Attachment A

Return with Camera-Ready Manuscript

Cover Information for ASM Technical Papers
(For inclusion in the ASM Proceedings)

In order to ensure that the correct title, author names, and affiliations appear on the cover and title page of your paper, please provide the information requested below.

Type the exact title of the paper, the author(s), and affiliation(s)—include city and state—in upper and lower case (For example: New High-Temperature Alloys, J. Jones, ABC Inc., Cleveland, OH). ASM will have the copy set in bold headline type for placement on the first page of the paper. Do not include author's title (Prof., Dr.), position (president, research engineer), or degrees (Ph.D., M.E.).

Paper Title (upper and lower cases)

Metallography at the Met Lab - the First Fifty Years

Author(s) (upper and lower case)	Affiliation(s) (city & state) (upper and lower case)
1. Richard H. Lee	1. Energy Technology Div., Argonne Natl. Lab. 9700 South Cass Ave., Argonne, IL 60439
2.	2.
3.	3.
4.	4.
5.	5.

Conference: 28th Annual IMS Convention, July 23-26, 1995, Albuquerque, NM

Note: Materials submitted for publication will not be returned unless requested by the author.

Attachment B

Part A - Copyright Transfer

Conference: 28th Annual IMS Convention, July 23-26, 1995, Albuquerque, NM
Title Date

Manuscript Title: Metallography at the Met Lab - the First Fifty Years

Author(s): Richard H. Lee

Copyright, title interest and all rights to the Article are hereby assigned and transferred to ASM International, Materials Park, Ohio 44073. U.S. Government employees whose work is not subject to copyright may certify such employment status in Part B below.

However, the author(s) reserves the following:

1. All proprietary rights other than copyright, such as patent rights.
2. The right to make oral presentation of the same material in any form.
3. The right to reproduce figures and extracts from the Article with proper acknowledgment.

In the case of work performed under U.S. Government contract, ASM International grants the U.S. Government royalty-free permission to reproduce all or portions of the Article and to authorize others to do so for U.S. Government purposes.

Author's Signature	Print Name	Date
Richard H. Lee	Richard H. Lee	10/10/95

In the case of "work made for hire," (a work prepared by an employee within the scope of his or her employment or commissioned as a work for hire by written agreement), an authorized representative of the employer should sign below.

Signature and Position	Print Name	Date

Part B - U.S. Government Employee Certification

I certify that all authors of the article [illegible]nt and performed this work as part of their employment; therefore, the Article i[illegible]otection.

The submitted manuscript has been authored by a contractor of the U. S. Government under contract No. W-31-109-ENG-38. Accordingly, the U. S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U. S. Government purposes.

Authorized Signature	Title
Name of Government Organization	Date

This transfer document must be signed by each author unless excepted above and be received by the Confernece Office before the Article can be published. Please mail the original of the document to the following address:
Conference Administrator; ASM International; Materials Park, Ohio 44073

METALLOGRAPHY AT THE MET LAB - THE FIRST FIFTY YEARS

Richard H. Lee
Energy Technology Division
Argonne National Laboratory
Argonne, IL 60439

Abstract

The Met Lab at the University of Chicago was established to build the world's first nuclear reactor. The object was to see if a pile (CP-1) could be built to create a sustained chain reaction, i.e., controlled nuclear fission. New materials of the very best quality were needed and people of many skills worked together to achieve the goal as quickly as possible. This is the story of a select group of people who were scientific and engineering pioneers in this new field. Research continued at new sites on more advanced reactors and cooling systems. Many problems were encountered in the fabrication of reactor components, and metallography was a crucial method of analyzing the reactions and quality of consolidation. 1996 will be the 50th anniversary of the beginning of the National Laboratories, so it is appropriate to commemorate and recall some pioneering achievements.

MANY DEVELOPMENTAL DISCOVERIES were made in the days when the first nuclear reactor was being built at the University of Chicago. Several examples are recalled by the people who were there and who pioneered the development and fabrication of the necessary new materials, many of which were not available in fabricated form. Because of neutron absorption, the reactor materials had to be extremely pure, and the fabrication procedures were experimental. New fissile materials, such as U_{235} and Pu 238,239 were discovered to be the desired materials for compact uncontrolled fission devices (bombs). Shortly after the first controlled fission experiment at the University of Chicago, a decision was made to produce the fissionable material elsewhere, and work on the military project was largely moved to Los Alamos, New Mexico. The focus of this presentation is the people (and their experiences) who were either at the first reactor in Chicago or at Argonne National Laboratory (ANL), beginning in 1946.

New Materials

The new materials included carbon (graphite), aluminum, uranium, plutonium, zirconium, and their alloys. Early in 1942, Arthur Compton of Columbia University was placed in charge of the graphite-uranium lattice reactor concept, and the Columbia and Princeton research groups were transferred to the University of Chicago where the "Metallurgical Laboratory" (Met Lab.) was established. This was a code name for this military project, part of the "Manhattan Project".

Impurities in the uranium and the graphite moderator would capture neutrons and make them unavailable for subsequent reactions, and these reduce the neutron reproduction factor (called "k"). To obtain a reproduction factor >1.0, maximum purity would be required of the pile materials (1).

Three manufacturers began supplying higher-purity uranium in November 1942, in cooperation with Frank Spedding of Iowa State University in Ames. Westinghouse, Metal Hydrides Company, and Mallinckrodt in St. Louis supplied up to 30 tons per month of refined metal and oxide. National Carbon Company produced improved graphite by doubling the furnace time in converting coke. In the same month, construction of the main pile began under the west stands of the University's Stagg Field. Until this time, uranium was only an obscure curiosity. "Never has an industrial material been made this pure" we were told (2).

Graphite was machined into blocks under the direction of Walter Zinn, who had six young physicists and an able carpenter to do the work. Eventually, 45,000 blocks were made for the pile, each weighing 8.6 kg. The uranium was inserted as 5636 kg of metal and 36,632 kg of oxide into the lattice. Enrico Fermi himself helped in the machining of graphite, and one day was observed to appear like a coal

Fig. 1 - Artist's conception of CP-1.

miner covered completely with fine black dust and perspiring profusely.

Uranium ore was immediately available because Kenneth Nichols told General Groves about a recent discovery of ≈1250 tons of pitchblende concentrate in the Belgian Congo. From there, it had been secretly shipped by the French to the U.S. to prevent it from falling into German hands (3).

Overview of Metallography

Metallography evolved at the Met Lab and ANL in support of the reactor development programs. We decided that reliability, quality, and operability were essential for long-term stability. Several classes of new materials were studied and developed for fuels - U,Th, and Pu; moderators - C, Be, and water; control rods - Cd, B, and Hf; structural cladding - Al, Zr, and stainless steel; and (5) structural containment - steel and concrete. These materials would have to function in hostile environments, i.e., under conditions of corrosion, high temperature, and radiation damage. One of the first surprises was the embrittlement of pressure vessel steels. In fabricating reactor materials, researchers learned that impurities had large effects and that stable fuels were needed. The latter need led to alloyed uranium and plutonium fuels. The philosophy with respect to weld quality was that we needed zero defects and had to use the best tools available, i.e., corrosion testing, nondestructive testing, and metallography, to secure this need. Simulated welds and routine metallographic examinations became standard procedure.

Fabrication Research

ANL engineer David Walker relates his experience with materials, beginning in 1946, when he first came to "Site B" to work on ways of protecting uranium from oxidation, e.g., by electroplating. Protection by plating was not sucessful, but a bright electropolish solution and a procedure to bring out the macro grain structure were developed. Being able to see the crystal structure gave a big boost to the researchers who were trying to produce single crystals of uranium. It allowed them to determine the critical strain required for recrystallization and grain growth. Aluminum was the first material used for "canning" the uranium fuel, but it had a relatively high reactivity both to the fuels and cooling water. So, while research continued on aluminum, better materials were evaluated, mainly by corrosion testing in pressurized water autoclaves. Preliminary tests showed that pure zirconium had excellent resistance to high-temperature water and desirable (low) neutron absorption. Researchers at Oak Ridge National Laboratory had found zirconium to have a high neutron capture cross section, but that was because it contained hafnium as a major impurity (4).

Aluminum was easy to fabricate, but difficult to bond to the uranium fuel rods. No zirconium tubing was available, in fact zirconium had only recently been produced at the Bureau of Mines by a new reduction process. One of the first fabrication problems was to make some zirconium tubing for cladding. A decision was made to try to make tubing by rolling strips of sheet and then tungsten-inert-gas weld a seam. Samples of the welded tubing were then sectioned, mounted, and polished for examinations of weld quality.

Because it was difficult to polish and etch, ANL staff consulted with others who had experience with it, and learned to give it a good strong etch to bring out the grain structure. The properties of zirconium are similar to those of titanium, except that it has better corrosion resistance and therefore requires an etch with more oxidizer (nitric acid) and a strong reducing agent (hydrofluoric acid).

Later, with ANL cooperation, seamless zirconium tubing was made by commercial fabricators Trent Tube and Wolverine Tube. The first material made by the Bureau of Mines was like spring steel and so tough it was called "Hoover Plate." Later, in the 1950s, crystal bar material became available from Foote Mineral Company; this material was much easier to work because of its higher purity. Because of the increasing demand of the Naval Reactor Program, ANL personnel showed commercial fabricators how to extrude zirconium tubing, including a process for copper-coating the blanks for easier drawing (4).

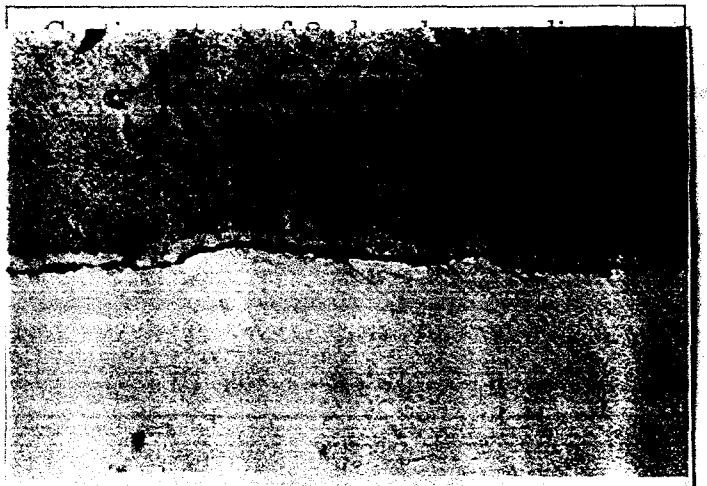
Uranium was another metallic material that required extensive developmental work. Uranium was so rare at that time that it was literally worth 100 times its weight in gold. Because researchers had little experience with it, one of the first problems was reducing it to a size for fuel rods. The first ingot was placed in a furnace and allowed to heat to soften it for extrusion. When the furnace was turned off, the billet was rapidly extruded and the mechanical energy was converted to thermal energy, nearly melting the uranium. As it came out of the press, it reacted with air and burned like fireworks. Melting and casting was initially performed in graphite molds, which caused about 10 at.% pickup of carbon and resulted in a hard, fibrous structure that was revealed by etching (5).

At ANL site B, an old brewery, the properties of uranium were studied and it was discovered that with thermal cycling, it grew in length. ANL assistant Andy Van Echo found that highly oriented uranium could grow to 10 times its original length by repeated thermal cycling through the transformation temperature range. ANL first developed the beta heat treatment to stabilize the alpha growth and realized texture was very important (6,8).

The first slugs of uranium for the Savannah River reactor were canned in aluminum by a novel method called "submarine style," which was developed by Frank Yaggee. To bond the aluminum cladding to the uranium fuel, a silicon-aluminum eutectic alloy was used. The uranium metal was dipped into the molten alloy to form a thin reaction layer and then the aluminum can was quickly dipped over the fuel and the assembly was removed to cool. ANL applied for a patent on the process, but at a meeting at Hanford shortly thereafter, they kept such good notes that they also applied for and received the patent (6). ANL later transferred the technology to DuPont and became the center for reactor development in the United States. ANL also formed an international school to teach nuclear reactor metallurgy (7,9).

For the first experimental breeder reactor, EBR-1, in 1949, uranium-melting safety was uncertain because the critical-mass properties were based only on calculations. Frank Yaggee, a metallurgist assigned to the casting project, carefully made the calculations and to prepared for melt and cast the uranium ingots for extrusion. When casting, the volume is usually reduced and care must be taken not to approach a mass that would become critical, resulting in possible death of personnel. Frank melted the required uranium and when the time came for casting, to his surprise, everyone else in the room and vicinity had disappeared. So Frank proceeded to cast the liquid uranium alone, without incident. For EBR 2, in 1959, the uranium was made to fill the quartz molds by lowering the molds into the molten metal in a vacuum furnace and then simply pressurizing the chamber with inert gas, forcing the metal to fill and freeze in the molds (9).

Worried about self-welding of reactor components at high temperatures, Lowell Lloyd and Howard Kittel studied diffusion between potential construction materials in 1947. Metallography was essential to determine the extent of reaction between materials and the formed phases. From the experimental data , reaction rate constants could be calculated at various temperatures. Figure 2 shows a typical diffusion reaction between U&Zr materials after polishing the cross section and etching. This data showed which materials were compatible and what phases would form, in a manner similar to that used in phase diagram studies (10).



No. 5510 1000x

Fig. 2 - Diffusion-experiment reaction layers between U-Zr after six days at 700C.

Fabricating Fuel Plates and Components

Several early reactor designs required fuel components to operate at higher temperatures to produce power. Two fuels, metal and oxide, were tried. Two coolants water and liquid metals, were selected. At site B, a liquid metals group was started under Larry Kelman to investigate the heat transfer and corrosion properties of sodium, lithium, and the Na-K eutectic. The NaK was also used to bond the metal fuel to steel cladding and metallographically it was observed that the NaK dissolved the carbon from steel (12). The first reactors for the Navy submarine were designed at ANL with Captain Hyman Rickover and two competing contractors. Westinghouse selected the water-cooled design and General Electric worked on the liquid metal design. The reactor that was selected, a pressurized water reactor, used high-pressure water as the coolant. The high pressure was thought to be absolutely necessary because the belief was that if water was allowed to boil off the fuel plates, cooling would be reduced, resulting in possible melting. General Electric began building boiling water reactors because ANL, under the leadership of Samuel Untermeyer, showed that the water coolant did not required pressurization. In fact, when boiling starts, reactivity decreases and the reactor is self-regulating. Untermeyer reasoned that, for the reactor to function, it must have a water moderator and when water changes to steam its density approaches zero and its moderation is gone. A series of so-called BORAX experiments was conducted by ANL in

Idaho in the 1950s. In these experiments, U-Th mixed oxide fuel was used.

The EBR-1 was also designed to see if fuel could be created faster by neutron irradiation of natural uranium. We later designed a special core to join cladding to the fuel by diffusion bonding. The EBR-1, which had a novel cam locking design to prevent movement of core assemblies, was made from the very first heat of "reactor-grade" stainless steel. Crucible Steel Company of America made a 20-ton heat of Type 347 stainless steel that was used to make all of the components of the core. This allowed assessment of radiation damage without trying to factor in effects of steel chemistry (9,11).

The "Argonaut " reactors (zero power) for teaching experiments were designed with pure aluminum cladding and an aluminum-uranium oxide (U_3O_8) fuel. They were extruded to flat plate with the final cladding thickness of only 2.5^{-4} to 1.25^{-3} mm (0.001-.005 in.). After extrusion, the density of the fuel was nearly 100% of theoretical and the oxide fuel did not react much with the pure aluminum because the aluminum formed a thin protective oxide, Al_2O_3, that was in contact with UO_2. The oxidation/reduction process then stopped. Oak Ridge National Laboratory used a hot roll-bonding process with Al-UO_2 fuel and experienced fuel reactions that formed blisters in the plates. Metallography also revealed perforated interfaces during operation (9).

The coating and jacketing group at ANL under Bob Noland also developed the world's first automatic TIG-welder (tungsten-inert-gas) with controlled current, RPM, and time of welding. Cecil Stone and David Walker developed these welders, as well as a pulse-bonding technique to join uranium fuel rods to the cladding with 1.25^{-3} mm Zr foils. The radio-frequency (RF) coil and inert gas made a perfect bond in seconds, making fuel rods that were much stronger.

Distant early warning (DEW-line) reactors were needed for independent and reliable sources of power and heat and ANL was asked to contribute a design: commercial fabricators were asked for fuel plates but no one could fabricate them successfully, because the plates would be subject to blistering. Nelson Beck developed a nondestructive test called "ultrasonic through transmission." With this method, the signal (indicating a good bond) would be picked up only if the ultrasonic wave passed through the assembly. It was superior to visual examination for blisters, which could miss some defects.

ANL also developed a unique method of bonding aluminum cladding to U-Al alloy fuel; it was called silicon pressure bonding of aluminum. The U-Al fuel was assembled into an aluminum "picture frame" and the top and bottom Al cladding plates were covered by a thin layer of pure silicon before being sandwiched together with the U-Al fuel in the aluminum frame. Under elevated pressure and temperature, a eutectic alloy formed, instantly melting and "sweeping" out interfacial oxides. The bond was so good (and thin) that it could not be seen with conventional metallography (9).

Argonne also witnessed the world's first (known) reactor core meltdown of EBR-1 at Idaho in 1955. The fully enriched core was designed to test and determine the reason for a positive temperature coefficient. When the temperature went up, neutron reactivity would also go up. It was thought that the core was bowing inward when more power was extracted, thus making the core have a smaller volume than normal and causing excess reactivity. A new core was designed that would not allow any bowing or change in volume. It was with this core that power excursions were run by deliberately pulling out the control rods and to see how fast it would go. Unfortunately, one test went so fast it could not be stopped, resulting in melting of the small core assembly. Metallography was used extensively to determine what had occurred in the fuel and analyze the temperatures that were encountered. Figure 3 is one example (10,13).

Researchers at ANL pioneered the technology of remote control metallography with steel-filled cement block "caves" to learn remote handling. Then building 301 was built to do the metallography of the EBR-1 and Fermi-reactor-melted cores. Later, an alpha-gamma (up to 10^{+6} Curie)-shielded hot cell was built in Building 212.

Subsequently, a second breeder reactor (EBR-2) was built, and a new fuel based on used fuel reprocessing was developed; U-Pu-Fs (uranium-plutonium-"fissium"). Researchers at ANL decided to use only inert-gas glove boxes for plutonium metallurgy and developed the first techniques for plastic pouching to transfer such materials. Large pouches (300 mm) were needed, but large dielectric sealers were not available. One day, a janitor was watching the experimenters sealing pouches and suggested that they simply overlap the heat seals. This turned out to be an easy and efficient method (14).

Fig. 3 - EBR-1 core-melting photomicrograph of U-Fe eutectic phases.

Other pioneering work was done at ANL, such as vanadium alloys for sodium-cooled reactors, but that is a subject for a future publication.

Acknowledgment

I would like to thank the retired ANL staff: William Sturm, Larry Kelman, Jim Schumar, Frank Karasek, David Walker, Howard Kittel, and Howard Rhude for their cooperation. We have only scratched the surface of their memories. I regret the lack of figures because nearly all of the early photomicrographs have been discarded.

References

1. "The First Reactor," DOE/NE 0046, 40th anniversary publication, NTIS, Springfield VA, 1982.
2. Private communication, W. Sturm, Argonne National Laboratory, 1990.
3. Rhodes, R., The Making of the Atomic Bomb, Simon and Schuster, New York, 1986.
4. Private communication, F. Karasek, LaGrange IL, 1995.
5. Private communication, J. Schumar, Hinsdale, IL, 1995.
6. Foote, F. G., Early Uranium Metallurgy in Chicago, J. Nucl. Mat. 100, North Holland, Amsterdam, 1981.
7. Wilkinson, W. D., and W. F., Murphy, Nuclear Reactor Metallurgy, Van Nostrand, Princeton, NJ, 1958.
8. Private communication, L. Kelman, Argonne National Laboratory, 1989.
9. Private communication, D. Walker, Argonne National Laboratory, 1995.
10. Kittel, J. H., Layer Formation by Interdiffusion Between Some Reactor Construction Materials, Argonne National Laboratory Report, ANL-4937, 1949.
11. Noland, R. A., and D. E. Walker, Special Heat of Austenitic Stainless Steel Used in the Experimental Breeder Reactor (EBR-1), Argonne National Laboratory Report ANL-5548, 1956.
12. Kittel, J .H., M. Novick, and R. F. Buchanan, The EBR-1 Meltdown - Physical and Metallurgical Changes in the Core, Argonne National Laboratory Report ANL-5731, 1957.
13. Schumar, J. F., The Development of Uranium Fabrication Techniques for Plutonium Production Piles, J. Nuclear Materials 100, North Holland, Amsterdam, 1981.
14. Private communication, H. Rhude, Tinley Park, IL, 1995.

DISCLAIMER