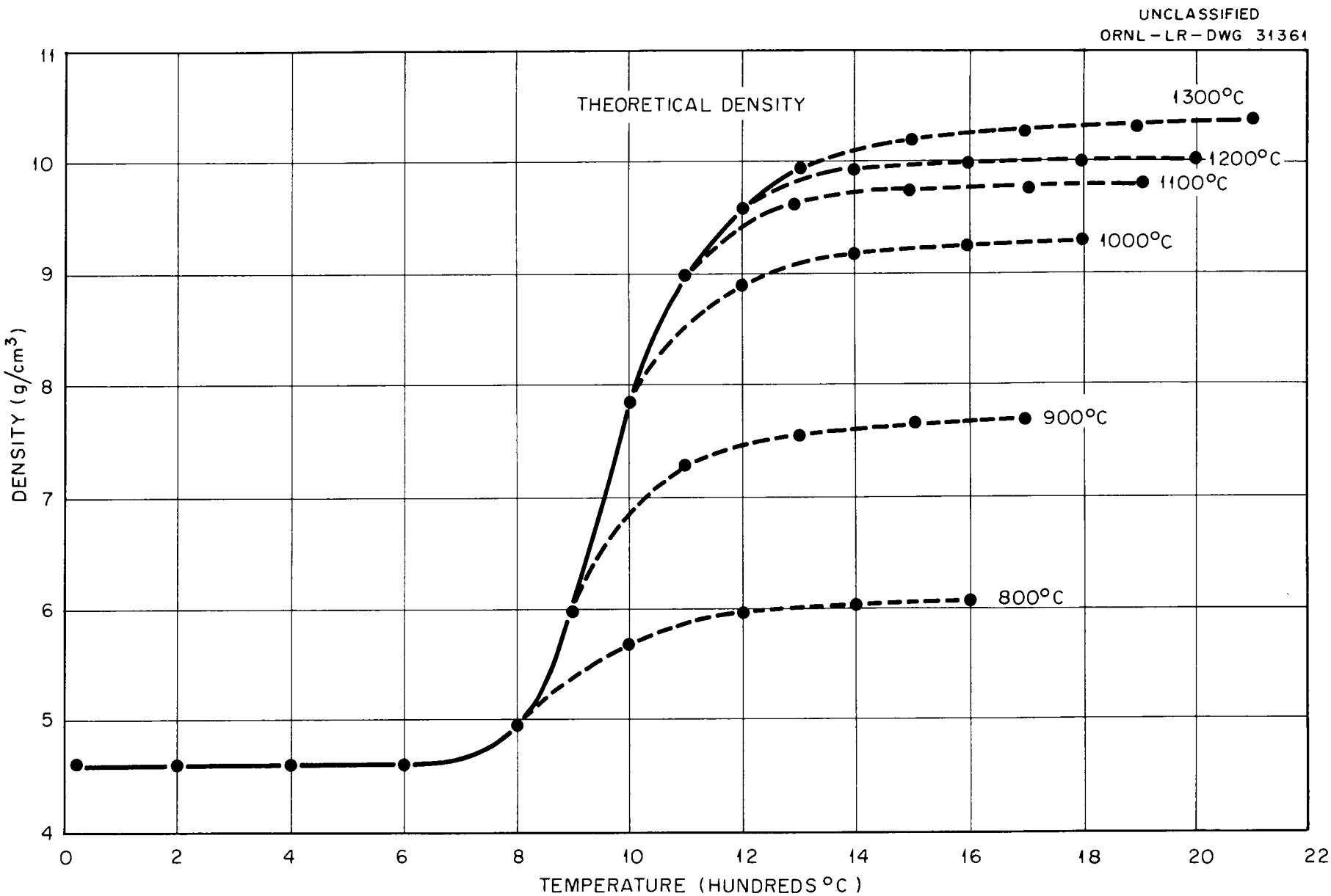


Fig. 4. Sintering of $UO_{2.18}$ in Argon and Nitrogen.

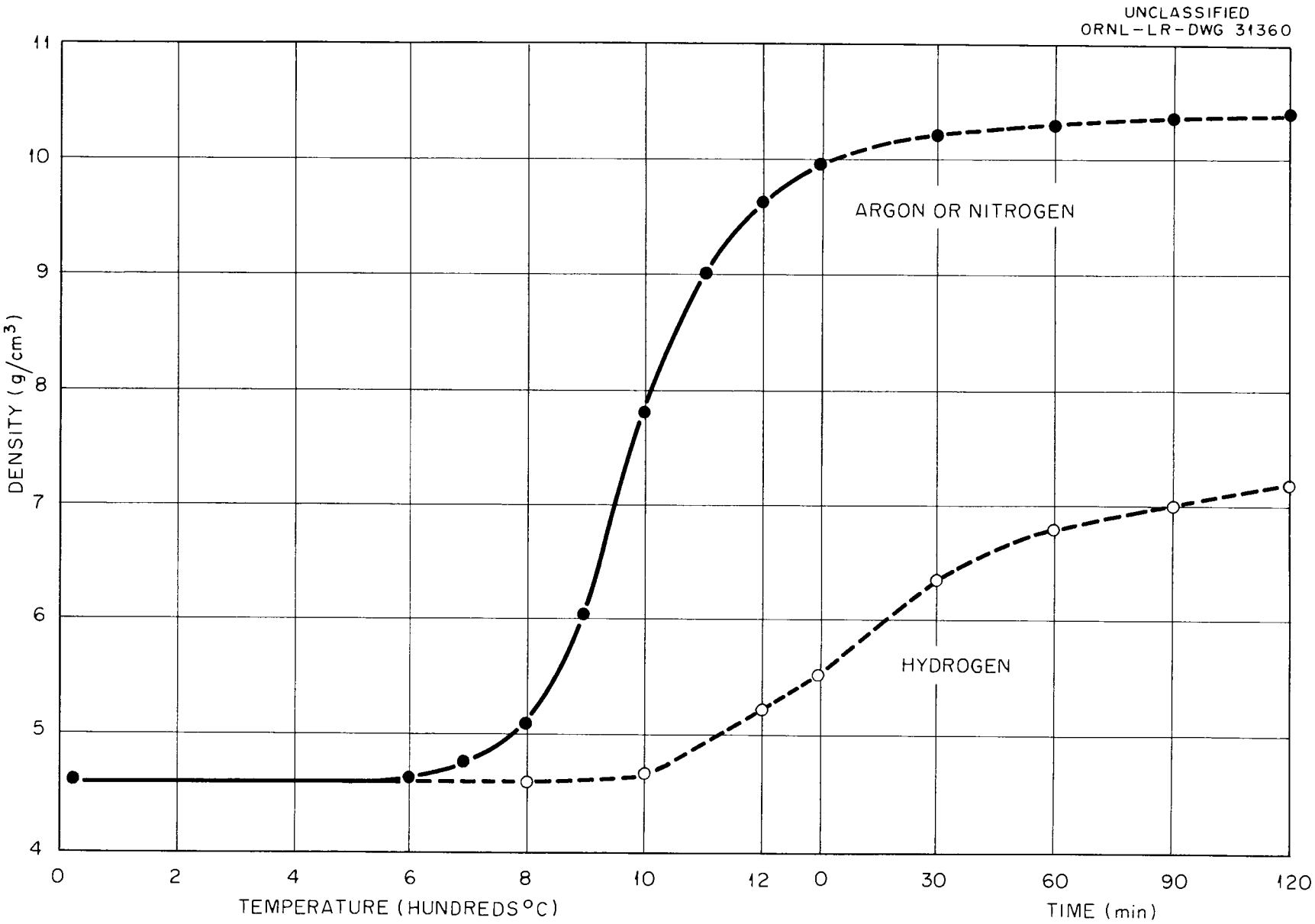


Fig. 5. Sintering of $UO_{2.18}$ in Argon, Nitrogen and Hydrogen (by Dilatometer).

[redacted]

[redacted]

[redacted]

operation, and (8) the increased oxygen-uranium ratio due to the entrapment of the air in the can.

Cadwell from Hanford discussed their swaged fuel element program and reported that the swaging process has merit. They were able to develop good can enclosures; the filling was not too difficult; and the radiation experiments have shown the element to be stable. In addition, the equipment investment is quite small, since it costs approximately $10,000 to buy a Norton Swager. They were able to obtain a density of ~ 90% of theoretical. It was pointed out by S. Pugh that this element was for a water-cooled reactor and was not necessarily the same type of element that is needed for a gas-cooled reactor.

Dimensional control and density tolerances on the uranium oxide bodies were then discussed. Everyone agreed that the density should be in the neighborhood of 95 ± 1%. In the ORNL GCR-2 design, a diametrical tolerance of ±0.005 as-sintered and ±0.001 as-ground was allowed. The personnel at Springfields are considering a ±0.002-in. tolerance on the diameter as-sintered and ±0.0005 in. after grinding for the AGR fuel elements. Since they have already made some 10,000 pellets using the Harwell procedure and have had no difficulty in maintaining these dimensional tolerances, they feel that this is conservative. The ±0.0005 dimensional tolerance on the diameter is quite important in the AGR fuel element in order to keep the center temperature down to a minimum, since the center temperature and the over-all uranium oxide body temperature should be kept as low as possible to minimize the fission-product escape. They calculate that the center temperature of the UO_2 body is ~ 1800°C.

Les Grainger feels that the AGR fuel elements as-designed have a conservative surface temperature and life time. They will test the fuel elements, studying the effects of variables such as the diameter, gas pressure, fuel element temperature, diametric clearances, etc. on the over-all life of the element in the AGR, to give them some feel for what they can do in future reactor designs.

It was pointed out that a document[2] has been issued which reports that the thermal conductivity of uranium oxide is fairly similar to Kingery's data.

Under item B-1 of the agenda, "The influence of stoichiometric ratio on fission-gas retention and radiation damage," Lewis told about some experiments in which there was little gas release from bodies with standard oxygen-uranium

[2] The Thermal Conductivity of Uranium Oxide, M/R 2526.

[redacted]

[redacted]

ratio of 2.02 until a temperature in excess of 1000°C was reached. At 650 – 700°C very little gas was released and above this temperature the release increased rapidly. When nitrogen was used in place of hydrogen as the cover gas, the release of fission products was increased due to oxygen contamination.

S. Pugh told about some studies on the effect of oxygen-uranium ratio on fission-gas release. They were unable to determine whether the difficulty with the fission-gas release experiments was due to the oxygen-uranium ratio or to the large temperature differential, since they observed something similar to central melting in the body. It is planned to irradiate pellets prepared by the AGR technique in Chalk River. Tests of hot-pressed material are also in process. However, it is their feeling that the big difficulty in their experiments in the past was due to their not having the proper O/U ratio.

The Harwell workers described some irradiation tests on uranium oxide-magnesium oxide compacts in which small particles of UO_2 were used; approximately 2% of the fission gases were released in the temperature range of 200 – 250°C, and virtually all were released at 800°C. Using UO_2 with a particle size of 100 μ, once again a 2% fission-gas release was observed in the 200 – 600°C range, but there was a much smaller release of the fission gases at 800°C.[3]

On the basis of their studies, the Harwell personnel feel that uranium diffusion is the controlling factor in the fission-gas release. They have determined that the activation energy for release is ~ 60,000 kcal, which agrees closely with the activation energy for uranium diffusion in the body rather than oxygen diffusion, which has an activation energy of ~ 30,000 kcal.

Under item B-3, "The effect of radiation damage on such properties of UO_2 as the thermal conductivity, vapor pressure, and melting point," the Harwell representatives had little definitive information. They felt that radiation damage had a very serious effect on the vapor pressure, but the melting point would not be affected until a burn-up in excess of 10,000 Mwd/T was reached.

Under item B-4, "The effect of high atomic burn-up > 10,000 Mwd/T on fission-gas retention," they had no data.

Under item B-5, "Mechanisms of diffusion and escape of fission gases from internal voids," there was no additional information presented other than B. Lustman's analysis of the problem.

[3] AEREC/M-343.

[redacted]

[redacted]

Bob Barnes discussed some work on heating a thorianite mineral which fragmented to powder when heated to 950°C; this was due to the helium formed there by radioactive decay. However, the amount of gas is equivalent to something like ten times the burn-up that would occur in the uranium oxide bodies at 10,000 Mwd/T exposure.

S. Pugh referred to some work that is being done by the Canadians in an effort to determine the effect of grain-boundary diffusion and lattice diffusion on fission-product gas release. This is being done by Mr. B. T. Childs and co-workers. Mr. Childs is also performing a calorimetric determination of stored energy in uranium oxide at the Chalk River Site. It is their opinion that at an irradiation temperature of 1000°C energy would not be stored in the body.

Grainger and the others were asked a question about the difference between a cored or a solid pellet. A solid pellet was selected for the AGR because of its simplicity. Consideration was also given to using a cored element which utilizes a high-porosity core for the extra voidage for the fission products. They may consider using a UO_2 body with a low ID/OD ratio and a pure MgO core to facilitate the fission-gas problem.

The "ratcheting" effect between the UO_2 and the can and the breakup of UO_2 due to thermal stresses were discussed briefly. If the length of the body is approximately two diameters or if the pellets stick together, ratcheting may occur with the stainless steel can. This ratcheting results from movement of the can in relationship to the uranium oxide body. They feel that the AGR beryllium can will not distort around the uranium oxide, and therefore, there will not be a ratcheting effect. There will be approximately 0.010-in. clearance between the uranium oxide body and the beryllium can. However, they are concerned about the possibility of ratcheting and failure of the can made of stainless steel, since the 0.010-in. can wall will collapse around the UO_2 body at 750°C.

Cracking of the UO_2 body has been observed by the Harwell personnel as well as by the personnel at Westinghouse and Chalk River, but no powdering of the uranium oxide under irradiation has been detected. It is their feeling that the breakup of the uranium oxide body has been clearly due to the thermal stresses set up in the body and not due to any induced radiation damage.

[redacted]

[redacted]

Reaction of UO_2 with CO_2 was mentioned. Below a temperature of 800°C the thermal reaction was not measurable. However, under irradiation some oxidation occurred, with the UO_2 being oxidized first to U_3O_8 and then to UO_3. They summarized this by saying:

1. In a high-temperature capsule series, it was determined that the end product found in the capsule is solely a function of the temperature of the experiment.
2. In some loop experiments, it was found that irradiation increased the rate of the oxidation-reduction reaction, but the actual equilibrium was not shifted.

However, if there is a small percentage of oxygen as an impurity, the UO_2 will oxidize.

The effect of fission-product damage on some crystals of lithium fluoride and sodium fluoride has been studied, and marked surface effects were observed. For instance, with the exposure of lithium fluoride at 80°C, an etch-pit pattern was found that differed with increasing irradiation level of the material.

We asked about the beryllium-uranium oxide reaction, but the British did not have any information. However, Ed Hayes of DuPont spoke about some Be + UO_2 extrusions that were processed at 1000°C by Nuclear Metals in which alloying did occur.

We asked the approximate number of people doing uranium oxide work in the United Kingdom, and were informed that there are approximately twenty people in the Industrial Group at Risley, twelve people at Harwell, and eight people doing radiation damage work at various sites.

Jack Williams spoke about the plastic deformation of uranium oxide in which they were performing some simple beam-bending measurements over a range of temperatures. The rate of the deformation is similar to a metallic creep-type curve. From his work, Mr. Williams has determined that the ΔH value for such a phenomenon for a UO_2 body with an oxygen-uranium ratio of 2.00 is 95,000 cal, whereas for an O/U ratio of 2.08, it is 70,000 cal, and for a ratio of 2.16, it is 65,000 cal. The explanation for this is the rapid change in the viscosity as a function of the oxygen content.

Jack Williams also described some other experiments in which the plasticity of UO_2 was seen to increase with increasing O/U ratio, and at a O/U ratio of

[redacted]

[redacted]

[redacted]

2.08 the UO_2 body could be bent. Also, a UO_2 body with an O/U ratio of 2.16 has a modulus of elasticity one-half that of oxide with an O/U ratio of 2.04. It was stated that uranium oxide with an O/U ratio of 2.16 at 975°C has the plastic properties of aluminum at 240°C or glass at 500°C.

Uranium

We asked the British to give us their latest information on the behavior of uranium under irradiation. This was summarized for us by Finniston. He feels that the effects are real and that what he presented to us was not only a qualitative picture but also a semi-quantitative study.

1. The growth of uranium stops at approximately 500°C. The British formerly thought that it stopped at 400°C.
2. Swelling commences below 500°C.
3. There is no definite correlation between the degree of swelling and the temperature level between 400 – 600°C. He mentioned that there was a wide scatter in results, and since there were few tests at any one temperature, it is quite difficult for them to get any general picture of what happens between 400 – 600°C.
4. When a sample is held above a certain critical temperature, the swelling increases quite readily with burn-up at approximately 5000 Mwd/T.
5. With samples showing a density decrease of 20 – 30%, the fission-product gases were released from the uranium, whereas at a 2 – 5% change in density the fission gases are retained in the uranium lattice.
6. Alpha-uranium alloys offer no help in stopping the fission-product swelling. In the gamma-uranium alloys, if considerable quantities of molybdenum are added, some help in minimizing the fission-product swelling can be utilized.
7. Thermal cycling greatly accelerates the swelling. An example of this was given in that a pure-uranium sample with a ±X temperature cycle swelled 146%, while one with a ±0.1 X temperature cycle showed approximately 4% swelling. The sample that showed 146% swelling had 0.34% burn-up while the sample that swelled 4% had an 0.41% burn-up.

When asked about this in more detail, we found that they were talking about a base temperature of 600°C with pure-[redacted]es. The sample that swelled 146% had a ± temperature cycle of ~ 10°C whereas the other one had a ±2°C

[redacted]

[redacted]

variation. When Stan Pugh was carefully reviewing the data from a report to tell us about this experiment, he remarked that there was no significant difference in swelling over the temperature range of 450 – 800°C, but there was a considerable scatter of the data. He also emphasized that the greater the temperature cycle the greater the amount of swelling.

The British commented on the American radiation-damage experiments with uranium. The temperature has been calculated in the United States experiments, and it is actually higher than is reported. The temperatures of American experiments are low as compared to the British temperatures. The Argonne National Laboratory reports less swelling than the British for equivalent burn-ups at temperature. The British work, as discussed, is based upon tests of 1000 Mwd/T burn-up in Calder Hall and tests of 2000 – 3000 Mwd/T burn-up at Harwell.

They were asked if any serious difference in their conclusion would occur if a large temperature gradient exists and they did not know how to analyze the problem. S. Pugh said that there was no pattern or correlation between the formation of the fission-gas voids in the crystals of uranium and the inclusions that already existed in the uranium, since under irradiation the entire inclusion pattern changes. The carbides disappear and seem to reappear on the surface of the uranium bar. In describing similar experiments, specifically specimen 217 of Harwell series, swelling did not occur and the carbide network, or structure, remained unchanged throughout the experiment. Of the ones that swelled, the carbides disappeared and then reappeared on the outside surface of the uranium bar.

Manly then specifically asked the question: "Wouldn't this give you a clue that the lack of swelling was because the carbide network would minimize the creep of the uranium?" He also asked what would cause the movement of the carbon. The answer to the first question was that they did not feel that a conclusion could be drawn that the carbide network minimizes swelling. Next they did not know the reason for the carbide disappearance. They suggested the possibility that hydrogen in the uranium could react with the carbon to move the carbide phase from the center to the outside. We were referred to a report by Greenough.[4]

In the early days, considerable discussion centered upon using a heavy-walled can to restrain growth. Cottrell and co-workers studied this and determined

[4] Greenough, The Volume Increase in Irradiated Fuels and the Effect of Thermal Cycling, AERE M/R 2609.

[redacted]

[redacted]

that the strength needed in the can could not be obtained with the present canning materials in the British program. The British then spoke about the alloy addition studies in which they made minor additions to uranium and then irradiated them under supposedly carefully controlled conditions. Over 100 alloys were made and the samples were exposed to 0.001% and 0.1% burn-up. With the exception of the gamma-phase alloys with molybdenum additions, none of the alloys seemed to have any effect on swelling.[5]

It was their feeling that there was considerable lack of control in the previous experiments and they now know what needs to be controlled to get meaningful results. Therefore, serious consideration is being given to preparing all of the alloys and repeating the experiments and then correlating the results with data from a creep testing program on these alloys.

They also want to determine what happens during the cracking of materials at large burn-ups. One thing that bothers them a great deal about their uranium work is that they cannot explain why they observe a 20% density change but detect only 10% porosity in the uranium.

It is Grainger's belief that uranium-metal fuel elements can be used in the swelling range. Both he and Monty Finniston think that maybe we are moving too soon into the uranium oxide technology, since we may run into difficulties with UO_2 fuels in the same manner we did with uranium metal. There is a lot known about uranium-metal fuels and this is one of the reasons that we are aware of the difficulties. Both Les Grainger and Monty Finniston believe that much more can be accomplished with uranium-metal fuel elements at elevated temperatures with the proper design to allow for the swelling.

Beryllium

A discussion on beryllium was started and Jack Williams described their beryllium fabrication program. Harwell started making sheet material the middle of last year, and were able to get reasonable ductility by cross-rolling the forged material. The beryllium sheet exhibited approximately 25% ductility similar to what Kauffman has reported. The size of specimen used and their testing methods had quite an effect on the amount of ductility reported. If the test is made with an 0.1-in.-wide beryllium strip, a much higher ductility figure is obtained than if an 0.2-in.-wide beryllium strip is used. The British must make beryllium tubes ~ 0.3 in. in diameter with a wall thickness of

[5] AERE M/R 2004/A.

[redacted]

[redacted]

0.040 – 0.060 in. and length of approximately one foot for the AGR. Industrial firms, such as Imperial Chemical Industries and Tube Investments, are ready to quote prices on producing tubes of this type.

The procedure for making beryllium tubes was developed by Harwell and is as follows:

1. An electrolytic process is used to prepare beryllium powder from a Na-K-Be chloride eutectic. This procedure produces beryllium flakes similar to the Clifton beryllium flakes used in the early days of the American program.
2. The beryllium is sintered into an extrusion billet at 1200°C with a slight pressure. A carbon mold with a beryllium oxide wash contains the beryllium.
3. The billet is coated with silver approximately 0.002 in. in thickness as the extrusion lubricant.
4. The billet is extruded at 700 – 750°C with extrusion ratios from 1:6 – 1:20. They probably will use an extrusion ratio of 1:16 in production. A colloidal dispersion of graphite wash is used on all the dies. A high-speed die steel is used for the mandrel, and a high-tungsten type steel is used for the die container.
5. The container is heated continuously at 700°C to prevent the beryllium from cooling during the extrusion.
6. The tubes are machined to final size, ID and OD, then cleaned and inspected.

The British have done some redrawing of beryllium and approximately 5% reductions per pass can be tolerated if the beryllium is warm, i.e. 400 – 500°C. The big difficulty is gripping the tube since this causes the material to break up.

Their first objective is to make the beryllium tube for the AGR. After this has been accomplished, their efforts will be directed toward producing beryllium tubing that has ductility in all directions.

Williams discussed the type of textures that is developed in the beryllium tubes. At an extrusion ratio of 1:11, there is considerable change in the texture. They have studied the textures of beryllium with reductions of 1:6 – 1:50. Below a 1:11 extrusion ratio the basal plane is circumferential around the tubing. Above a 1:11 extrusion ratio the basal plane is radial...

[redacted]

[redacted]

The British have an effort of approximately 20 to 30 people on the beryllium program performing their studies with the Industrial Group at Risley, Springfields, and Culcheth and with the Research Group at Harwell. In addition, they have considerable effort under contract in the various industrial concerns.

The British are now producing one to five tons of beryllium a year, and they hope to raise this figure approximately a factor of ten within the next year. Their plans are to get the cost of beryllium to approximately $100/lb for fabricated tubing for the AGR, with a possible reduction to $30 – $60/lb for fabricated tubing in the future. This was the figure that was quoted by the Consortia people.

Bob Barnes, who works with Cottrell, spoke about the work on the in-pile testing of beryllium to determine the extent of the n, α, 2n reaction. This has a very small cross section and the beryllium must be in a reactor a long time to show much effect. To do this in a more expeditious manner, they injected 40-mev alpha particles into the metal until approximately 1 at. % helium was produced in the beryllium. This is equivalent to a dose of approximately 10^{22} fast neutrons. Great care was exerted to keep the beryllium cool during the injection of the helium with the Cyclotron so that the formation of helium bubbles in the metallic lattice could be studied with heating experiments. The bubbles grew in size at higher temperatures and decreased in number. As the bubbles began getting larger, the beryllium started swelling. When the bubble is small the surface tension of the bubble keeps it from exerting pressure in the beryllium. Studies were also made of cast beryllium with a large grain size, and the same general behavior was observed. However, it was necessary to heat the metal to a higher temperature in order to initiate the movement of the helium for bubble formation to start swelling.

Mr. Barnes explained that vacancies were required for the process, and these vacancies are produced at grain boundaries and must move into the grain. This then allows the helium that is produced in the grain to diffuse. A temperature of approximately 700 – 800°C must be reached before much vacancy movement occurs. This explains why the bubbles will not be formed until a temperature of ~ 700°C is attained. The larger the grain size the longer the diffusion path for vacancies to move to get into the grain. This will minimize the helium mobility; therefore, the swelling and bubble formation will be less severe with a large grain-size material.

[redacted]

[redacted]

One of the bubbles that precipitated in beryllium which was heated for 3 hr at 700°C was measured and found to be 1000 angstroms in diameter. This was extruded beryllium, and upon heating, bubbles appeared in the matrix at approximately 700°C. Below 700°C the helium was in solution in the beryllium and only affected its etching characteristics, since specimens etched much faster in those portions which were exposed to the helium injection. In the injection experiments the helium comes to rest at approximately 0.001 in. below the surface.

Mr. Barnes discussed their heating experiments with pieces from the MTR shim rod. The dosage was approximately 5×10^{21} nvt for the shim rod material. Gas analyses were performed on the samples, and different amounts of gas were released from the various pieces. This was due to the difference in the manufacturing of the material, or differences in the level of irradiation. At a temperature of 600 – 700°C, the beryllium shim rod material had an etched effect due to the beginning of bubble formation. Once again, at higher temperature the bubbles appeared larger in size and fewer in number, and at 600°C the beryllium started swelling due to the helium coming out of solution. The swelling reached a maximum at approximately 1000°C with a 30% increase in volume. Above this temperature it is believed that the gas is lost due to interconnected holes, so a larger increase in volume is not obtained. (See Fig. 6.)

The bubbles that are growing are predominantly at the grain boundaries. The bubbles that appear in the grains remain approximately the same size once they are formed. The British are concerned about using beryllium above a temperature of ~ 600°C because the bubbles would line up and form a continuous path for the fission products to escape from the tube. Also, this would completely weaken the beryllium structure from a load-carrying standpoint.

Studies have not been made of the effect of the n, α reaction on the mechanical properties of beryllium. The hardness of the MTR shim rod beryllium was approximately two times what it was thought to be in the cold condition. A decrease could be seen in the hardness of the material after heating 1/2 hr at 700°C; this is presented in Fig. 7.

At 900°C the beryllium is quite soft. This is probably due to the many large holes, but there is no ductility at all in this type of material. It was explained that small holes in the beryllium increase the hardness whereas the large holes make it softer.

[redacted]

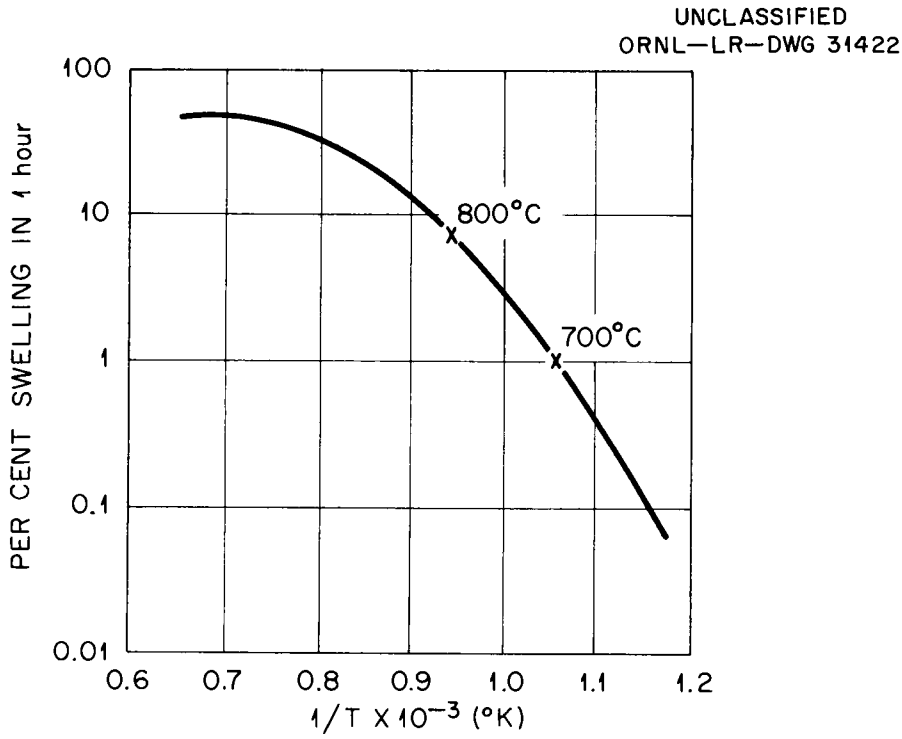


Fig. 6. Swelling Produced after 1 hour Annealing at the Temperature for MTR Shim Rod, Berylium.

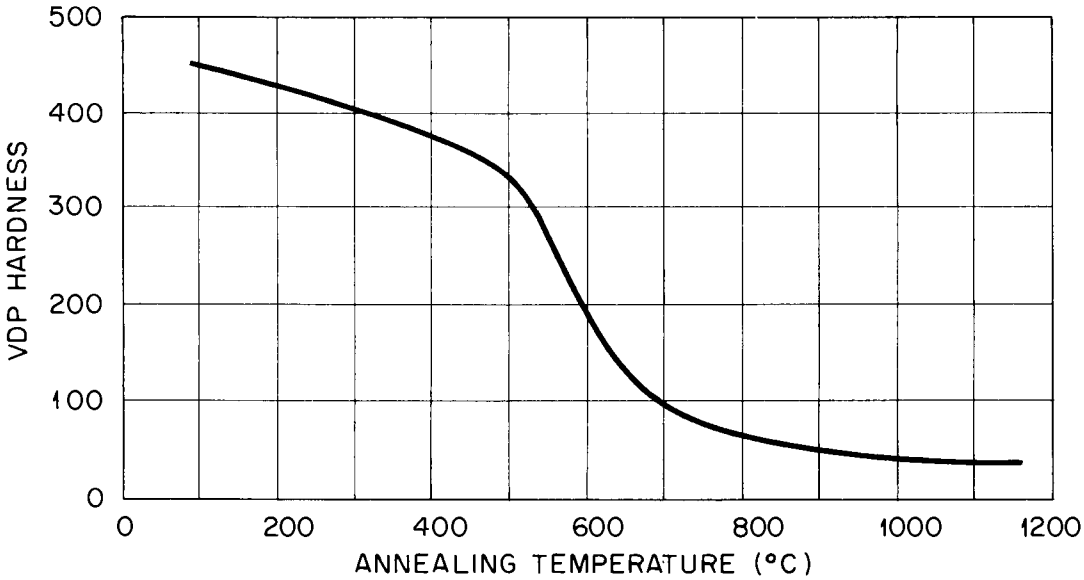


Fig. 7. Annealing Out Radiation Hardness of Berylium from MTR Shim Rod.

[redacted]

The lower temperature limit at which the bubble effects would be observed with the beryllium can be raised by increasing the grain size. In beryllium that has been cold-worked considerably, the bubbles can be seen throughout the grains, since dislocations formed within the grains can act as nucleation centers for the bubbles. In a well-annealed material the precipitation of bubbles can occur only at grain boundaries.

In the MTR shim rod beryllium, it was difficult to correlate radiation information with density changes and evolutions of gas due to unknowns in the flux, unknowns in the original density of the beryllium, and very poor experimental control. It is planned to repeat these experiments with more carefully controlled starting material and radiation behavior. Creep studies will also be made on the beryllium after irradiation. It is their belief that the helium evolution for the MTR shim rod material was some place between 4 to 16 cc/cc, and a comparable quantity of hydrogen was evolved. It is Mr. Barnes feeling that 400°C is about the maximum temperature that can be used for long-time applications. However, in the AGR the specifications call for the beryllium to operate at a temperature of 550°C and not to exceed 600°C.

Barnes and co-workers have completed injection studies of helium into cold-worked and annealed copper, using high-energy alpha particles to determine if the same type of bubble formation occurs. They were quite pleased with the correlation between the copper and beryllium results, and the explanation of the vacancy movement as the mechanism which allows the helium to escape from the grains was upheld.[6] They have also written reports on the beryllium studies.[7,8]

Jack Williams described their beryllium welding and joining studies, to seal end caps in beryllium tubes. Brazing was tried, but due to the extremely low-melting brazing materials and the brittle nature of these materials the results were not impressive. The magnesium-aluminum and the aluminum-silicon brazing alloys were considered, as well as the silicon-beryllium eutectic. The latter was not successful because silicon formed a continuous brittle phase.

---

[6] I. S. Barnes, G. D. Redding, and A. H. Cottrell, "The Observation of Vacancy Sources in Metals," Philosophical Magazine, 3, 25, 97 (January, 1958).

[7] Injection of Helium in a Beryllium Cyclotron Target, M/R 2332.

[8] Injection of Helium into Cast Beryllium, M/R 2332A.

[redacted]

[redacted]

Fusion-welding was studied and believed to be a reasonable method since it can produce a gas-tight weld; however, this type of weld would not have any mechanical strength due to the extremely low ductility of the weld material, and it must be kept completely unstressed under all operation conditions. Finally, the selection made by the British for sealing the AGR tubing is the pressure-welding technique. The end cap and the inside and outside diameter of the beryllium tube in the region of the weld are carefully cleaned by pickling.

The cap is inserted into the can and the tube is heated and outgassed in a vacuum system. The cap is pressure-bonded to the tube at 850°C with approximately a two-minute stroke on the ram using a few tons per square inch pressure. The equipment used for the pressure-bonding is seen in Fig. 8 and typical closures are shown in Fig. 9. The pressure of upsetting varies with the type of beryllium and the reduction ratio that was used in extruding the beryllium tubing.[9]

In case the British tubing-extrusion program is not successful, tubing can be produced by machining of extruded rod. Experiments of this type have been performed and 0.3-in.-ID by 0.060-in.-wall tubing, 21 in. long, has been made by such a technique, with a 40% use factor of the metal. However, if for some unknown reason the beryllium is not available, stainless steel tubing will be used for the AGR. Mr. Williams stated that the beryllium can for the AGR will be 0.3-in. ID x 0.060-in. wall, 6 to 9 in. long, operating with a surface temperature of 550°C. If stainless steel is used, the fuel cans will have an 0.010-in. wall and will operate at a surface temperature of 750°C.

The Harwell personnel are quite interested in the creep properties of beryllium. They informed us that most of their creep work on beryllium, in the absence of radiation, has been performed at the Admirality Materials Laboratory at Holton-Heath, Poole, Dorset. They would appreciate receiving any data from this country on the creep properties of beryllium to assist them in their over-all program.

We had hoped to be able to talk to Mr. G. N. Walton, who is impregnating graphite with various fuel-bearing materials, but he was not available.

Liquid Metals

Mr. G. W. Horsley, who is studying container materials for bismuth and bismuth-lead systems, described his work with miniature loops operating at

[9] Muneo and Williams, The Pressure Welding of End Plugs and Beryllium Tubes, M/M-189.

[redacted]

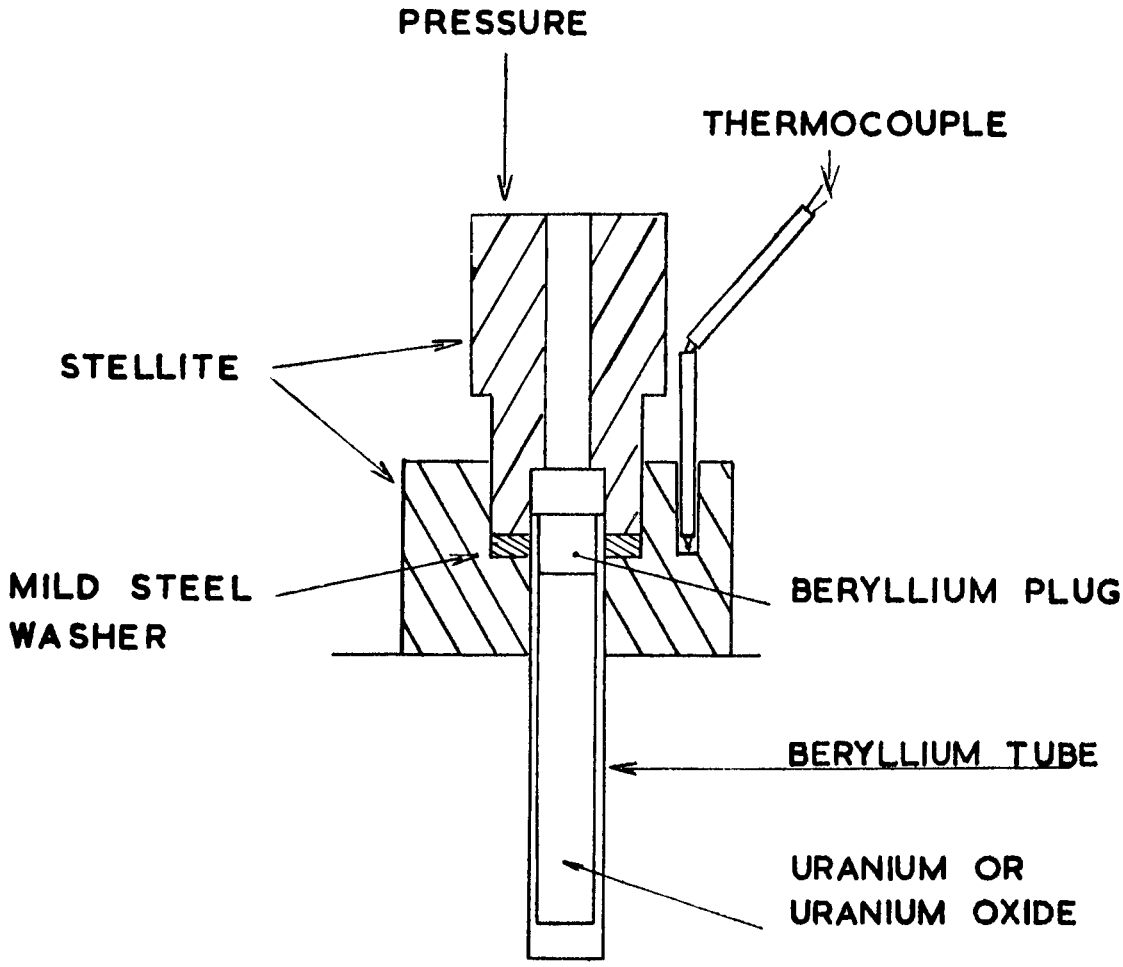
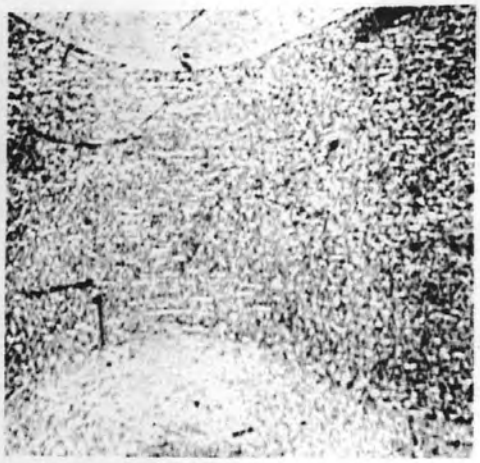
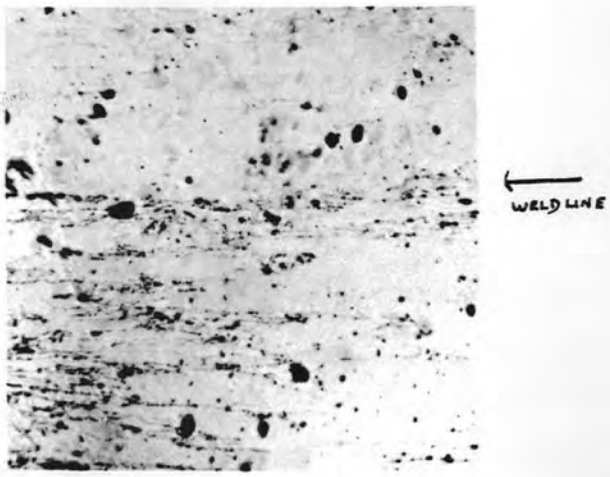


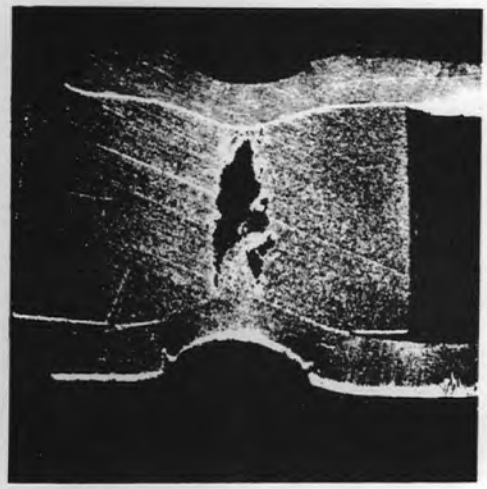
Fig. 8. PRESSURE WELDING APPARATUS



Section of Pressure Weld 6x

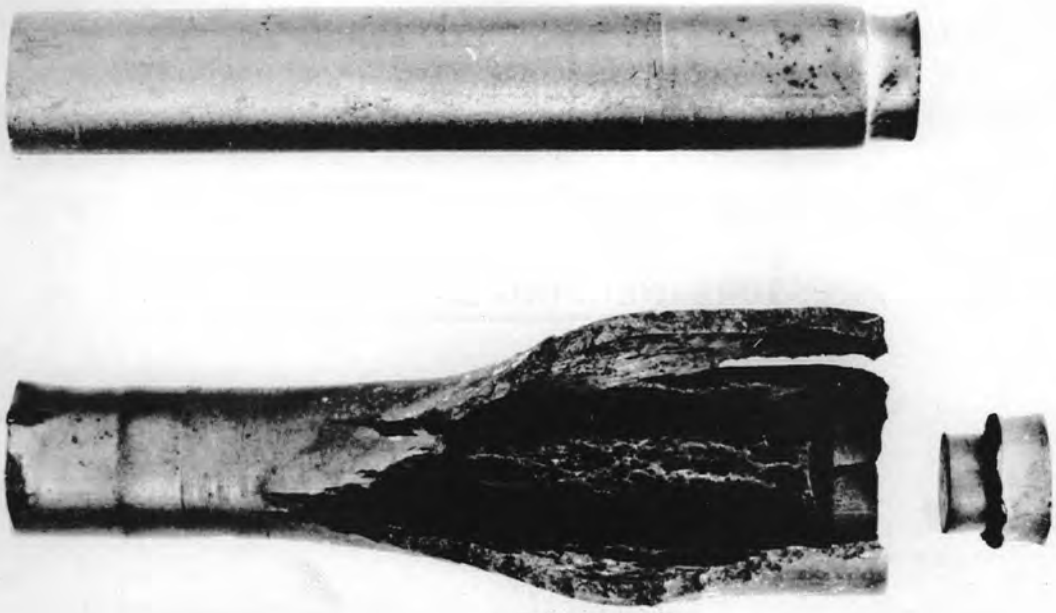


Weld Line in Pressure Weld



Section of Pressure Weld Showing Failure of End Plug 3x

Section of Pressure Weld Showing Failure of End Plug 10x



Can As-Welded, and also after Failure on Thermal Cycling

Fig. 9 Typical Pressure-Bonded Closures.

[redacted]

600°C on the hot leg and 400°C on the cold leg. His work is quite similar to the studies and findings of the people at the Brookhaven National Laboratory. He is determining the effectiveness of diffusion barriers in inhibiting mass transfer by additions of zirconium to form zirconium carbide or zirconium nitride on the surface of the steel. In an effort to obtain a type of coating, or barrier, that will be self-protective, he has given considerable thought to making alloys containing zirconium, nitrogen, and carbon.

He is preparing a series of alloys using an iron-2-1/2% Cr base and an iron-1% Mo base, to which he will add considerable quantities of nitrogen. He will then screen this material by nitriding the inside surface of a small thermal-convection loop (3/8-in.-OD tubing, loop 3-3/4 in. high by 2 in. wide) and placing the alloy specimen in the hot leg of the loop.

Mr. Horsley is hoping also to make some steels of iron, zirconium, and carbon, and the zirconium-carbon ratio will be varied in an attempt to get good dispersion of zirconium carbide in the steel. Mr. Horsley feels that the zirconium nitride and zirconium carbide repels the lead-bismuth eutectic and prevents it from wetting the surface.

Harwell is sponsoring considerable liquid-metal research at various organizations. The Metro-Vickers of Manchester are determining the molybdenum-uranium-bismuth solubility in the high-uranium part of the system. The General Electric Company of England is studying thorium oxide slurries in bismuth. Hawker Siddley at Slough is performing loop tests and determining the effect of inhibitors on loop corrosion. The Associated Electrical Industries at Aldermaston are studying isothermal mass transfer in various circuits and the compatibility of lead, lithium, aluminum, and bismuth. They have just released much of this information.[10] The British Thomson-Houston Company, Ltd., at Rugby are doing compatibility tests and in-pile tests with bismuth, and are designing pumps and constructing pump loops. They are able to place twenty specimens in the hot leg of their pump loops by suspending them with molybdenum wires. Fulmer Research is conducting fundamental research on liquid metals and is determining the activity of uranium in bismuth solutions. Oxford University is doing equilibrium studies on bismuth and lead. Cambridge University is studying the influence of a third element to the solubility of metal A in Metal B, such as the effect of bismuth on the solubility of iron

[10] J. Nuclear Industry.

[redacted]

[redacted]

in the lead-bismuth system. Nottingham University is studying various surface effects in liquid metals and the fundamentals of wetting. Birmingham University has a contract, but Mr. Horsley was not familiar with their work. Imperial College, London University, is working on the uranium-lithium system and the solubility of various materials in the uranium-bismuth system.

Slurries

Mr. G. H. Broomfield described their work with slurries, and we were informed that their studies on sodium-uranium oxide slurries had been stopped. The only slurries that they are working on now are uranium oxide in lead and lead-bismuth eutectic and thorium oxide in lead and lead-bismuth eutectic. The British terminated slurry studies with metallic uranium or thorium due to the nucleation and growth of the particles in the cold zone. They also have given up studying semi-soluble slurries, such as intermetallic compounds of uranium and bismuth, for the same reason. Mr. Broomfield informed us that it takes considerable stirring to get the thorium oxide into the bismuth. They add calcium and/or magnesium to the bismuth which helps in promoting wetting.

The GEC people have performed gas analyses on samples of the thorium oxide and uranium oxide by pumping off the gas and have found considerable water and CO_2. The CO_2 acts as a wetting barrier and must be removed. The maximum concentration of thorium oxide in bismuth that has been obtained is 30%, using a particle size of approximately 1 to 2 microns. The wetting by the bismuth will break down any agglomeration of material that has occurred. The slurries will settle upwards due to trapped gases that are taken into the system with the uranium oxide or thorium oxide particles.

In circulating systems at 550°C with ΔT of 100°C, calcium reduces the thorium oxide. It is thought that magnesium will do this also. A thorium oxide slurry in bismuth with 0.5% magnesium added had an operating time 3000 times greater than that at which a loop using a thorium bismuthide slurry would plug. No evidence of mass transfer was found when the loop was examined. A mechanical properties loop has been operated, using a slurry of 5% thorium oxide in bismuth; after 1000 hr of service, no sign of erosion was observed on the steel pump impeller.

Some experiments have recently been initiated on slurries of uranium oxide in lead, using an O/U ratio of 2.08. These slurries are quite stable in an iron container. The effect of fission-product gases on the stability of the slurry is unknown.

[redacted]

[redacted]

Mr. Broomfield suggested several reports to get a more up-to-date picture of the British slurry work.[11,12,13,14]

Considerable work on heavy-metal oxide slurries in bismuth is being performed by an outside concern. This is under Contract No. 13/5/165/887, under blanket No. 6/Atomic/155 by Chitty, Brown, and Woereys. The United States Atomic Energy Commission personnel may request this information through the US AEC London Office to Mr. V. J. Freestone at Harwell. Mr. Horsley has had a great deal of his work published[15,16,17,18,19,20] and he also quoted the work of Davis and Draycott.[21]

---

[11] Eten, Greenwood, and Broomfield, AERE M/R 2144a and 2144b.

[12] Barton and Greenwood, AERE M/R 2310.

[13] Broomfield and Mathews, AERE M/R 2623.

[14] Greenwood and Sharp, AERE M/R 1829.

[15] G. W. Horsley and J. T. Maskrey, "The Corrosion of a 2-1/4% Cr-1% Mo Steel by Liquid Bismuth," J. Iron and Steel Inst., 189, 139 – 148 (June, 1958).

[16] G. W. Horsley, "The Preparation of Bismuth for Use in a Liquid Metal Fueled Reactor," J. Nucl. Energy, 6, 41 – 52 (1957).

[17] G. W. Horsley, "A Filtration and Vacuum Distillation Unit for the Purification of Alkali Metals," J. Appl. Chem., 8, 13 – 18 (January 8, 1958).

[18] G. W. Horsley, "Corrosion of Iron by Oxygen-Contaminated Sodium," J. Iron and Steel Inst., 182, 44 – 48 (January, 1956).

[19] G. W. Horsley and J. T. Maskrey, "Redetermination of the Liquidus of the System Lead-Magnesium in the Region 0 – 3 Weight Per Cent Magnesium," J. Inst. of Metals, 86, 446 – 448 (1957–58).

[20] G. W. Horsley and J. T. Maskrey, "The Determination of the Solubilities of Beryllium and Molybdenum in Liquid Bismuth," J. Inst. of Metals, 86, 401 – 402 (1957–58).

[21] Davis and Draycott, Compatibility of Reactor Materials in Flowing Sodium, IGR-TN/C-87.

[redacted]

[redacted]

Graphite

Information on the behavior of graphite in reactors was exchanged with members of the UKAEA and several of the members of the UK Nuclear Power Groups.

RISLEY DISCUSSIONS - JULY 8 AND 9

Participants: H. Kronberger
Director Research and Development
UK AEA, Industrial Group

M. T. Kavanagh
Overseas Manager, Risley

R. E. Nightingale

Dr. Hans Kronberger commented upon the recent burning experiments completed at Windscale. They have characterized the conditions under which sustained burning of graphite may occur in air. Their experiments confirm the theoretical model developed by W. B. Hall and J. Woodrow. This work was discussed in more detail with J. S. Nairn at Windscale and a technical description of the work will be given later in the report.

Mr. J. Moore (physicist-Windscale) observed the recent stored energy release in the Brookhaven graphite reactor. He also discussed the burning experiments being done there and concluded that since the Brookhaven experiments were not adiabatic, the worst possible case was not studied. Kronberger believes that a reactor fire may approach adiabatic conditions. The author is not completely familiar with the Brookhaven experiments and therefore requested that Kronberger send his comments directly to people at BNL.

Because the CO_2 graphite reaction is endothermic there is no concern for a catastrophic fire in the Calder Hall reactors as occurred with the Windscale No. 1 pile.

KNUTSFORD - AEI JOHN THOMPSON - JULY 9

Participants - Members of the Gas-Cooled Reactor Team and personnel of AEI John Thompson

Only a short time in the afternoon was spent at Knutsford by the author. Consideration is being given to the use of graphite sleeves in the reactor now being designed (following the Berkeley reactor). Their advance design would have an inlet and outlet CO_2 temperature of 190°C and 380°C, respectively. An individual discussion with Mr. J. R. M. Southwood after the meeting brought out the fact that they have consid[redacted]°C as the practical limit for

[redacted]

[redacted]

the maximum graphite moderator temperature. They do not worry too much about leaks in the heat exchanger even though the pressure of the CO_2 will be less than the water pressure. They will use moisture detectors with a sensitivity of < 0.01% H_2O and quick-acting valves (~20 sec closing).

WINDSCALE LABORATORIES - JULY 11

Participants - J. C. Bell and J. S. Nairn (Windscale)
W. J. Lindsey (Production Division, AEC)
R. E. Nightingale

The work on graphite-gas reactions at Windscale is largely in support of the Windscale and Calder Hall reactors. Mr. J. S. Nairn is in charge of the oxidation work. About eleven people are working in his group. The thermal-oxidation rate has been determined at two temperatures for Calder Grade A graphite in a static system. These and data under other conditions are presented in a report[22] for the forthcoming Geneva Conference.

Mr. Nairn stated that after two years of Calder operation there has not yet been any positive evidence of a back reaction $2\ CO = CO_2 + C$ except possibly at the higher CO concentrations recently attained in Calder I Reactor.

This conclusion was reached after: (a) experiments in which weighed and polished pieces of platinum, steel, and graphite placed in the main gas stream of the heat exchanger did not collect any carbon deposits after 21636 Mwd and 59,000 Mwd of Calder I operation; (b) parts of the reactor circuit examined after various periods revealed no evidence of carbon deposits; (c) the measured CO concentration in the coolant could be predicted quite satisfactorily from known initial oxidation rates (no back reaction) and estimated coolant leak rates.

During the first major shutdown of Calder I modifications were made which reduced the leak rate from ~ 2 tons/day to ~ 1 ton/day. The effect of reducing the leak rate was to increase the steady-state concentration of CO, but not quite to the extent expected if no back reaction had occurred. This, together with certain other observations, could be explained by a small back reaction at the higher CO concentrations (0.67 volume per cent). The extent of the back reaction is small under these conditions, however, and seems to justify no concern from an operational standpoint.

[22] J. Wright (Editor), Chemical Studies of Carbon Dioxide and Graphite Under Reactor Conditions, Paper 303 for the 1958 Geneva Conference (June 13, 1958).

[redacted]

[redacted]

Mr. Nairn was asked if any graphite oxidation limits had been considered for the Calder reactors, and if so, what they might be. He mentioned three factors which might limit the oxidation allowed: (1) loss of moderator strength, (2) loss of neutron moderation, (3) carbon mass transfer. Apparently no limits have yet been set but it is felt that at least 5% oxidation over the lifetime of the reactor would be easily tolerated. The present Calder rate is ~ 1/2 ton/yr based upon CO analyses or about (1/2)(1/600)(100) = 0.08%/yr. This would amount to a very nominal amount (1.6%) in 20 yr. Nairn has been unable to get an opinion from the physicists on the effect of loss in moderating power. He is not concerned with carbon transport under present operating conditions.

The oxidation rate of graphite samples placed in the coolant stream was found to be little different from those inserted into the bulk of the moderator. This indicates that one must be concerned with strength losses of the moderator itself, since reactions are not confined to the surfaces at these temperatures. Similar conclusions have been reached from experiments conducted at Hanford.

When the Calder Hall reactors were constructed, a large number of weighed graphite specimens were distributed throughout the moderator structure. This allows the monitoring of the weight loss of the moderator throughout the reactor lifetime. Data thus obtained will also be useful in predicting the behavior of graphite in more advanced reactors. This procedure should be followed in any GCPR constructed in the United States. Careful consideration should be given to the placement, size, and number of samples and methods by which they may be removed without invalidating weight losses by oxidation.

Results have been published which indicate that the thermal reaction rate is significantly reduced by treatment of the graphite with a small amount of phosphate. Nairn stated that such treatment is not effective in reducing the rate in the reactor. Apparently the phosphate, which may occupy active sites and reduce the thermal reaction, is changed by irradiation or active sites are rapidly created. Time was not available to discuss these experiments in detail, and he also seemed reluctant to do so. It would appear that further work should be done on the effect of irradiation on phosphates or other oxidation inhibitors. Nairn believes that there is some hope of getting a very pure graphite which will have a significantly lower oxidation rate. This belief stems from the rather large variations in thermal rate observed in graphites

[redacted]

[redacted]

with different purity. Thermal oxidation rates are always tested as part of the specifications of reactor graphite.

Nairn preferred to say nothing on their graphite coating work since it was scheduled for discussion at Harwell later in the trip.

The graphite-air burning apparatus described by Dr. Kronberger has been dismantled. Some graphite pieces which had been used were examined. The samples consisted of about a half-dozen graphite cylinders some 6 in. in diameter containing 15 to 20 small coolant channels. The cylinders were stacked to form continuous air channels some 3 ft in length. These were surrounded with heaters to maintain the graphite at the temperatures of the adjacent air. The conditions under which the graphite oxidation became self-sustaining were studied. The experimental results were not described in detail, but it was stated that they agreed very well with the model developed by W. B. Hall and J. Woodrow. A copy of their report is included in Appendix B from which the experimental conclusions may be deduced.

Radiation damage in graphite was discussed with Mr. J. C. Bell, who is in charge of the graphite radiation damage work. This is a group of some eight people. The author had an opportunity to visit the Windscale laboratories. New equipment for measuring stored energy has been built. A new calorimeter for measuring total stored energy has been constructed at Capenhurst and is scheduled to arrive at Windscale soon. The methods of measuring stored energy are very highly developed both at Windscale and Harwell. The "linear rise" apparatus which is programed for automatic temperature rise is a particularly nice instrument. Electrical circuits for the programs will be made available to any U. S. group interested in constructing such equipment.

The technical people at Windscale are still quite interested in the graphite sample exchange discussed during the UK/US Graphite Meeting in London in December, 1957. The UK would like to extend their studies to higher exposures on samples from Hanford. A request has been made through the Hanford Operations Office to send a series of samples to Windscale and the status of this request will be determined by the author. Such an exchange would also be useful in the Hanford graphite work since it would allow comparison with Windscale of the measurements of several physical properties.

[redacted]

[redacted]

[redacted]

HARWELL - JULY 14

Participants - J. F. Jackson, P. W. Mummery, J. H. W. Simmons
T. M. Fry, J. Wright – all of Harwell

M. T. Kavanagh - Risley

E. L. Brady - US AEC, London

W. J. Larkin, A. M. Perry, R. E. Nightingale – all of US

The proposed co-operative graphite program was discussed. The basis for the discussion was the letter included in Appendix C. As a matter of philosophy it was generally agreed that both the US and UK must proceed with their required programs without depending upon the other side for information. However, the free exchange of results should be encouraged. Determination of the scope and content of the graphite information exchange required more time than was available during this meeting. This will be explored further with the Harwell people by Dr. E. L. Brady of the London AEC office.

Mr. Wright briefly described the work of his group (~ 10 people) which is studying gas-graphite reactions under irradiation. A group of similar size, under the supervision of Mr. J. J. Hutcheon, is studying the gas-graphite system in the absence of irradiation. This work is generally considered fundamental in nature and many of the results could likely be made available to the US on an exchange basis. There appears to be no question but what the gas-graphite work at Harwell is now far more advanced than anywhere in the free world.

Work on the H_2-graphite reaction was described briefly. Methane is the main product. The thermal-reaction rate is comparable to the CO_2 graphite rate at some 100 – 1500°C higher temperature. The rate is enhanced by irradiation. The kinetics of the H_2-graphite system is somewhat different, however, in that only short-lived active species, presumably hydrogen atoms, are formed. In contrast to the CO_2-graphite reaction carried out in the same type of apparatus, reversing the flow in the quartz in-pile loop, did not change the reaction rate. Further details are available in a recent report.[23] There appear to be no effects found which would prevent the use of hydrogen as a reactor coolant from a graphite compatibility standpoint.

Other gases which show promise as reactor coolants will be studied at Harwell. Wright estimated that perhaps one gas per year would be studied in

[23] N. S. Corney and R. B. Thomas, The [redacted] Radiation on the Reaction between Hydrogen and Graphite, AERE C/R 2502 (June, 1955).

[redacted]

[redacted]

about the same details as was done wi[redacted] apparatus is now being tested for research on the helium-graphite system, and will test the possible formation of helides which could conceivably transport carbon in a flowing system. In-pile conditions will be simulated out of the pile by the use of a microwave generator. It will be possible to put more energy into the gas than could be done in the pile. If the active species formed do not transport carbon, it will be assumed that this will also not occur in the pile.

A small amount of work has been done with N_2. An experiment at 850°C in DIDO at 5 to 6 x 10^{13} nvt (thermal) showed no trace of cyanogen or other carbon-nitrogen compounds. A second experiment at 600°C also resulted in no carbon-nitrogen compounds, no carbon weight loss, and no carbon transfer after six weeks.

There was little discussion on graphite coatings. Some work has been reported in the Geneva Conference paper[24] and they were reluctant to say much more. In addition to the coating work at Harwell, work is also being done by Mr. O'Driscoll at Culcheth. It was the author's opinion from individual discussions with Kronberger and Nairn that there has as yet been no decision on whether to coat the Advanced Graphite Reactor (AGR) graphite. It is doubtful that they have yet developed a satisfactory coating. The decision on the AGR graphite will probably be deferred as long as possible.

Work was briefly described on annealing kinetics of graphite being conducted by a group of twelve people under the direction of Mr. J. H. W. Simmons. The annealing studies are largely concerned with stored energy release. An empirical equation has been used as the basis for correlating the experimental results:

where $ds/dt = f(s)e^{-A(s)/T}$

s = stored energy released to time, t

T = annealing temperature

A(s) = activation energy

In order to simplify the treatment, A(s) has been taken as a constant and seems to work satisfactorily for low annealing temperatures. This equation giving the rate of heat generation has been coupled with the heat transfer details and programed for an IBM 704 computer. The experimental data is being obtained from four adiabatic stored energy calorimeters. The program has been

[24] J. Wright (Editor), Chemical Studie[redacted] Dioxide and Graphite Under Reactor Conditions, Paper 303 for the 1958 Geneva Conference (June 13, 1958).

[redacted]

[redacted]

underway only about three months, so few results are available as yet. However, it appears that the program, plus the effort at the Windscale laboratories will result in a much better understanding of the stored energy release in graphite.

Because more information is needed on irradiation damage from 100° – 400°C, Simmon's group is building some controlled temperature equipment to irradiate samples in DIDO at these temperatures. These are somewhat similar to experiments about to start at Hanford. Information will be exchanged on the results.

No data were yet available on the Mark II Model channel experiment to be conducted at 500°C. A report[25] on the design of the original Model Channel (350°C) was provided and steps were taken to make a copy of the Mark II design report[26] available at an early date.

The author attended part of the discussions on philosophy and design of high-temperature, gas-cooled reactors. The Industrial Group at Risley is responsible for the AGR in which the CO_2 graphite system will be pushed to the temperature limit. Harwell has responsibility for the High-Temperature Gas-Cooled (HTGC) Reactor. This is to be a reactor experiment built on as small a scale as is possible, probably with a power rating of about 10 Mw. Both carbon and beryllium are being considered as the moderator. The present feeling seems to favor carbon. The coolant will be determined from the research work now being done. They feel helium could be used and no opinion has yet been formed on N_2. If beryllium is the moderator it may be possible to use CO_2 as the coolant. Coolant temperatures will be the order of 800°C. Experiments are underway to determine the graphite contraction and other effects from irradiation in the temperature range 900 – 1000°C.

Additional reports[27,28,29] of interest were made available to us.

---

[25] J. F. Ablitt, The CO_2-Cooled PIPPA Model Channel in BEPO, TRDC/P106 (July, 1955).

[26] Stephen, Mark II Design, POC Data 58 (1956).

[27] H. W. Davidson and H. H. W. Lasty, The Effect of Neutron Irradiation on the Mechanical Properties of Graphite, Paper 64 for the 1958 Geneva Conference.

[28] G. H. Kinchin, Stored Energy in BEPO Graphite, AERE R/M 169 (February, 1958).

[29] G. W. K. Ford, D. M. Harris, and D. Pantall, Gas Lubricated Self-Acting Journal Bearings, AERE ED/R 1662 (1955).

[redacted]

[redacted]

SACLAY (CEN) JULY 18

Participants - H. Hering, Chief of Physical Chemistry
P. Leveque, Assistant to Hering
A. Herpin, Solid State Physics
G. Perruo, Solid State Physics
G. Jehanno, X-ray Studies
F. M. Lang, Chemistry of Graphite
Mr. Darras, Corrosion and Chemistry
Jack Sutton, Radiation Chemistry
R. E. Nightingale, Hanford

H. Hering explained that the purpose of the Saclay Center of Nuclear Studies is to provide research and training services for the Commissariat A L'Energie Atomique. Employing about 2500 people, Saclay has become the most important of the three nuclear research centers, the other two being at Fontenay-aux-Roses and Grenoble.

Following these introductory remarks the discussion proceeded directly to more technical subjects. The French are concerned at the moment with stored energy in the G-1 pile. This is an air-cooled pile very similar to the Brookhaven reactor built primarily for plutonium production (~ 12 kgs Pu/yr). It has never been annealed. Present plans are to anneal the reactor sometime in September or October by heating air after the fuel has been discharged. They are concerned with the oxidation of the graphite during the anneal and need to know the temperature which the graphite will attain when the stored energy is released. Air flow through iron pipes has picked up rust and deposited it in the graphite channels. There is concern that the oxidation will be catalyzed with possible burning during the stored energy release. Samples from the moderator have been removed and stored energy releasable up to 400°C has been determined by dipping and differential thermal analysis techniques. The maximum stored energy they have found to date is 140 cal/g. Stored energy decreases from the air inlet side where the graphite temperature is about 50°C to the exit side where the graphite temperature is approximately 180°C. They estimate the temperature rise through the graphite from the channel to the center to be about 5°C.

Stored energy determined by differential thermal analysis technique has revealed a peak in the stored energy release curve at about 340°C. This was somewhat of a surprise to them although such a peak has also been found from studies at Hanford on stored energy and other properties. The stored energy

[redacted]

[redacted]

curves obtained at Saclay precipitated further discussion on the characteristics of stored energy release. The experiments on thermal-annealing of radiation damage from work at Hanford and other US sites were discussed.

The French have given a lot of thought to the prediction of graphite expansion from thermal expansion measurements. They have found that:

$$\frac{\Delta C_{th}}{\Delta C_i} = \frac{\Delta l_{th}}{\Delta l_i} = \text{a constant}$$

where

ΔC_{th} is the thermal expansion of the crystal (C_o)

ΔC_i is the irradiation damage expansion of the crystal (C_o)

Δl_{th} is the thermal expansion of the sample length

Δl_i is the irradiation damage expansion of the length.

The experience at Hanford has been that while this works reasonably well for a particular graphite, one cannot reliably predict length changes from low-temperature irradiation by a consideration of only thermal expansion on a number of different graphites.

Oxidation work was discussed with F. M. Lang. It appears that they are now most interested in the oxygen reaction. Attempts to reduce the oxidation rate by a treatment with phosphates (organic, inorganic, P_4O_{10}, etc.) have not been too successful. The rate with oxygen is only reduced by a factor of 2. More success has been reported by the British with the CO_2 reaction in reducing the rate by such treatments. The French have apparently done very little work on coatings for graphite as yet, but apparently are quite interested in this work.

Sutton has started some oxidation experiments with carbon dioxide. These have included capsule experiments and circulating experiments. Irradiation was accomplished in a "converter," an irradiation facility next to fuel placed in the reflector. The equilibrium concentration of carbon monoxide in the capsules at 400 – 450°C was found to be 1/2%. In their recirculating experiment CO concentrations on the order of hundredths of per cent were found. They have no explanation for such a large difference but concluded from this that more flow experiments are needed to supplement the capsule irradiations.

[redacted]

[redacted]

Part of the afternoon was spent touring the EL-2 and EL-3 reactors with Mr. Guy H. Berges. The EL-3 reactor appears to be a useful test reactor in that convenient access to the core is available through vertical experimental holes. The graphite work has suffered from lack of adequate irradiation facilities in the past. The EL-3 should partially correct this problem.

In summary, it would appear that graphite radiation damage studies in France, while currently not developed to the extent found in the US or UK, will make significant contributions in the near future. The French appear to be considerably behind the UK as far as CO_2-graphite reactions research is concerned, but interest in this work will certainly result in an increased effort soon.

[redacted]

[redacted]

APPENDIX A

ORGANIZATION OF THE METALLURGY DEPARTMENT AT HARWELL

H. M. Finniston, Director
A. H. Cottrell, Deputy Director

Applied Physics - G. E. Bacon
C. S. Lees - Nondestructive Testing

Chemical Metallurgy - P. Murray
J. Williams - Fabrication Development
Harry Lloyd - Fuel Element Fabrication

Reactor Metallurgy - S. F. Pugh
M. B. Waldron - Plutonium Metallurgy
G. W. Greenwood - Irradiation Section
G. W. Horsley - Liquid Metal Work
G. H. Broomfield - Slurry Development

Metal Physics - A. D. LaClaire
Robert Barnes - Solid State Studies with A. H. Cottrell

It has been stated that when A. H. Cottrell goes to Cambridge University, P. Murray will become the Deputy Director. J. Theulis, who was the Assistant Director, has moved to the Central Administrative Group. Mr. B. R. T. Frost, who was in charge of all liquid metal work, is now in charge of Metallurgical Studies of the Controlled Thermonuclear Program. Mr. J. A. Robertson, who was in charge of the Irradiation Section, is now at Chalk River.

[redacted]

[redacted]

APPENDIX B

OXIDATION OF GRAPHITE IN AN AIR STREAM

W. B. HALL and J. WOODROW

(a) Conditions under which oxidation process becomes unstable

The heat generation due to oxidation may be expressed as

$$H = V.a\ e^{-b/\theta} \quad \text{cal sec}^{-1} \qquad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(1)$$

and the heat transfer to the air coolant as

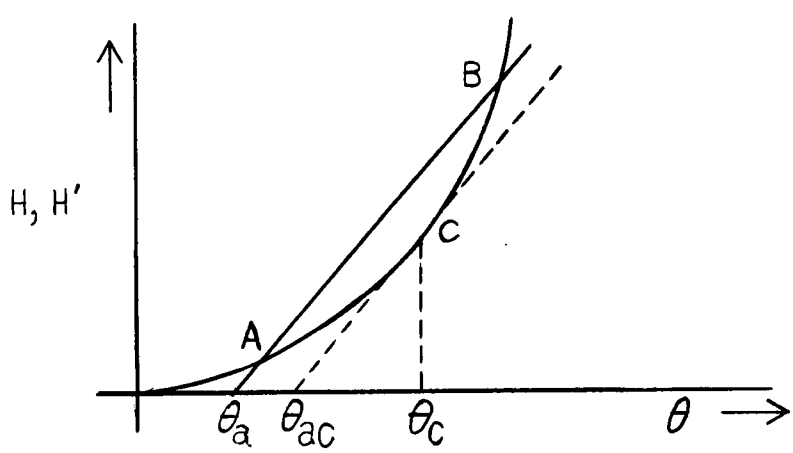
$$H' = Sh(\theta - \theta_a) \quad \text{cal/sec} \qquad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(2)$$

where V = volume of graphite undergoing oxidation (cm^3)
S = surface area for heat transfer (cm^2)
h = heat transfer coefficient from graphite to air cal sec^{-1} cm^{-2} $°C^{-1}$
θ = graphite temperature °C
θ_a = air temperature °C
a, b are constants defining the combustion process.

For the graphite and air to be in equilibrium, $H = H'$. For this state of equilibrium to be stable, $dH^2/d\theta > dH/d\theta$, so that a small increase in graphite temperature increases the heat loss by a greater amount than it increases the heat generation due to oxidation.

i. e. for stability $\quad Sh > V\dfrac{ab}{\theta^2}\ e^{-b/\theta}$

Equations (1) and (2) are illustrated in the following sketch. Two points of equilibrium are shown, point A being stable, and point B unstable.



[redacted]

[redacted]

(b) Oxidation of a graphite channel carrying an air flow

Provided that the channel is long enough, the air will reach such a temperature that the oxidation process becomes unstable. If, in the above diagram, θ_a represents the temperature of the air at inlet to the channel, the corresponding graphite temperature and heat generation will be given by point A. The air temperature will increase along the channel until the line defining the heat transfer from the graphite is in the position shown by the dotted line. Any further increase in air temperature would result in a situation where the increased heat generation could not be matched by greater cooling, and the graphite temperature would rise uncontrollably.

The length of channel required to raise the graphite temperature from its value of θ, at the inlet to the critical value θ_c may be determined as follows: -

The air temperature θ_a at any point along the channel is given by

$$wc\,\frac{d\theta_a}{dx} = Aae^{-b/\theta} \qquad \text{.....(3)}$$

and θ_a and θ are related by

$$\theta - \theta_a = \frac{Aae^{-b/\theta}}{h.p} \qquad \text{.....(4)}$$

where w = mass flow of air along channel
c = specific heat of air
x = distance along channel from inlet
A = cross sectional area of graphite associated with channel
p = perimeter of channel

Eliminating θ_a between (3) and (4), we have:-

$$\left[e^{b/\theta} - \frac{Aab}{h.p} \cdot \frac{1}{\theta^2}\right] d\theta = \frac{Aa}{wc}\,dx \qquad \text{.....(5)}$$

Defining the critical graphite temperature θ_c by

$$e^{b/\theta_c} = \frac{Aab}{hp}\,\frac{1}{\theta_c^2}$$

and the distance along the channel at which this condition occurs as x_c, the distance $(x_c - x_1)$ which is required in order to produce unstable conditions with a graphite temperature of θ, at the channel inlet is given by:-

[redacted]

$$x_c - x_1 = \frac{wc}{Aa} \int_{\theta_1}^{\theta_c} \left[e^{b/\theta} - \frac{Aab}{h.p.} \frac{1}{\theta^2} \right] d\theta \qquad \ldots\ldots\ldots\ldots\ldots\ldots\ldots(6)$$

(c) <u>Particular case</u>

$w = 25$ gms/sec.
$h = 5 \times 10^{-4}$ cal sec^{-1} cm^{-2} °C^{-1}
$A = 360$ cm^2
$p = 32$ cm
$c = 0.24$ cal gm^{-1} °C^{-1}
$a = 2.55 \times 10^{1}$ cal sec^{-1} cm^{-3}) data provided by J. Wright
$b = 2.00 \times 10^{4}$ °K) for irradiated graphite.

$$\frac{Aab}{h.p.} = 1.143 \times 10^{19}$$

$$e^{\frac{2 \times 10^4}{Sc}} = 1.143 \times 10^{19} \times \frac{1}{\theta_c}$$

$$\theta_c = 655°K \quad (382°C)$$

The corresponding air temperature is given by

$$\theta_c - \theta_{ac} = \frac{Aae^{-b/\theta}}{h.p.}$$

where $\frac{Aa}{hp} = 0.572 \times 10^{15}$

whence $\theta_{ac} = 623°K \quad (350°C)$

The following relationship between graphite temperature at inlet to channel θ, and the distance along the channel after which conditions become unstable is then found from (6)

θ_1 °K	655	650	640	630	620	610
$x_c - x_1$ (cm)	0	9	73	238	581	1244

(d) <u>Maximum temperature which the graphite may be allowed to achieve before full coolant flow is inadequate</u>

Suppose that the graphite has reached the unstable condition, and its temperature is rising with time. What temperature can it be allowed to reach before the full flow of coolant is inadequate to check the temperature

[redacted]

rise and return the graphite to stable conditions? The following problem can be formulated in order to clarify this point. Suppose the graphite is at a uniform temperature (above the critical value) and that the full coolant flow is then switched on, the temperature of the air at inlet to the channel remaining constant (at about 20°C). The following equations represent the problem:-

Graphite

$$A\rho c_p \frac{\partial\theta}{\partial t} + ph(\theta - \theta_a) = Aae^{-b/\theta}$$

Air

$$wc \frac{\partial\theta_a}{\partial x} = ph(\theta - \theta_a)$$

Eliminating, θ_a, we have

$$\frac{A\rho c_p}{ph} \frac{\partial^2\theta}{\partial x \partial t} + \frac{A\rho c_p}{wc} \frac{\partial\theta}{\partial t} + \left(1 - \frac{Aab}{ph} \frac{e^{-b/\theta}}{\theta^2}\right) \frac{d\theta}{dx} = \frac{Aae^{-b/\theta}}{wc} \quad \ldots\ldots\ldots\ldots\ldots(7)$$

where ρ = density of graphite
c_p = specific heat of graphite
t = time

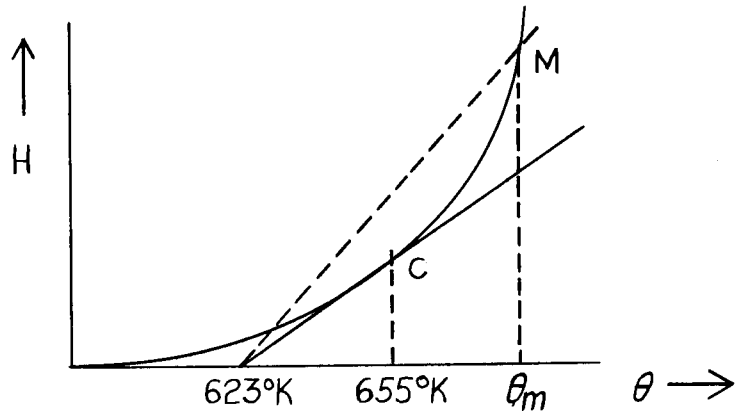
the other quantities being as in (b) above.

Simplified problem

Suppose that the graphite has become unstable at the end of the channel. The air temperature remains constant and the graphite temperature rises with time. Now suppose we merely increase the heat transfer coefficient between graphite and air: (in practice, switching on the main blowers would decrease the air temperature at the end of the channel as well as increasing the heat transfer coefficient).

[redacted]

The state of affairs is shown in the following diagram.



The point at which the graphite becomes unstable is denoted by C; the graphite temperature then follows the curve towards point M. Now, if we increase the heat transfer coefficient keeping the air temperature constant, the cooling is as shown by the dotted straight line rather than the full straight line. The graphite temperature must not be allowed to reach the intersection of this dotted line with the heat generation curve (point M). Using a value of $h = 4.3 \times 10^3$ (appropriate to full coolant flow) the value of θ_m is 728°K (455°C).

It is highly likely that this temperature can be increased when the solution of equation 7 has been obtained.

[redacted]

[redacted]

John A. Hall, Director
Division of International Affairs

W. Kenneth Davis, Director
Division of Reactor Development

COOPERATIVE GRAPHITE PROGRAM WITH THE U. K.

During the recent US/UK Graphite Conference held in London, December 16 – 19, 1957, the desirability of further sharing of technical information, exchange of irradiated graphite samples and other cooperative activities involving graphite were discussed among the technical representatives.

This was followed by a proposal dated January 20, from B. F. J. Schonland, Deputy Director of Harwell, to E. L. Brady, USAEC Scientific Representative, requesting the exploration of the feasibility of a joint research program involving a considerable amount of irradiation of graphite specimens in high fast flux U. S. reactors. This proposal, as well as other areas in which US/UK cooperative graphite programs might be undertaken, was discussed with representatives of the Divisions of Production, Research, and Reactor Development. Following this discussion, in which it was agreed that there were areas in which a joint program on graphite would be of mutual value, the Division representatives prepared statements amplifying their interests. These interests are incorporated in this memorandum.

The following cooperative graphite program is recommended to be of the greatest value to the U. S. by the Divisions of Research, Production, and Reactor Development.

1. Fundamental research

   a. Basic mechanisms of radiation damage, energy storage stored energy release.

   b. Basic mechanisms of gas-graphite reactions.

   c. Thermal and radiation annealing processes, repeated annealings, high temperature energy storage under conditions of frequent low temperature annealing.

   d. Physical properties of irradiated graphite such as thermal and electrical conductivity, hardness, strength, crystalite and dimensional variations.

   e. Gas-graphite reactions involving CO_2, N_2, H_2, and other potential atmosphere or reactor coolant gases or mixtures, thereof. These would include studies in the presence of reactor radiations with adequate provision for control measurements. Also, the effects of impurities in the gases and the graphite as well as the effect of cladding materials on the graphite

[redacted]

[redacted]

2. Technical information [redacted]

   a. Operating experience - on all graphite reactors where information pertains to reactor safety.

   b. Methods for measuring stored energy.

   c. Methods of measuring gas-graphite reactions.

   d. Stored energy releases.

   e. Prevention and control of fires in graphite reactors.

   f. Pyrophoric properties of irradiated uranium and graphite.

3. Exchange of technical information

   a. Publications

   b. Periodic technical meetings, symposia

   c. Laboratory visits, as appropriate, by technical and scientific personnel.

4. Exchange of samples of unirradiated and irradiated graphite

   a. Exchange of experimental data and interpretations of measurements made with these samples (covering radiation damage, stored energy, and gas-graphite reactions).

5. Sharing of specialized facilities

   a. U. S. high flux reactor irradiations as proposed by Dr. Schonland.

   b. Gas-graphite oxidation loops in U. K. reactors.

The cooperative program should include studies covering a broad range of temperatures, including values at least as high as 800°C, high neutron fluxes, greater than 10^{14}nv, and exposures exceeding 10^{22}nvt.

Maximum benefits should derive to both countries if free and unrestricted exchanges of information are permitted. By treating graphite as a material in the manner common to scientists, this free exchange should be possible in all areas. The areas of conflict involving the exchange of information, relating to U. K. graphite power and production and U. S. graphite production reactors, are primarily those of design, specifications, operating conditions and performance characteristics. The broad coverage of the proposed cooperative program will reduce these areas of conflict and, as the basic mechanisms become better understood and the gaps in scientific and technical knowledge

[redacted]

[redacted]

are closed, there will be no need to refer to the actual conditions of these reactors for information. The initiation and conduct of the recommended cooperative program does not directly involve operating and performance specifications of them involved. Hence, exchanges considered restricted to these reactor types may be treated by separate agreements.

* * * * * *

[redacted]