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RADIATION EFFECTS OF
HIGH-ENERGY ELECTRONS

by

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ABSTRACT

Six electron-irradiation experiments are described. In one, copper was irradiated with 1.25-Mev electrons at about 80° K. Electrical resistivity, measured as a function of amount of exposure and after annealing at temperatures above the irradiation temperature, indicates the extent of radiation damage and annealing recovery. A similar irradiation is in progress using nickel. As the result of a third study, the threshold displacement energy for nickel is determined to be 34.5 ± 1 ev near 4.2° K. An irradiation of coldworked copper at temperatures between 100 and 150° C is described in which the electrical resistivity decreased as a result of "radiation annealing". In a fifth experiment, Young's modulus is found to increase rapidly as a function of electron irradiation. The irradiation of graphite with 1.25-Mev electrons is also described. Here it is seen that no annealing effect occurs below 80° K.

Another series of experiments are reported, involving radiation and pyrolytic stability of terphenyls and polyphenyls. It is concluded that para-terphenyl is the most stable in both respects and that addition of or radiation-induced generation of minor quantities of the most radiation-stable compounds may yield a mixture of polyphenyls with enhanced stability. Reported decomposition rates for terphenyls are the lowest reported in the literature for any organic compound under comparable conditions and suggest high-temperature applications in radiation fields.

INTRODUCTION

The present major effort at Atomics International involves the construction and operation of the SRE and OMRE reactors as part of the Atomic Energy Commission reactor development program. An important part of the operation of these reactors will include radiation damage investigations of the fuel, moderators and structural materials.

In order to aid in the general problem of understanding radiation effects in materials, two groups in this laboratory have made radiation damage studies during the past several years in graphite, metals and possible organic moderators with the general goal of helping to build that science which will permit selection of the optimum materials of reactor construction. This report describes the most recent work of the solid state physics and radiation chemistry groups in which high energy electrons were used as a source of radiation. The electrons, obtained from a Van de Graaf machine, have the advantage over other types of radiation in that the energy of the bombarding particles can be more accurately controlled and measured. They produce in some cases slightly different types of damage in materials and much can be learned from observing and understanding these differences. A particular effort is being made to identify the crystal defects produced in solids by radiation and to gain an insight into such defects' behavior and their relationship to physical properties. In the possible organic moderators the goal is to determine thermal and radiation decomposition rates and the decomposition products.

SOLID-STATE PHYSICS

The six solid-state studies to be described have this in common: each made use of electron irradiation of solids. The resulting damage or annealing was then measured and analyzed as to extent and mechanism, and annealing behavior was investigated. The objectives of these experiments were two-fold: (1) to contribute to better knowledge of technologically important properties of solids, thereby aiding future reactor engineering and design; and (2) to add to basic physics concepts of the nature of solids.

Copper Resistivity Studies¹

In one of the experiments, copper was irradiated with 1.25-Mev electrons at about 80° K. Electrical resistivity was measured first as a function of exposure and then after annealing at temperatures above 80° K.

The data from these irradiations are shown in Fig. 1 (Resistivity change during irradiation) and Fig. 2 (Recovery of resistivity after heating). The significant results of this investigation are listed on next page.

- a). The slope of the exposure curve, shown in Fig. 1, was found to be 4.2×10^{-28} ohm-cm/per electron/cm².
- b). On heating at an average rate of 3° C/min., 30 percent of the radiation-induced resistivity increase recovered between 80° and 233° K; 50 percent recovered between 233° and 373° K; and 20 percent recovered between 373° and 573° K. These data are shown in Fig. 2.
- c). The activation energy for the recovery state between 233° and 373° K was found to be 0.60 ev and an analysis of an isothermal recovery curve at 293° K showed that it obeyed a second order chemical rate equation.

The authors attribute the 0.60 ev annealing state to a recombination of interstitials and vacancies by the migration of interstitials. They suggest that the higher temperature recovery (above 100° C) is a result of the migration of the few remaining radiation-induced vacancies and thermally created vacancies to more complex defects such as multiple interstitials or dislocations.

Nickel Resistivity Studies²

The preliminary data from this work are presented in Fig. 3. The recovery state which begins near 100° C has not yet been analyzed to determine the activation energy for the process or to determine whether it obeys a second order chemical rate equation as was the case for copper. There is an interesting difference between the recovery in nickel (Fig. 3) and in copper (Fig. 2) in that all of the radiation-induced resistivity increase recovers in nickel during the "interstitial annealing".

Threshold Displacement Energy in Nickel³

Nickel foils, 10,000 Å thick, were irradiated at 4.2° K with a variable energy electron beam. The displacement energy in nickel was determined by measuring the minimum energy of the electrons required to produce an increase in electrical resistivity. The electron energy threshold was found to be $0.59 \pm .01$ Mev; this corresponds to a threshold displacement energy in nickel of 34.5 ± 1 ev at 4.2° K.

Preliminary annealing data were obtained by heating the specimen from 4.2° K to 373° K. About one third of the resistivity increase annealed out below 50° K. A similar annealing state was observed by Cooper, Koehler and Marx⁴ in copper and silver following irradiation with 12-Mev deuterons at temperatures below 10° K.

Radiation Annealing of Cold-Worked Copper

Cold-worked copper was irradiated⁵ at temperatures between 100 and 150° C following a two-hour anneal at 170° C. The resistivity decreased as a function of exposure; the rate of decrease was lower at the lower temperatures. Data which demonstrate this effect are shown in Fig. 4. This surprising behavior can best be explained by assuming that interstitials and vacancies migrate to dislocations, thereby causing a rearrangement of dislocations which decreases their contribution to the electrical resistivity. An activation energy for vacancy migration of 1.28 ev was determined from this experiment based on a model which assumes that at a given temperature a steady state condition is achieved in which the fraction of newly created interstitials which go to dislocations is determined by the steady state vacancy concentration. The steady state vacancy concentration will be lower at the higher temperatures; consequently a larger fraction of the interstitials will be annihilated at dislocations. It is desirable to do equivalent experiments in which mechanical property measurements are made, since these properties are primarily determined by the dislocation density and arrangement.

Effect of Irradiation on Young's Modulus⁶

Young's modulus in copper was observed to increase rapidly and saturate early as a result of exposure to 1 Mev electrons at irradiation temperatures between about -100° C and +100° C. Typical data which demonstrate this effect are given in Fig. 5. This figure also illustrates a slow decrease of modulus following the initial saturation and the annealing behavior.

The initial increase and saturation has been explained by assuming that interstitials and vacancies produced by the irradiation can pin the dislocations which lower the modulus in slightly cold-worked metals. The later decrease is attributed to the bulk lattice distortion; the interstitials are presumed to be the primary contributors to this distortion.

Graphite⁷

Graphite was irradiated with 1.25-Mev electrons at temperatures near 4° K and subsequently annealed to determine the mobility of the lattice defects introduced by irradiation. In particular the investigators wanted to determine whether any radiation damage would recover at temperatures below 100° K, since the lowest previous irradiation had been made at this temperature and there had been some recovery observed at slightly higher temperatures.

The resistivity recovery data are shown in Fig. 6. It can be seen that there is practically no effect below 80° K, and that previous irradiations made at higher temperatures had failed to observe all of the damage produced. The amount of recovery which had gone unob-

served before, however, was not significant enough to require substantial revisions to a radiation damage model⁸ which has proven to be quite satisfactory in understanding irradiated graphite.

Discussion

The previous studies have contributed not only to our knowledge of radiation damage, but also to a better understanding of the nature of solids. Many workers in the field of radiation damage feel that nuclear reactor materials studies should be concerned as much with improving our knowledge of the physics of solids as with obtaining engineering data. With electron irradiation, we feel that both purposes are being achieved.

The resistivity studies described here, when compared with similar studies which have been made using cyclotron particles⁴ for bombardment, demonstrate that both types of radiation produce about the same radiation effects. There are, however, slight differences which if explored further might yield new knowledge of the nature of solids and their interactions with radiation. This knowledge in turn could lead to a scientific approach to many metallurgical problems which at present can be solved only by trial and error.

These studies give promise of providing a method of answering many perplexing questions which arise in both radiation damage and crystalline imperfection investigations. Some of these are discussed below.

The displacement-energy experiments should be extended to metals of different crystalline structure and to non-metallic refractory materials which will be used more and more in future reactors. It is desirable to determine the cross section for atomic displacements as a function of energy using electrons of energies up to several Mev. It is desirable to explain all the details of the resistivity recovery such as that in Fig. 2 and 3. At present, the role which impurities or dislocations play in determining these details is not known. It seems likely that an understanding of the nature of dislocation and imperfection interactions will aid greatly in explaining recovery and recrystallization effects.

The relatively new phenomena of modulus increase due to irradiation and of "radiation annealing" offer additional and more direct methods of understanding interactions between crystalline imperfections in metals. Both these studies and the resistivity studies should be continued in a wider range of materials and over a wider range of irradiation temperatures.

RADIATION CHEMISTRY

Currently much consideration is being given to the development of relatively stable organic materials for use in nuclear reactors as circulating fluids (coolants and/or moderators). Reactor evaluation studies in this laboratory have indicated the feasibility and advantages of hydrocarbon moderated and cooled reactor systems. This has resulted in contract between Atomics International and the Atomic Energy Commission for the construction and operation of a diphenyl moderated and cooled reactor. Atomics International will share an appreciable fraction of the cost of this experiment. This effort is known as OMRE (Organic Moderated Reactor Experiment). Experiments are described which provide pyrolytic and radiolytic decomposition rate studies in various organic materials.

Radiation Stability of Terphenyls and Polyphenyls at 30° C and 350° C.⁹

The terphenyl isomers were chosen for study as they are among the most stable organics known with respect to both radiation and pyrolytic damage. In addition they possess the advantages of being commercially available, having low vapor pressure and low viscosity at elevated temperatures. G gas (molecules of gas formed per 100 ev) values were measured at various temperatures as a function of irradiation with 1.0 Mev electrons. Typical data are shown in Fig. 7 and 8.

The following conclusions can be made from these studies:

1. Para-terphenyl is the most radiation-stable terphenyl isomer, with ortho- and meta-terphenyl being approximately equal in radiation stability.
2. At 30° C, biphenyl is about as stable as ortho- and meta-terphenyl. Biphenyl was not investigated at elevated temperatures, as its vapor pressure is considerably higher than the terphenyls.
3. Para-quaterphenyl is more stable than any of the terphenyls but is less desirable as a coolant because it is high melting and is unavailable commercially.
4. Results on p-terphenyl - m-terphenyl and para-quaterphenyl - m-terphenyl mixtures indicate that the addition of minor quantities of the most radiation-stable compounds to other polyphenyl materials may yield a mixture possessing enhanced stability. The increased radiation stability of the various single polyphenyl compounds with increasing dosage is attributed to a similar protective action, in this case rendered by higher polyphenyl polymers formed during the irradiation.

The low radiation decomposition rate of the terphenyls as well as their low viscosity and vapor pressure at elevated temperatures indicates the suitability of these materials as circulatory fluids in nuclear reactor systems.

Pyrolytic and Radiolytic Decomposition Rates of Terphenyls at 400° C and 450° C¹⁰

This investigation is similar to the one described above⁹ except the temperature range and the measurement of different properties: G_{POLYMER} (molecules of polymer formed per 100 ev), temperature stability, and melting point. Typical data are shown in Fig. 9 and 10.

Both the pyrolyses and radiolyses experiments indicate that para-terphenyl is the most stable isomer, with ortho- and meta-terphenyl being approximately equal in radiation stability at 400° C. The stability of these polyphenyl materials increases with dosage at 450° C for both meta- and para-terphenyl, an almost constant decomposition rate (stability) being approached at the higher dosages (50 μ amp-hr/gm). Presumably the polymers formed tend to exert a stabilizing influence on the mixture.

At the lower temperature (400° C), p-terphenyl decomposes at substantially a constant rate regardless of the cumulative irradiation dosage. Para-terphenyl is not rendered more stable at 400° C by the polymer formed in the irradiation. It appears that p-terphenyl is of the same order of stability as the higher polyphenyls formed in the decomposition process.

The melting point curves obtained on the irradiated meta- and para-terphenyl samples indicate the nature of the material as it decomposes. With both terphenyls, the final melting point of the mixture decreases with increasing irradiation temperature, presumably due to the greater degree of complexity of the mixture at higher temperatures. Meta-terphenyl becomes a liquid after about 20 μ ah per gram at 450° C and after about 40 μ ah per gram at 400° C. It is interesting to notice that the final melting point of damaged para-terphenyl assumes a constant value after an irradiation dose of about 30 μ ah per gram (approximately 260° C at irradiation temperature of 450° C and 240° C at irradiation temperature of 400° C). The decreasing melting points of the mixtures with increasing radiation dosage indicate that the polymers formed are lower melting. Complete fluidity is achieved for meta-terphenyl at about 35 percent polymer formation. Ortho-terphenyl becomes a liquid after only a few percent polymer formation. The high melting isomer, para-terphenyl, damaged to 40 percent polymer formation, is still completely solid. Destructive pyrolysis (at about 500° C) and/or extended irradiation will render any of these polyphenyl materials into an infusible coke-like material.

The decomposition rates found here for the terphenyls are the lowest values reported in the literature for any organic compound under comparable conditions of temperature and radiation and suggest possible high-temperature applications for these materials in a radiation field.

Zirconium Hydride

Measurements¹¹ of the dissociation pressure of zirconium hydride were made at elevated temperatures as a function of exposure to 1 Mev electrons. These experiments are in agreement with those of other investigators who have studied the radiation stability of the metallic hydrides. In no case has there been any evidence for either reversible or irreversible changes in dissociation pressure due to radiation.

References

1. Meechan, C. J. and Brinkman, J. A. "An electrical resistivity study of lattice defects introduced in copper by 1.25-Mev electron irradiation at 80° K". (NAA-SR-1571) (1956)
2. Kenworthy, H. M. and Meechan, C. J. (Unpublished data).
3. Kenworthy, H. M. and Neely, H. H. Threshold displacement energy in nickel. NAA-SR-1580 (1956)
4. Marx, Henderson and Cooper, "Irradiation effects in Cu, Ag and Au near 10° K." Phys. Rev. 97, 599 (1955)
5. Meechan, C. J. "Annealing of Cold-Worked Copper by Electron Irradiation", NAA-SR-1654 (1956)
6. Dieckamp, H. and Sosin, A. (unpublished data)
7. Austerman, S. B. and Hove, J. E., "Irradiation of Graphite at Liquid Helium Temperatures". Phys. Rev. 100, 1214 (1955)
8. Hennig, G. R. and Hove, J. E., "Interpretation of Radiation Damage in Graphite". Geneva Conference on Peaceful Uses of Atomic Energy.
9. Colichman, E. L. and Gercke, R. H., "Radiation stability of the terphenyls and other polyphenyl materials as measured by gas evolution". NAA-SR-1288 (1955)
10. Colichman, E. L. and Fish, R., Pyrolytic and radiolytic decomposition rate studies on orth-, meta-, and para-terphenyls. NAA-SR-1287 (1955)
11. Gilbert, Jr., P. T. and Goeddel, W. V., "Zirconium Hydride". NAA-SR-1399 (1956)

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1. Electrical Resistivity Changes in Copper During a 1.25 Mev Electron Irradiation at 80° K

Fig. 2. Recovery of Electrical Resistivity of Copper After 1.25 Mev Electron Irradiation at 80° K (Heating Rate --- 3° K/min)

Fig. 3. Tempering of Nickel Irradiated at About 80 $^{\circ}\text{K}$ With 1.25 Mev Electrons.

Fig. 4. Rate of Resistivity Decrease With and Without Irradiation, Average Beam 6×10^{14} electrons/cm²-sec with 1.25 Mev energy.

Fig. 5. Extended Irradiation and Annealing of 99.999% Copper.
Specimen Maintained at Each Temperature for 5 Minutes.

Fig. 6. Tempering of Graphite Irradiated at 4° K with 1.25-Mev electrons. Resistivity Measured at 4° K.

Fig. 7. G_{Gas} vs Dosage at 30° C

Fig. 8. G_{Gas} vs Dosage for Terphenyls and P-Quaterphenyl at 350° C

Fig. 9. Statitron Irradiated p-Terphenyl at 450° C
(1-Mev Electrons)

Fig. 10. G_{polymer} for Meta- and Para-Terphenyl at 450° C

RADIATION DAMAGE OBSERVATIONS
AT THE MTR

by

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The MTR has operated successfully for over four years without failure of components that could be attributed to radiation damage. Long time exposure of metals used in construction of the core of the MTR produced changes in their properties but not to a functionally damaging extent. Failures of fuel elements have been attributed to excessive hydraulic forces. High pressure, elevated temperature equipment has functioned satisfactorily although the design does not fall within areas covered by existing pressure vessel and piping codes.

RADIATION DAMAGE OBSERVATIONS AT THE MTR

The Materials Testing Reactor went critical in March of 1952 and after a short time at low power has operated since that time on a continuing basis at power levels of either 30 or 40 megawatts. While some evidence of radiation effects on MTR fuel assemblies and structural components has been observed, there have been no failures of MTR components and no reactor down time that has been attributed to malperformance of MTR metal components arising from radiation damage.

The fact that the MTR to date has not been troubled by failure of components from radiation damage does not mean that the future will be as pleasant nor does it mean that other reactors will be trouble free in this respect. Actually there is a great deal of concern at the MTR about radiation effects and the probability of trouble from these effects. The non-fuel-bearing metal components are in an environment of high thermal resonance and fast flux. It is likely that some of the MTR components now have been exposed to more neutrons than any other known

reactor materials. Fuel elements, while replaced frequently, are subject to all the damaging effects of light particle radiation plus that of massive fission fragments. While fission product leaks have occurred in MTR fuel assemblies, there is no evidence that radiation damage was responsible. Experiments conducted at the MTR are of constant concern, particularly when it is realized that the characteristics of the in-pile experimental equipment are constantly changing and failure of any one of many of these experiments could seriously damage the MTR. Similarly the characteristics of control rods and safety shim rods are changing constantly in a direction of reduced safety and must be replaced as the cadmium burns out.

Efforts at the MTR have been directed toward operating the reactor efficiently, safely and on a permanently continuing basis. The investigations of radiation damage have been directed along lines that would assure accomplishment of these goals. The balance of this discussion covers the investigations aimed at evaluating the condition of MTR metal components and observations of radiation damage that have been made during the operating lifetime of the MTR.

CHANGES IN PROPERTIES OF METAL COMPONENTS

The metal components of the MTR that are not fuel bearing and are either in the core or very near the active lattice are constructed of one of four materials--A.I.S.I. Type 347 stainless steel, QMV beryllium, aluminum alloy 1100 (2S) or aluminum alloy 356. It was feared, at the time of reactor start up, that radiation induced changes in these materials might cause failures and malfunctioning of the reactor. The components of particular concern were those in the regions of high thermal and fast flux and to assess the changes occurring in these components, it was decided to install appropriate test coupons some of which could be removed after exposure in the reactor for 1 year, 2 years, 4 years,, etc. It was felt that the condition of these coupons at the time of test should approximate the condition of the reactor components constructed of similar materials. Coupons of beryllium, type 347 stainless steel, 2S and 356S aluminum were placed in a modified beryllium piece next to the core of the reactor. This position, L-43, providing one of the highest thermal and fast flux regions in the reactor, may be located in Figure 1.

Two types of coupons were placed in the modified beryllium piece. One type consisted of a series of washers and was used to measure corrosion performance of the materials used in the reactor. The second type of coupons consisted of small cylindrical tensile coupons with reduced sections one and one-half inch in length and diameters of the reduced sections of 0.252 inches. Four tensile coupons, one of 347 stainless, one of 2S aluminum, one of 356S aluminum, and one of beryllium, were supported in each set of coupons by means of end spacing plates as shown in Figure 2.

FIG. 1
REACTOR TANK
CROSS SECTION,
SHOWING POSITION OF FUEL AND L-43

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FIG. 2
TENSILE SPECIMEN HOLDER

Six sets of tensile coupons were loaded in the reactor on October 15, 1952. The first set was removed on February 3, 1953 after 2756 MWD (megawatt days) exposure. A second set was removed on September 9, 1953 after 8387 MWD exposure. A third set was removed on April 30, 1956 after 31048 MWD exposure; the last 6108 MWD being at 40 MW operation and all earlier at 30 MW. The accumulated neutron flux, of 100 ev and high energy neutrons, to which these samples were exposed was about 2.4×10^{21} , 7.2×10^{21} , and 2.6×10^{22} respectively for the first, second, and third sets of samples¹. The thermal neutron flux to which these samples were exposed was also about 2.4×10^{21} , 7.2×10^{21} , and 2.6×10^{22} nvt. The ratio of flux above 1 Mev to that above 100 ev was about 1 to 10 for the radiation location of these specimens¹.

The first and second sets of coupons removed from the reactor were tested at Oak Ridge National Laboratory² as hot cell facilities were not completed at the MTR at that time. The last set of coupons was tested at the MTR. Of six sets of coupons placed in the reactor in October of 1952, three have been removed and tested, one remains in the reactor, and two were damaged during routine unloading operations and were discarded.

The changes in properties that took place in samples of aluminum alloy 1100 are summarized in the following Table I and in Figure 3.

TABLE I

PROPERTIES OF 2S ALUMINUM BEFORE AND AFTER REACTOR EXPOSURE

Total nvt (>100 ev)	Density	Rockwell Hardness	Stress at 0.2% Offset psi	Ultimate Strength psi	% Elong. in 1 inch	Red. of Area
0	2.713	6.3F	18,100	20,150	23.0	81.0
0	2.713	10.4F	18,700	20,350	21.6	81.5
2.4×10^{21}	2.712	36.5F	23,200	26,950	25.5	78.0
7.2×10^{21}	2.712	46.9F	23,550	29,200	20.2	75.0
2.6×10^{22}	---	42.3F	26,500	30,400	23.0	73.0

Changes in properties of aluminum casting alloy 356 are summarized in the following Table II and in Figure 4.

FIG. 3
RADIATION DAMAGE
TO
ALUMINUM ALLOY 1100

TABLE II

PROPERTIES OF 356 ALUMINUM BEFORE AND AFTER REACTOR EXPOSURE

Total nvt (>100 ev)	Density	Rockwell Hardness	Stress at 0.2% Offset psi	Ultimate Strength psi	% Elong. in 1 inch	Red. of Area
0	2.663	29.6B	24,000	32,800	2.9	--
0	2.667	29.0B	24,100	32,000	2.4	--
2.4 x 10 ²¹	2.660	60.6B	42,400	44,300	0.4	--
7.2 x 10 ²¹	2.659	64.3B	51,000	54,000	0.9	--
2.6 x 10 ²²	---	46.6B	36,600	45,000	1.5	--

Changes in properties of QMV beryllium are summarized in the following Table III and in Figure 5.

TABLE III

PROPERTIES OF QMV BERYLLIUM BEFORE AND AFTER REACTOR EXPOSURE

Total nvt (>100 ev)	Density	Rockwell Hardness	Stress at 0.2% Offset psi	Ultimate Strength psi	% Elong. in 1 inch	Red. of Area
0	1.847	70.6B	23,200	35,000	1.4	--
0	1.847	71.5B	25,800	36,400	1.4	--
2.4 x 10 ²¹	1.847	88.8B	--	51,400	0.2	--
7.2 x 10 ²¹	1.847	97.4B	--	50,400	0.2	--
2.6 x 10 ²² *	---	88.7B	> 47,800	> 47,800	--	--

*Coupon broke in grips

Type 347 stainless steel exhibited the properties shown in the following Table IV and in Figure 6 after various neutron exposures.

TABLE IV

PROPERTIES OF TYPE 347 STAINLESS STEEL BEFORE AND AFTER REACTOR EXPOSURE

Total nvt (>100 ev)	Density	Rockwell Hardness	Stress at 0.2% Offset psi	Ultimate Strength psi	% Elong. in 1 inch	Red. of Area
0	7.890	16.4C	66,300	101,800	57.0	74.5
0	7.907	12.4C	56,900	89,800	69.4	81.5
2.4 x 10 ²¹	7.896	20.5C	98,000	109,000	51.6	78.5
7.2 x 10 ²¹	7.892	20.6C	97,300	109,700	50.6	77.8
2.6 x 10 ²²	---	21.8C	100,800	107,250	44.0	65.0

FIG. 4
RADIATION DAMAGE
TO
ALUMINUM ALLOY 356

FIG. 5
RADIATION DAMAGE
TO
BERYLLIUM

FIG. 6
 RADIATION DAMAGE
 TO
 STAINLESS STEEL
 AISI TYPE 347

- ULTIMATE STRENGTH
- ◉ YIELD STRENGTH
- ▲ ROCKWELL C HARDNESS
- ELONGATION

Dimensional measurements made on the coupons before radiation and after various exposures failed to reveal any changes in original dimensions.

While changes are evident in the properties of the metals used in construction of the MTR, these changes have not been great enough to date to impair the functioning of any component in the MTR. Beryllium and 356 aluminum are brittle materials in their unirradiated condition and the design and operational practices of the MTR have taken this fact into account. The further reduction in ductility that has taken place with service in the reactor has not influenced the performance of the 356 aluminum support castings nor the beryllium reflector pieces of the MTR. Aluminum alloy 1100 and stainless steel, type 347, were ductile material before irradiation and to date the embrittlement of these alloys from neutron radiation has not been great.

It is encouraging to note that these tests confirm other earlier tests in showing that while there are significant changes in properties of materials exposed to neutrons, these changes occur most rapidly during early exposure to neutrons and change slowly after initial low level exposures. The distinction between the effects of fast neutrons and their disturbance of original crystal lattice and the effects of slow neutrons and gradual addition of impurity atoms to a metal with long time exposures has not been established by this investigation.

Corrosion coupons, previously mentioned, have been removed from time to time in an effort to determine corrosion losses. Unfortunately the coupons were poorly designed with large weight to exposed surface ratios and other than permitting the general observation that corrosion rates are not objectionably high, the coupons have been of little value.

MTR FUEL ELEMENTS AND CONTROL RODS

The MTR fuel assembly has evolved from an 18 plate element with 140 grams of U²³⁵ to a 19 plate element with 200 grams of U²³⁵. In the process of making these changes engineering errors were made and as a result some fuel elements have failed and fission product breaks occurred.

For two years, from start-up in 1952 to July of 1954, all fuel elements functioned as intended. During this time several elements were intentionally subjected to much larger than normal fuel burn up. While the maximum intended lifetime of the 140 gram fuel assemblies was one MTR cycle (3 to 4 weeks per cycle), heavier fuel loaded elements were visualized as operating during two or three cycles. To check the radiation effects at the higher burn-ups demanded by longer reactor service, 140 gram elements, and 168 gram elements that were now coming into use, were retained in the fuel lattice for 5 to 10 cycles. After this time the elements were visually examined and appeared to be perfectly capable of continuing operation had sufficient fuel remained in

them. High priority programs required these elements to be dissolved before measurements of final mechanical properties could be made.

The conditions causing failures in MTR fuel assemblies and the corrective measures taken have been described in detail at an earlier AEC Metallurgy Meeting³. Briefly, the account is this. In June of 1954, the activity in the MTR process water began to rise at a steady but not alarming rate. Within a few days it became apparent that the reactor had suffered a fission break. On careful examination of the fuel assemblies, a number of elements were found to have developed blemishes on the surface of outside fuel plates. Typical blemishes are shown in Figures 7 and 8.

The blemishes generally started from one to two inches from the bottom of the fuel and extended upwards from one to ten inches. They were generally elliptical in shape, but varied considerably. They were characterized color wise by a light grey zone surrounding a darker region. Smears of a number of these blemishes and an analysis of I¹³³ showed conclusively that the defects penetrated the cladding and exposed the alloy core.

Significantly, blemishes were only found on the outside surfaces of outer fuel plates and all inside plates were free of blemishes. Further work revealed that the lower portions of the outside concave plates of the elements buckled outward during time in the reactor. The buckling is shown schematically in Figure 9. Concave plates deflected enough to touch the mating convex plate of the adjacent element (Figure 9). Overheating apparently resulted, causing the cladding to corrode away rapidly and fission breaks occurred.

It is interesting to consider why, after two years of operation without the slightest trouble from any fuel assembly, the MTR was suddenly plagued by these fission breaks.

FIGURE 7

BLEMISH ON OUTSIDE MTR FUEL PLATE

FIGURE 8

ENLARGED VIEW OF SURFACE BLEMISH

FIG. 9
DEFLECTION
OF
FUEL ASSEMBLY PLATE

The reason probably lies in the fact that a number of changes were made in the design of the fuel assembly and the pertinent ones contributing to these failures are shown in Figure 10.

The thickness of the fuel plates was reduced, the thickness of the side plates was reduced, and the opening in the lower fuel element casting was made smaller. To confuse the problem, the fuel content had increased from 140 grams to 168 grams, and at the time the first break occurred, to 200 grams. There were two effects of these changes. First, the element was weakened through thinning of fuel and side plate and second, the water pressure within the element was raised by restricting flow through the lower end box.

Figure 11 shows results of pressure measurements made in different elements under dynamic flow conditions in the reactor in 1952 and 1954. Lateral pressure differences of about 10 psi were found to exist across the lower end of the outside concave plate of elements of the type that were failing. Pressure tests of their plate assemblies showed that the concave plate would buckle under about 9 psi.

Remedial measures taken were (a) the outside fuel plates were increased in thickness from 0.050" to 0.062", (b) the side plates were increased from 0.106" to 0.188", and (c) the cross-sectioned area of the water exit channel of the lower end box was restored to the original 3.7 sq. inches. These changes reduced the lateral pressure differences to less than 5 psi and increased the element's resistance to buckling from 10 psi to about 20 psi.

There was no evidence that radiation damage and accompanying changes in mechanical properties of fuels was responsible for the fission breaks that occurred. The core and cladding had hardened from initial values of about 26-39 DPH for both core and cladding to about 155 DPH for core and about 40 DPH for cladding after service⁴.

Fuel sections of shim rods are exposed to very long service in the reactor. Fuel burn out has been very high and the fuel sections have been found to be in satisfactory condition, except with less than necessary fuel, when discharged.

Regulating rods and shim rods containing cadmium as the controlling poison are used in the MTR until control effectiveness is lost due to burn out of Cd¹¹³. Figure 12 shows a transmission curve for cadmium sections removed from shim rods after various time in the reactors⁵. The cadmium that was completely transparent to 0.05 ev neutrons could not be distinguished from the cadmium that was totally black to neutrons by visual examination or by bend tests. More detailed examination of the mechanical properties are in progress.

LOWER END BOX

ORIGINAL

a - 0.060
 b - 0.060
 c - 0.1875
 A - 3.7 SQ. IN.

INTERIM

.050
 .050
 .106
 3.2 SQ. IN.

FINAL

0.0625
 0.050
 0.1875
 3.7 SQ. IN.

FIG. 10
 DIMENSIONAL CHANGES IN MTR FUEL ELEMENTS

TOP SECTION A-A

BOTTOM SECTION B-B

FIG. II
STATIC PRESSURES IN MTR FUEL ELEMENTS IN PSI.

FIGURE 12

CHARACTERISTICS OF CD IN SPENT SHIM RODS

HIGH PRESSURE EQUIPMENT AT THE MATERIALS TESTING REACTOR

A number of experimental holes in the MTR have been in the past and are now occupied by elevated temperature, high pressure water loops. All of the loops have been constructed of austenitic stainless steels, types 304, 347, or 316 with the exception of one constructed of carbon steel, ASTM A201 grade B. These loops are constructed to provide irradiation facilities simulating conditions that are expected to be encountered in planned reactors. Generally the loops are intended to test corrosion and radiation performance of fuel assemblies; however, the loops themselves are in a sense under test. The systems operate with water temperatures to 650°F and pressures up to 2050 pounds per square inch. If a failure of any one of these loops occurred in an in-pile area, there would exist a real danger of damage to the reactor core.

In view of the potential danger that exists from failure of one of these loops, each of the sponsoring experimenters has conducted a rigorous stress analysis of the in-pile components. In some instances, the reliability of the pressure containing equipment has been established by constructing the equipment in conformance with interpretations of the ASA Code for Pressure Piping. Other designs that could not be constructed to fit the requirements of existing codes have been based upon the criteria, described by B. F. Langer⁶, which permits relaxation of peak surface stresses in consideration of allowable operating conditions.

The in-pile sections of these loops vary in design, but generally consist of either straight through tubular sections or of cylindrical sections closed with hemispherical caps. A typical design might be that shown in Figure 13. The walls of cylindrical sections may be tapered and the thickness of the hemispherical section varying from a maximum at its junction with the cylindrical portions to a minimum at the furthest extension of the hemispherical closure. The reason for varying wall thickness is that at the region of maximum gamma heating wall thickness must be reduced to minimize thermal stresses. In lower gamma heating areas additional metal for strength purposes may be added without increasing objectionable thermal gradients. While an in-pile pressure tube is heated uniformly across its walls from gamma radiation, it is usually cooled only from the inside surfaces by the high pressure water circulating through the tube. The high metal temperature on the outside surface of the tube and the lower temperature of the inside fibers give rise to thermal stresses that are in tension on the inside surfaces and if no pressure stresses were in effect are in compression on the outside surfaces. A hypothetical case showing stress conditions with pressure and thermal stresses operating singly and combined is shown in Figure 14. If the maximum stress permitted is determined from simple addition of the thermal and pressure stresses at the inside surfaces, the design is restricted in the sense that only the inner fibers are at maximum load and the rest of the structure is loaded at lower stresses.

IN-PILE PRESSURE THIMBLE

FIGURE 13

TYPICAL IN-PILE PRESSURE THIMBLE

VARIATION OF PRESSURE AND THERMAL
STRESS ACROSS CYLINDER WALL

FIGURE 14
STRESS CONDITIONS IN IN-PILE TUBE SECTION

Stresses of typical loops that have been in operation at the MTR are summarized in Table V showing the longitudinal stress conditions at the outside and inside surface of the walls at points of maximum stress.

TABLE V

THERMAL AND PRESSURE STRESSES IN IN-PILE PRESSURE EQUIPMENT

	Pressure Stress Inside Fibers	Thermal Stress Inside Fibers	Combined Stress Inside Fibers
Loop No. 1	+ 15,000 psi	+ 26,900 psi	+ 41,000 psi
Loop No. 2	+ 15,800 psi	+ 7,500 psi	+ 23,300 psi
Loop No. 3	+ 13,600 psi	+ 12,700 psi	+ 26,300 psi
Loop No. 4	+ 14,900 psi	+ 8,000 psi	+ 22,900 psi

After several months of operation (178 days) an in-pile section of one of the stainless steel loops was removed from service and destructively tested at Argonne National Laboratory⁷. This tip had been exposed to about 3×10^{20} nvt (> 1 Mev). The tip was in good condition and perfectly capable of continued service. The hardness at the tube tip was about 61 Rockwell A (21Rc) and appeared to decrease uniformly as the distance from the tip increased until about 8 inches back from the tip, the hardness was down to about 58 Rockwell A. Hardness readings taken in the weld that was located about 6 inches back from the tip varied from RA 57 to RA 62. Segments were cut from welded and unwelded portions of the tube, clamped in a vise and flexed until failure. The samples were bent through a total angle of about 160° (80° in each direction from normal) for 10 to 12 cycles without failure. When failure did occur, they were ductile in nature. From this, it may be concluded that the metal in the tube tip had reached the saturation conditions that have been predicted⁸.

Similar tests were conducted on the in-pile portion of a carbon steel loop that had operated at 2050 psi and about 600°F ⁹. Tensile coupons, crush rings and hardness strips were machined from the loop after exposure of about 3×10^{19} nvt (> 1 Mev).

The location of the various coupons are shown in Figure 15 and the partially machined tip is shown in Figure 16. The data derived from the tensile coupons are tabulated in Table VI.

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FIG. 15

LOCATION AND TYPE OF TESTING
COUPONS CUT FROM CARBON STEEL LOOP

FIGURE 16

PHOTOGRAPH OF PARTIALLY MACHINED TUBE TIP

TABLE VI

PROPERTIES OF CARBON STEEL LOOP BEFORE AND AFTER SERVICE AT THE MTR

	Before Radiation	After MTR Service
Yield Strength	43,000 psi	46,500 psi
Ultimate Strength	72,800 psi	76,000 psi
Elongation, % in 2"	41.0	29.0
Red. of Area, %	53	50
Breaking Strength	127,000 psi	125,000 psi

A steep flux gradient existed along the tube, and the tensile data reflects the properties at the lowest exposure. Figure 17 shows a typical fracture of tensile coupon and the fact that it broke as all others at one end of the reduced section. This end at which fracture occurred had been in the lowest flux. Hardness varied from R_p 92 to R_p 72 along the tube as shown in Figure 18. A crush ring failed to exhibit the same flattening characteristics of the unirradiated tube; however, severe embrittlement to slow deformation was absent. A hardness strip cut through the hemispherical end of the tip was crushed flat without cracking (Figure 19), although a 2" wide crush ring cut about 3" away from the tip flattened from its original diameter of 4" to 1-1/2" before cracking. When failure did occur, it was with a sudden snap.

The fact that the MTR has operated for the length of time that it has without failures from radiation damage is encouraging. It is felt, however, that much remains to be done in establishing the very long time effects of radiation on metal components before design people can have complete confidence in structures intended to operate in neutron environment for the long times expected of power reactors.

FIGURE 17

FRACTURED TENSILE COUPON CUT FROM CARBON STEEL TIP

FIG. 18
HARDNESS PROFILE OF CARBON STEEL TUBE

FIGURE 19

CRUSH RING AFTER FLATTENING

BIBLIOGRAPHY

1. Hogg, C. H., and Lewis, W. B., Phillips Petroleum Company, Atomic Energy Division, Idaho Falls, Idaho, Private communication.
2. Wilson, J. M., Oak Ridge National Laboratory, Letter of August 3, 1954, ORNL-54-8-6, to R. L. Doan, Phillips Petroleum Company. "Results of Tensile Tests".
3. Burnham, J. B., Elgert, O. J., and Bartz, M. H., "Breaks in Cladding of MTR Fuel Elements", "Proceedings of Metallurgy Information Meeting", 1955, AEC Secret Document TID-7502 (Pt. II), pp 809--824.
4. Feldman, M. J., Richt, A. E., Manthos, E. and Long, E., "Metallographic Analysis of Sections 1, 2 and 3 of MTR Element I-700", ORNL-55-3-182, April 29, 1955, Secret.
5. Elgert, O. J., Phillips Petroleum Company, Atomic Energy Division, Private communication.
6. Langer, B. F., "Working Stress Criteria for Nuclear Power Plants", Trans ASME, July, 1955.
7. Freund, G. A., Private communication on Examination of First Pile Tube Tip from ANL-2 Loop.
8. Fromm, L. W., Design Evaluation of In-Reactor Tube for Argonne Water Loop at MTR, ANL-5403.