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# Mechanical Properties of Low Temperature Irradiated Magnesium

By

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Magnesium crystals were irradiated at 77 °K in the RA-1 reactor to neutron doses up to $4\times10^{18}$ ncm$^{-2}$, and the deformation properties were studied at this temperature without warm-up. The yield stress increased with dose to a value of $(465 \pm 45)$ p/mm$^2$ at $1\times10^{17}$ ncm$^{-2}$. For neutron doses greater than this value, it was found that the deformation mechanism changed with an upper yield point being formed with an accompanying yield drop. The upper yield point and yield drop increased with dose and became very substantial at $4\times10^{18}$ ncm$^{-2}$ with the resolved yield stress increasing to 1000 p/mm$^2$ and the yield drop to the order of 50%. The lower yield stress was independent of dose in the higher dose region. Annealing experiments showed an annealing peak centered at 240 °K. For large doses, recovery first occurred in the upper yield stress with the lower yield stress being unaffected until complete recovery of the upper yield stress occurred. Strain markings were studied and found inhomogeneous in character but different from a typical Lüders band.

Magnesiumeinkristalle wurden bei 77 °K im RA-1-Reaktor mit Neutronendosen bis zu $4\times10^{18}$ ncm$^{-2}$ bestrahlt und die Deformationseigenschaften bei dieser Temperatur ohne Aufwärmung untersucht. Die Streckgrenze steigt mit der Dosis auf einen Wert von $(465 \pm 45)$ p/mm$^2$ bei $1\times10^{17}$ ncm$^{-2}$ an. Für Neutronendosen, die größer als dieser Wert sind, wurde gefunden, daß sich der Deformationsmechanismus mit einer oberen Fließgrenze, die mit einem begleitenden Flußabfall gebildet wird, ändert. Die obere Fließgrenze und der Flußabfall steigen mit der Dosis an und werden bei $4\times10^{18}$ ncm$^{-2}$ sehr wesentlich, wobei die aufgelöste Streckgrenze auf 1000 p/mm$^2$ ansteigt und der Flußabfall auf die Größenordnung 50%. Die untere Streckgrenze war unabhängig von der Dosis im Bereich der höheren Dosen. Temperungsexperimente zeigten ein Temperungsmaximum bei 240 °K. Für große Dosen tritt Erholung zuerst in der oberen Streckgrenze auf, wobei die untere Fließgrenze unbeeinflußt bleibt bis vollständige Erholung der oberen Streckgrenze eingetreten ist. Deformationsmarkierungen wurden untersucht und es wurde gefunden, daß sie inhomogen im Charakter, jedoch von typischen Lüdersbanden verschieden waren.

## 1. Introduction

The effects of neutron irradiation on the mechanical properties of the face-centered cubic metals have received widespread attention in the past twenty years [1, 2]. The effects are quite pronounced in these metals with the yield stress increasing by a decade or more for moderate irradiations. By comparison the hexagonal metals have been neglected, as the only work reported has been that of Kunz and Holden [3]. They studied the effect of room temperature irradiations on the stress/strain curve of zinc. This work, which was largely of the survey type, demonstrated a substantial increase in the yield stress of the irradiated zinc.

The present work was undertaken to attempt to fill at least in part this information gap in the field of irradiation effects. Magnesium was chosen for this study because it has low level radioactivity after reactor bombardment, it can be obtained in high purity, and it is ductile at 77 °K.

## 2. Experimental Procedure

Single magnesium crystals were grown from Johnson Mathey 99.99% magnesium, using the Bridgman technique, in the shape of square tensile specimens of 9 mm² cross-section and 3 cm long with spherical gripping heads 6.35 mm diameter in split molds made from reactor grade graphite. Their crystallographic orientation was determined by the back reflection Laue-technique. Although there were a few exceptions, most samples were selected with orientations given by $X = 58°$, $\lambda = 33°$, where $X$ is the angle between the basal plane (0001) and the specimens axis, and $\lambda$ is the angle between the $\langle 1120 \rangle$ direction (slip direction) and the crystal axis. Their yield stress was $(40 \pm 5)$ p/mm².

The irradiation was performed in the cryogenic facility located in the core of the RA-1 reactor of C.N.E.A. In this cryostat, which was designed and built at Grenoble [4], the samples are bombarded in a bath of liquid nitrogen. Although a complete description of this device is available in the literature [4], it is worth noting that refrigeration is provided by liquid nitrogen utilizing a reflux cycle. The samples were loaded into the cryostat in an aluminum can that was completely immersed in liquid nitrogen in order to permit withdrawal of the samples without warmup. The liquid nitrogen in this can was capable of keeping the samples cold for a period of time, which greatly exceeded the removal time.

The neutron flux in this location was determined by the yield stress of copper single crystals. The yield stress, measured as a function of the effective bombardment time, was then compared with yield stress dose curves previously published

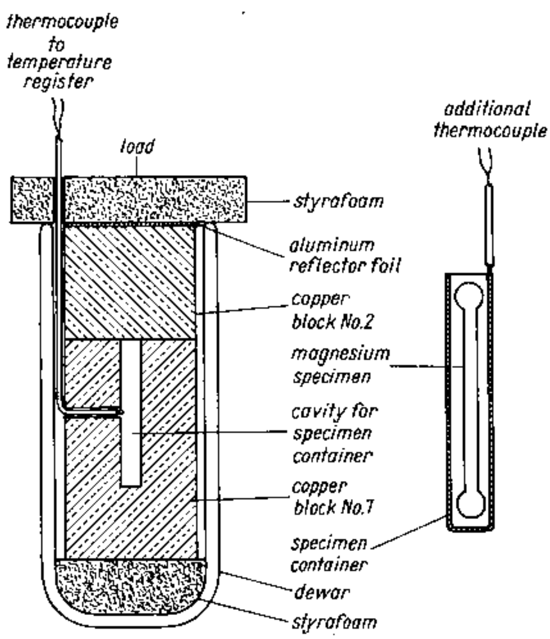



Fig. 1. Schematic diagram of the annealing cryostat. Both copper blocks were brought to the desired temperature. The sample, which is the specimen container and is stored in liquid nitrogen, is placed in the cavity of copper block No. 1. The heat capacity of the copper is so great compared to the sample that the temperature of the system was virtually unaffected by the insertion of the samples. A good square pulse can be obtained from this cryostat

to determine the flux. The dose values reported in this paper are then directly comparable to those reported in the Oak Ridge graphite reactor [1]. This technique yielded a flux of $3.3 \times 10^{12}$ reactor neutrons/cm$^2$ s. Since it is estimated that only about $1 \times 10^{12}$ n/cm$^2$s of these are above 0.5 MeV, the dose values reported here are about a factor of three greater than the fission neutron dose.

After removal from the cryostat, the crystals were stored in liquid nitrogen for a week or so to allow the activity to decay. They were then mounted in an Instron tensile machine (Model TTM) and deformed with a crosshead speed of $1 \times 10^{-3}$ cm/min. The mounting was performed with the sample immersed in liquid nitrogen to prevent warmup. On some samples, deformation markings were examined by photomicrography as the deformation proceeded. The samples were removed from the tensile machine and photographed while immersed in liquid nitrogen. Good images could be obtained through the liquid nitrogen if the liquid layer were less than 3 mm thick.

Recovery measurements were performed in a very simple manner. A high mass copper block with a sample cavity in it was thermally isolated at the desired annealing temperature (Fig. 1). The sample was removed from the Instron machine, immersed in liquid nitrogen, and inserted in the cavity for the desired annealing time. Since the heat capacity was very high in comparison to that of the specimen, insertion of the specimen did not appreciably change the block temperature. A typical annealing curve is shown in Fig. 2. Following the annealing pulse, usually of ten minutes duration, the sample was removed from the cavity and quenched in liquid nitrogen. It was remounted in the Instron machine while immersed in liquid nitrogen, and the effect of the pulse on the yield stress was determined at the reference temperature of 77 °K. Since the same sample was used throughout the anneal, the plastic strain required to determine the yield stress was kept as small as possible.

## 3. Results

Load elongation curves obtained on crystals made to various doses at 77 °K and tested at the same temperature without intermediate warmup are shown in Fig. 3. Clearly from these data a fundamental change in the yielding phenomenon occurs after a critical dose is reached.[1]) For low doses a "normal" yielding phenomenon occurs, whereas for high doses the yield stress is multivalued with an "upper" and "lower" yield stress. The critical neutron dose for this transition is of the order of $1 \times 10^{17}$ reactor neutrons/cm$^2$.

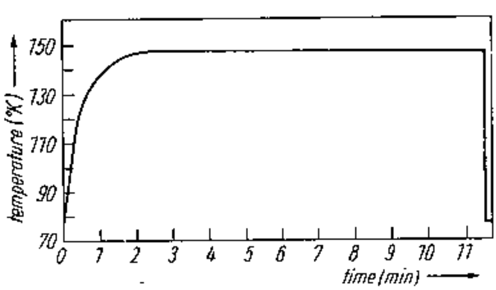



Fig. 2. Typical annealing pulse, $T = 148$ °K

[1]) The authors were guided by the results of our colleague, A. B. Gonzales, who made in situ measurements in the RA-1 reactor of the yield stress of magnesium as a function of dose. These results strongly suggested saturation effects and will be published soon.

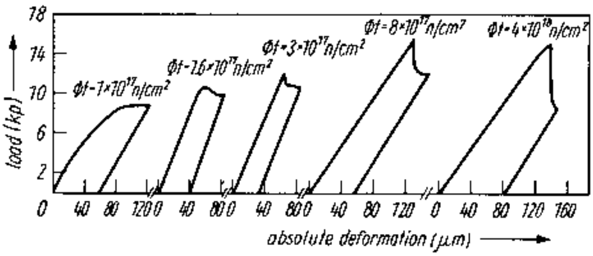
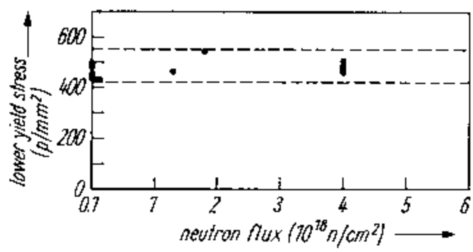



Fig. 3. Load–elongating curves at 77 °K for magnesium crystals bombarded to various doses. The samples have different orientations; it is obvious that the load drops increased as a function of dose

Fig. 4. The lower yield stress, measured on magnesium crystals bombarded and tested at 77 °K as a function of dose, shows that within experimental error, the lower yield stress is independent of dose



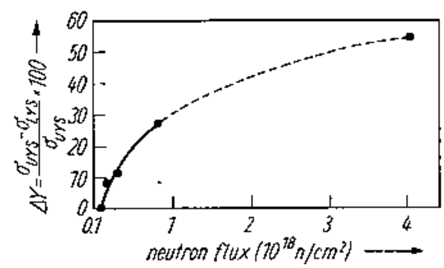




Fig. 5. The yield drop plotted as a function of neutron dose for magnesium crystals irradiated and tested at 77 °K

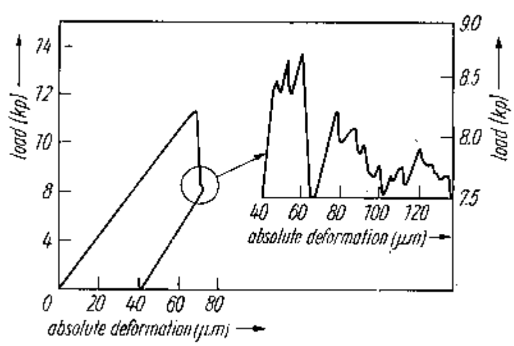



Fig. 6. The load drop for a sample bombarded to 4 × 10¹⁸ reactor neutrons/cm². The load-elongation curve for the lower yield stress was obtained on an expanded scale and is shown in the inset. The typical jerky flow makes it difficult to accurately define the lower yield stress

Consider the behavior of the crystals in the high-dose region, i.e. for doses greater than $1 \times 10^{17}$ reactor neutrons/cm². As seen in Fig. 4, the lower yield stress is independent of the neutron dose for doses ranging from $1 \times 10^{17}$ to $4 \times 10^{18}$ reactor neutrons/cm² with a value of $(485 \pm 45)$ p/mm² compared to a value of $(40 \pm 5)$ p/mm² for the annealed crystal. However, the upper yield stress, and consequently the yield drop, increases with neutron dose. This is shown in Fig. 5. The yield drop, $\Delta Y$, is defined as

$$\Delta Y = \left(\frac{\sigma_{\mathrm{uys}} - \sigma_{\mathrm{lys}}}{\sigma_{\mathrm{uys}}}\right) \times 100\,,$$

where $\sigma_{\mathrm{uys}}$ is the upper yield stress and $\sigma_{\mathrm{lys}}$ is the lower yield stress. Details of the "yielding" are shown in Fig. 6. Here a sample bombarded to $4 \times 10^{18}$ reactor neutrons/cm² is strained to the yield point, and the test stopped. The load cell of the tensile machine is then set on an expanded scale and the crystals tested again. Jerky flow follows, implying inhomogeneous straining and making lower yield stress difficult to define.

Annealing experiments seem to verify that the yield drop is a direct consequence of the defect concentration. That is, the removal of the defects by thermal migration reduces the yield drop without affecting the lower yield stress until the yield drop is completely recovered. The main annealing peak (for yield point recovery) occurs in the region above 200 °K with small, if any, recovery in the region from 77 to 200 °K. The annealing experiments were conducted by the study of the recovery of the yield drop and the recovery of the lower yield stress.

The yield drop experiments were rather limited in scope, as only two specimens were tested. Both were bombarded to $4 \times 10^{18}$ reactor neutrons/cm² and tested at 77 °K. The first crystal deformed after a pulse anneal for 10 min at 170 °K with the yield stress, both upper and lower, being unaffected with experimental error ($\pm 10\%$).

The second crystal was deformed after pulse annealing to 273 °K for 30 min. In this case, practically complete recovery of the yield drop occurred without affecting the lower yield stress. The load elongation curve for this sample is shown in Fig. 7.

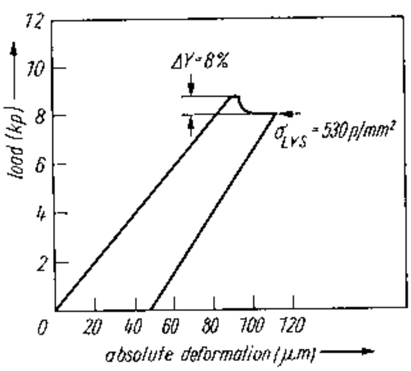



Fig. 7. The load–elongation curve for a sample bombarded to $4 \times 10^{18}$ reactor neutrons/cm² at 77 °K after being annealed for 30 min at 273 °K

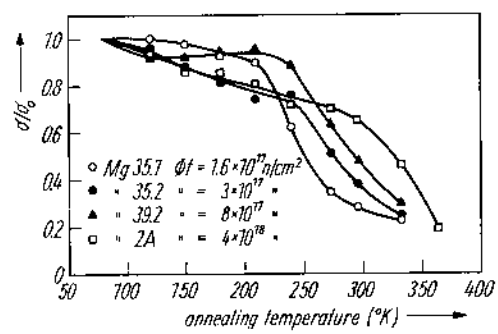



Fig. 8. Isochronal annealing curves of the lower yield stress in 77 °K bombarded magnesium. The samples were strained by about 0.2% at 77 °K prior to the recovery to remove the upper yield stress. Pulse annealing techniques were used. The temperature pulses failed to reactivate the upper yield stress

The effects of annealing on the lower yield stress were determined in a more detailed manner. Prior to annealing, all samples were prestressed to remove the upper yield stress with the aim of obtaining a reproducible lower yield stress. Pulse annealing techniques were used with 10 min pulses. Fig. 8 shows the yield stress at 77 °K as a function of the pulse temperature.

If the low-temperature annealing is ignored because of the lack of definition of the lower yield stress, apparently, a significant recovery of the yield stress occurs in the 200 to 350 °K temperature ranges. Furthermore, this annealing peak shifts to a higher temperature with increasing dose. The temperature shift of the annealing peak is in the wrong direction to be explained by the usual annealing kinetics, as either the peak should be unaffected by defect concentration (first order kinetics) or the peak should decrease with increasing concentration (second or higher order kinetics). This temperature shift can be explained by the concentration dependence of the lower yield stress and does, in fact, substantiate the annealing behavior of the upper yield stress.

In order to explain this shift, consider the lower yield stress to be unaffected by a defect concentration greater than a value $C_c$, which corresponds to that of a neutron dose of $1.5 \times 10^{17}$ reactor neutrons/cm². Assuming that the defect concentration $C$ is proportional to the neutron dose, a reasonable assumption for the doses considered, then a dose of $4 \times 10^{18}$ reactor neutrons/cm² will have a defect concentration of about $25\, C_c$. Consequently, the effects of annealing will not be observed until the defect concentration has been reduced to 1/25th of its initial concentration, pushing the recovery of the yield stress into the high-temperature tail of the annealing peak, which accounts for a temperature shift. With these assumptions, it was possible to synthesize the isochronal annealing curve from the temperature shift as a function of dose (See Fig. 9.).

An attempt to determine the activation energy was made. The crosscut method was used with isothermal annealing curves of the lower yield stress being measured. Samples were bombarded to $1 \times 10^{17}$ reactor neutrons/cm² so that the lower yield stress reflected the defect concentration. The isothermal

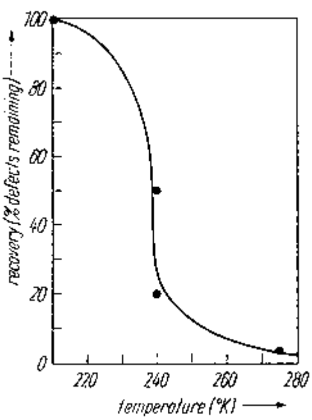



Fig. 9. An isochronal annealing curve of the defect concentration constructed from the dose dependence of the recovery as seen in Fig. 8

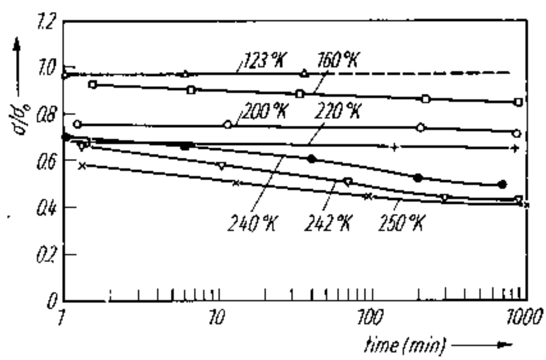
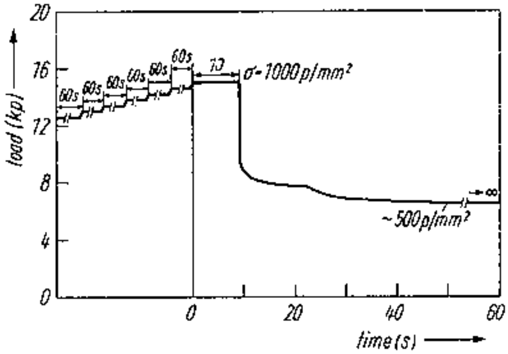



Fig. 10. Isothermal annealing curves of magnesium crystals irradiated to a dose of $1 \times 10^{17}$ reactor neutrons/cm². No upper yield was observed. The abscissa is the ratio of the yield stress ($\sigma$) to the initial yield stress ($\sigma_0$). All measurements were taken at 77° K. The ordinate is the annealing time $\sigma_0$ at liquid; nitrogen temperature as irradiated; isothermal annealing of 6 h irradiation at liquid nitrogen temperature

Fig. 11. Delayed yielding in magnesium crystals irradiated to $4 \times 10^{14}$ reactor neutrons/cm$^2$ at 77 °K. The sample was incrementally loaded with delays of 60 s between load increments until yielding occurred. Two distinct events occurred — each with a delay time of about 10 s. Note that the lower yield stress is the same order of magnitude in the stated test as in the dynamic case. The entire test was carried out at 77 °K



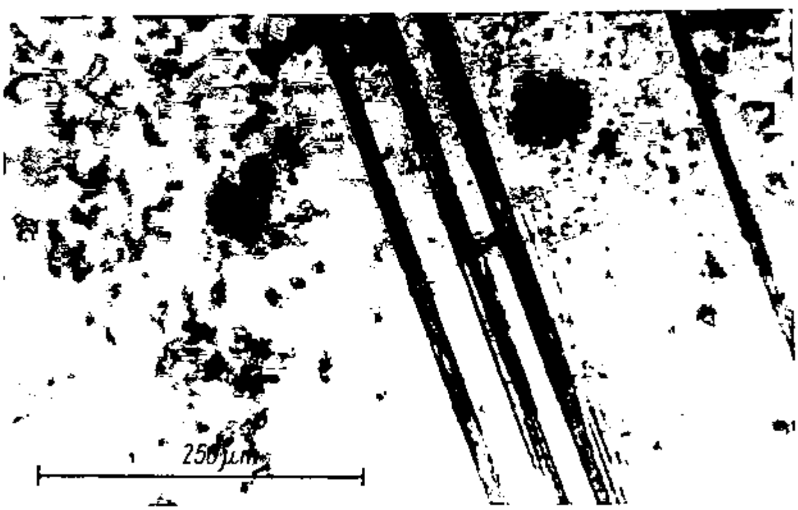




Fig. 12. The strain markings resulting from the yield drop shown in Fig. 11

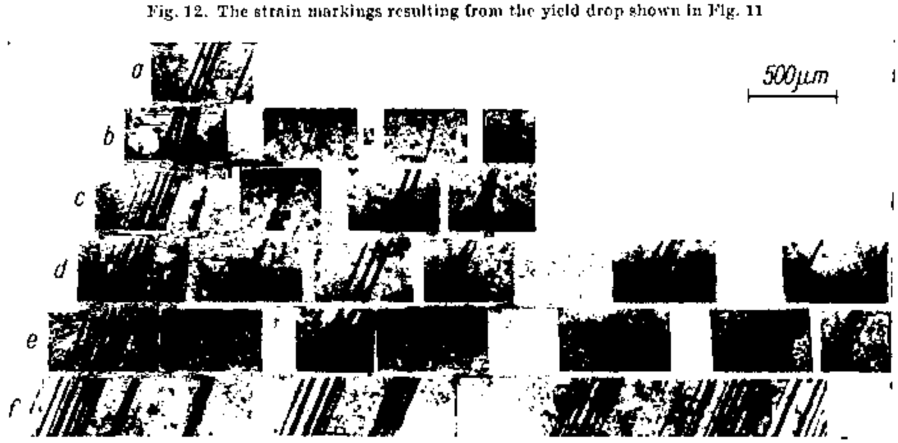



Fig. 13. Slip lines in magnesium irradiated to $4 \times 10^{14}$ reactor neutrons/cm$^2$ at 77°K as a function of the strain. The distance between successive markings is to scale. Deformation and photomicroscopy were performed at 77°K a) $\varepsilon = 0.13\%$, $\Delta l = 40$ µm, b) $\varepsilon = 0.36\%$, $\Delta l = 108$ µm, c) $\varepsilon = 0.77\%$, $\Delta l = 231$ µm, d) $\varepsilon = 1.0\%$, $\Delta l = 300$ µm, e) $\varepsilon = 1.6\%$, $\Delta l = 580$ µm, f) $\varepsilon = 2.0\%$, $\Delta l = 600$ µm

annealing curves are shown in Fig. 10. The shape of these curves makes it difficult to determine the activation energy, as it is hard to obtain overlap of the curves on a cut. It must be concluded that the activation is very high or the process is not uniquely activated. The latter conclusion is probably valid.

Some rough experiments were performed on the effects of crosshead speed on the yield stress. It was found that yield stress increased with the crosshead speed. This suggested that there might be a delay time in the yield point, and consequently, a search was made for it. The sample was loaded in increments with a waiting period of 60 s between each incremental increase in load. A delay time of the order of 10 s was observed. This experiment is illustrated in Fig. 11. Of particular interest was the second burst of slip lines that occurred some 10 s after the first with a stress of about one-half that of the yield stress.

Strain markings on the crystal were also studied. The photomicrographs of Fig. 12 were obtained after the time delay experiments of Fig. 11. It can be seen that four packets of slip lines were released during the two bursts, and most of the dislocations occurred during the first burst. Fig. 13 shows the progressive formation of the slip lines as a function of strain on a crystal bombarded to $4 \times 10^{18}$ reactor neutrons/cm$^2$. Notice that the strain is inhomogeneous, yet it does not behave like a typical Lüders band, as new slip line packets form at a distance of some 500 μm with satellite lines forming on the old packets well behind the new slip lines.

## 4. Discussion

The effects of irradiation on the mechanical properties of magnesium are obviously greatly different from those in face-centered cubic metals. Development of large yield drop induced by irradiation sets magnesium apart from aluminum, gold, silver, and copper. This phenomenon has most of the important characteristics of yield phenomena observed in body-centered cubic [5, 6] and hexagonal [7, 8] metals resulting from interstitial impurities, i.e. large yield drops (for larger neutron doses of the order of 50% of yield stress); a significant delay time; and, inhomogeneous deformation. However, there are other aspects of the yield of irradiated magnesium that differ from the interstitial doped body-centered cubic metals. For example, in magnesium, the dislocation locking occurs without aging, and after deformation its behavior is atypical as aging did not restore the drop. Furthermore, deformation, while inhomogeneous, did not have the usual Lüders band characteristics. Nevertheless, it can probably be assumed that the basic yielding mechanism in irradiated magnesium is the same as that in interstitial impurity doped hexagonal and body-centered cubic metals.

Then if this is the case, the yield drop in irradiated magnesium would be expected to be the result of dislocation locking by an atmosphere of irradiation-inducted defects. Since vacant lattice sites and interstitial atoms are created in equal numbers during this bombardment, the atmosphere could be caused by either of these defects. It can probably be concluded that the atmosphere is composed of interstitial atoms. The argument is as follows. Since the upper yield point (and therefore the locking) occurred at 77 °K without aging, the atmosphere must arrive at the dislocation either as a result of thermal migration of the defects or as the result of displacement cascades intersecting the dislocation lines. Since all the dislocations must be locked, and the probability of this occurring is small for the latter case at the doses considered, it seems reasonably certain that the defects responsible for this locking must be mobile at 77 °K.

Examination of isochronal annealing curves for resistivity changes in neutron-irradiated magnesium [9, 10] showed annealing stages at 50, 75, and 140 °K. Of these three, the highest temperature peak was the largest and showed second or higher degree order, as it shifted to lower temperatures with increasing dose.

Apparently, either or both the 50 and 75 °K peaks are the result of long-range defect migration, because measurements of the elastic modulus show that the dislocation mobility decreased as the result of neutron bombardment at 77 °K [9]. On the basis of this experiment, it is feasible to assume that defects are moving to dislocations at 77 °K or below. Consideration of the migration energy of the defects and the number of jumps involved would indicate that migration defects are interstitials.

It is difficult to understand why a further increase in yield drop did not occur when the specimens were heated through the 140 °K peak. Measurements indicate that the modulus increased as the annealing pulse went through the 140 °K peak, whereas the load drop was either unaffected or decreased. Furthermore, recovery of the modulus occurred between 500 and 600 °K, whereas the load drop for equivalent doses occurred at least 100 °K lower in temperature. It must be concluded that there is little correlation between dislocation mobilities determining modulus with those determined by yield drops.

The increase in the yield stress in low dose regions and the constancy of the lower yield point in high dose regions is difficult to understand in light of yield point studies. The lower yield stress is frequently interpreted as stress required to move the unlocked dislocation through the lattice [11]. The results of Fig. 10 would seem to support this viewpoint for it can be seen that in the first yielding process there is a tail in the load-elongation curve where dislocations continue to move at the lower yield stress. If this is taken seriously, then it can be deduced that for low concentrations ($< 1 \times 10^{17}$ reactor neutrons/cm$^2$) where the yield drop is zero, the defects impede the motion of an unlocked dislocation, as the stress is raised from $40 \pm 5$ p/mm$^2$ to as much as 465 p/mm$^2$ at $1 \times 10^{17}$ reactor neutrons/cm$^2$, whereas for doses above this value, defects lock the dislocation, as the upper yield increases. Apparently they do not further impede the motion of unlocked dislocations, as the lower yield stress remains at 465 p/mm$^2$. Therefore, it is difficult to avoid the conclusion that two different defect-dislocation interactions occur, one of which dominates at low defect concentrations, and the other at high defect concentrations.

Alternately, it can be assumed that the lower yield stress is a consequence of a stress magnification associated with the initial packet of the slip lines [12]. That is, the upper yield stress is required to move the locked dislocations, and the slip bands formed by this event act as a stress riser, enabling locked dislocating to subsequently move at a lower applied stress level. Since the back stress of dislocation pile-ups locks the dislocations generated in the slip lines, the only dislocations that are unlocked in any given instance are thus in motion. There is some evidence to support this model. For example, it was found in the pulse annealing experiments that the upper yield stress could not be restored by aging, implying that there were no unlocked dislocations available. Furthermore, the shift in annealing temperature of the lower yield stress with the dose offers collaborating evidence, as it suggests that defects were acting as locking atmosphere rather than as an impediment of their motion. While the uniform spacing of the new packets shown in Fig. 12 would tend to support a uniform lower yield stress, the large spacing between new slip packets makes it difficult to account for the magnitude of the stress magnification.[2])

All of this means that the sharp yielding phenomenon and yield drop are really not too well understood. It is hoped that irradiation effects in magnesium might ultimately prove to be a useful tool to study this phenomenon.

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[2]) Haasen and Leibfried [13] pointed out that a maximum stress magnification would occur at a distance from the slip line, which is about the same as the diameter of the dislocation pile-up. Something of this nature could account for these results. However, the separation between slip packets should then decrease with dose. Limited evidence suggests that this condition is not satisfied.