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THE USE OF ACCELERATORS TO STUDY IRRADIATION EFFECTS IN MATERIALS

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Summary

There is currently high interest in the use of accelerators to study irradiation effects in materials to be used in reactors. The effects produced by heavy ion irradiation (e.g., nickel ions irradiating stainless steel) and fast neutron irradiation are qualitatively similar. Time to achieve the damage state characteristic of end-of-life service is hours as compared to several years in available reactors. Some of the successes in this area are described herein as well as some of the many problems to be solved in order to establish quantitative relationships.

Introduction

The bombardment of solids with high energy particles causes significant changes in many important engineering properties and are of great concern to designers of reactors (both fission and fusion). Most of the changes can be attributed to the ultimate disposition of the atoms which are displaced from their normal lattice sites. Many come to rest in interstitial positions, leaving vacant lattice sites behind. If these vacancies agglomerate into voids (three-dimensional pores containing less gas than necessary to balance the surface tension) and the interstitials precipitate as dislocation loops, significant swelling occurs in fuel element cladding and reactor core structural components.

The economic consequences of swelling in fuel cladding and fuel duct material are detailed in a report prepared by the Bethe panel.[1] This group concluded that a solution to the swelling problem would shorten the doubling time of the liquid metal fast breeder reactor by a factor of two. Radiation-induced voids were first reported in neutron-irradiated stainless steel,[2] quickly followed by their detection in other metals and alloys. They typically exist in concentrations of 10^{20} to $10^{22}/m^3$ and are in the size range of tens to a few thousand angstroms (see Fig. 1). Void formation causes swelling due to precipitation of the displaced interstitials, and changes in mechanical properties. These changes have produced significant effects in research reactors after several years of operation.[3]

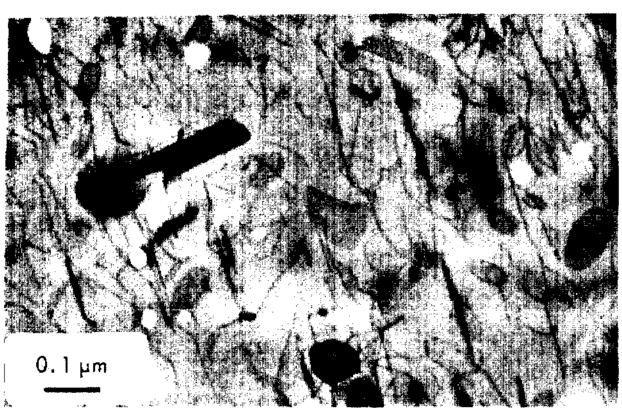

Fig. 1. Transmission electron micrograph showing voids and interstitial dislocation loops in type 316 stainless steel irradiated to $2 \times 10^{[illegible]}$ n/m^2 in EBR-II.

Simulation of Neutron Damage

The current high interest in accelerator-produced damage is a result of the time scale for selection of new cladding and structural materials for fast breeder reactors and the fact that there is no intense neutron source having a spectrum to be expected in a fusion reactor.

It is now realized that materials must be examined after irradiations corresponding to about 140 displacements per atom (dpa) for some components of fast reactors. Such doses can only be obtained by irradiation times as long as five years in the highest flux fast test reactors available. Even experiments designed to define trends in the behavior of commercial alloys which are conducted in EBR-II have a three- to four-year turnaround time. Hence, there has been a renewed interest in the use of heavy ion irradiations in accelerators. Fortunately, new techniques have been developed that were not available in the 1950's for examining specimens. As examples, there are transmission electron microscopy (TEM), small angle x-ray and neutron scattering (SAXS, SANS), and positron annihilation. These techniques allow one to examine specimens which are of the order of 1000 to 2000-A thick.

Radiation damage produced in metals during fast neutron irradiation can be qualitatively simulated by irradiation with energetic ion beams from accelerators.[4,5] The most pressing requirement is the establishment of quantitative relationships among the various kinds of irradiations.[6] There is need for both theoretical and experimental studies.

A fundamental requirement for the use of accelerators in any study is the production of a region of uniform damage within a metal specimen while the specimen is maintained at a selected temperature. The fluence (or dose) must be sufficient to create a comparable number of lattice defects within a given region to that produced in reactor components during fast neutron irradiation. However, the total irradiation time need be no longer than can be achieved conveniently using existing accelerators. An additional requirement is the demonstration that the damage region is not affected by the surface and the results are representative of the bulk material. The bombarding ion species must be chosen such that it does not produce adverse chemical or physical effects within the sample which might influence the formation of the neutron irradiation microstructure.

For neutron damage simulation, it is necessary to account for the transmutation gases which are produced within the material, for example, as a result of (n,γ) or (n,p) reactions. Trace quantities of helium, for example, should be uniformly implanted throughout the sample simultaneous with the damaging ions. If a dual source for simultaneous injection of helium is not available, a reasonable compromise is the sequential irradiation by the damaging ions and helium. However, most experiments to date have injected the total helium content (typically 10 to 30 ppm) in a separate irradiation before the displacement irradiation. There are now data which show that the early stages of the damage structure differs in the two cases (prior and simultaneous). The importance of these differences at high fluences has not yet been established.

Correlation or Calibration Studies

Some of the problems in development of a quantitative correlation among irradiation effects produced by bombardment with particles of varying species and energies will be briefly described. Most of the theoretical and experimental efforts of a fundamental nature have been directed towards determination of the number of point defects produced. Robinson[7] has calculated that approximately four times as many displacements are generated in some materials by 14-MeV neutrons as by 1-MeV neutrons. Logan[8] calculated that 16-MeV protons will produce displacement cascades having an energy spectrum that is similar at its high energy end to that produced by 14-MeV neutrons. A large fraction of the displacements are produced by lower energy cascades, however, which are not present in 14-MeV irradiation. There are a number of experiments planned or under way to measure point defect production by fission spectrum neutrons, 14-MeV neutrons, protons, electrons, self-ions [ions of the same element(s) as the target]. These experiments typically employ dilute alloys irradiated at about 4.2°K, and measure the increase in electrical resistivity as a function of flux, fluence, type of particle, and energy.

The crucial questions are: What is the nature of the displacement cascade? How many defects survive at elevated temperatures? What is the configuration? Most data thus far come from either screening tests or examination of irradiated reactor components. For example, Fig. 2 illustrates the temperature dependence of swelling for fast reactor (EBR-II) irradiation and 5-MeV nickel ion irradiation.[9] The amount of swelling is a gross measurement of the survival of point defects as voids and interstitial dislocation loops.

Figure 3 shows a summary of swelling as a function of the number of displacements per atom for several bombarding species.[10] The major differences can be accounted for by considering the survival probabilities due to various cascade configurations.[11]

Evaluation of just how well such experiments agree with each other and with models is complicated by an incomplete understanding of the effects of alloying elements (including impurities) and microstructure on the kind and amount of damage. For example, "high purity" 316 stainless steel (i.e., a Fe-Cr-Ni alloy) swells about fifty times as much as a commercial 316.[11] Variations in composition of type 316 stainless steel from one production lot to another although within nuclear reactor composition specifications, result in factors of 5 to 10 difference in swelling behavior. The complex questions raised by such results make apparent the need for careful experiments in which the large number of variables are considered first individually and then in various combinations.

Physics of Particles in Solids

Many details of the interaction of energetic-charged particles with crystals are still poorly understood. The step from understanding the basic physics of energy loss and partition, defect production and distribution, etc., to the description of what damage survives and in what form is large and will require an interdisciplinary effect between atomic physics and materials science.

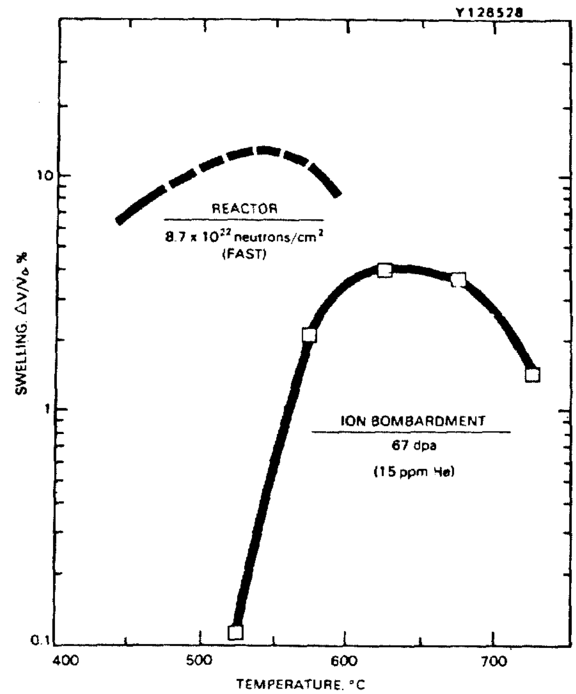

Fig. 2. Temperature dependence of swelling in annealed-type 316 stainless steel bombarded with 5-MeV nickel to a damage level of 67 dpa. The steel contained 15 ppm of cyclotron-injected helium. The temperature dependence of EBR-II core swelling is shown by the dashed curve. The displacement rates were $\sim 10^{-6}$ dpa/sec in EBR-II and 2×10^{-2} for the ion bombardment. (Johnson et al.[9])

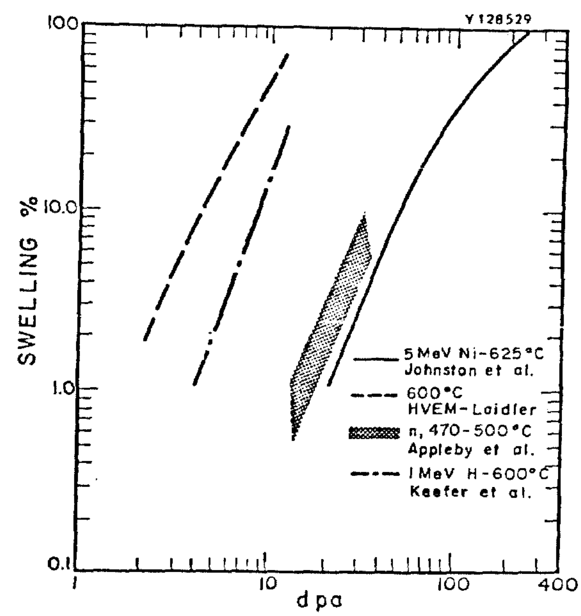

Fig. 3. Comparison of neutron-induced swelling in 316 stainless steel to that determined by H^+, Ni^+, and HVEM simulation studies.[10]

Production of Defects

One may employ accelerators to produce well-characterized defect structures for further study without regard to whether or not this simulates neutron damage. If reproducible numbers of defects can be created in similar configurations, many experiments can be conducted to relate physical, chemical, and mechanical properties to them. The manner in which metallurgical variables influence the defect state can also be studied.

Irradiation by electrons is especially useful in this type of study. Electrons with 1-MeV energy can displace atoms from their lattice positions in most metals with atomic weights less than about 110. Electron bombardment experiments permit the determination of the energy required to remove an atom from its equilibrium position. Another important use of electrons lies in the fact that as long as the energy of the electrons is close to the displacement threshold, single vacancy-interstitial pairs are formed. Thus, many radiation-induced phenomena can be analyzed in terms of isolated defects, and this avoids the complication attendant upon the generation of complex damage regions that occur in neutron and heavy charged particle irradiation. High voltage electron microscopes have been recently used in this fashion since they allow the in situ observation of the damage once the damage state (cluster of defects) reaches a size of 25 to 50 A. Electrical resistivity techniques are generally used to study the earlier stages, and have the advantage of being sensitive to low concentration of isolated point defects.

At high energies (25 MeV) in the electron accelerators, one may expect to generate some clusters of defects or cascades. The transition from single defect production to multiple defect production in these experiments can give important information for all types of irradiation.

Accelerator Probe Techniques

An area in which there are many opportunities for technique development and application is the use of accelerators as probes for analytical work. Some applications are based on scattering processes such as $(\alpha$-$\alpha)$, (p,p), (p,α) and channeling and blocking experiments. Among the major unknowns connected with radiation effects is the form and distribution of the transmutation gases (He,H) and how these change with time, temperature, and amount of damage. With the achievement of suitable sensitivity and resolution, such probes offer hope of defining the behavior of such gases. In controlled thermonuclear reactors (fusion), transmutation gases will be produced at rates which are orders of magnitude greater than in fission reactors and may be the limiting factor in determining the lifetime of reactor components. A knowledge of their distribution might enable one to design around their effects or to manipulate metallurgical parameters to put the gases into less detrimental forms.

Characteristics of Radiation Effects in Metals

Void formation occurs at temperatures where both vacancies and interstitials are mobile. Any point defect may be annihilated by recombination of vacancy-interstitial pairs or by absorption at a sink such as a void or a dislocation. During constant irradiation, quasi-steady state is reached in which the loss rate equals the creation rate.

Gas atoms are thought to stabilize a void in its very early stages of life; hence, transmutation reactions giving helium or hydrogen play a role. The association between the quantity of helium generated and the magnitude of swelling observed is an indirect one. Voids can form in the absence of helium; however, many investigators believe other gaseous impurities are involved in the nucleation step.

Voids can be produced in most metals and alloys and with any type of irradiation that displaces atoms. A useful and descriptive unit for representing a quantity of irradiation is in units of displacements per atom (dpa), the average number of times that each atom in the sample is displaced. The conversion of a neutron or other particle fluence ϕt, where ϕ is the flux and t the time, into a displacement dose by uniform procedures is important.[7] The problem is to determine the equivalence of a given particle flux to dpa and, therefore, into equivalent damage per unit flux.

The neutron fluences needed to produce voids in stainless steels, $\sim 10^{25}$ n/m^2, can only be obtained conveniently in a fast reactor. Voids occur in most pure metals, on the other hand, at relatively low fluences, $\sim 10^{24}$ n/m^2; it is practicable to investigate these using thermal test reactors. Factors such as neutron spectrum, dose rate and rate of helium production are important and care must be taken when comparing thermal and fast reactor data.

The temperature dependence was one of the first characteristics of void formation to be discovered. It was generally observed that void concentrations decreased with increasing irradiation temperature and that average sizes increased. Swelling occurs only in a limited temperature range and is a maximum at some intermediate temperature. Void shape is also affected by irradiation temperature. Other irradiation variables thought to be important include total dose (fluence), dose rate, and stress state. Most materials variables (composition, internal microstructure, etc.) seem to influence the damage state produced; hence, close attention must be given to these factors.

From an application point of view, alterations of mechanical properties during irradiation rank with dimensional instability as a major concern.

There have been hundreds of papers published describing the effects of irradiation at low temperatures, below 0.3 T_M (T_M = melting point on absolute temperature scale) on mechanical properties. The general understanding is summarized in reference 12. Irradiation and testing in this temperature range shows large increases in yield stress, small increases in ultimate tensile strength, and large decreases in both uniform strain and work-hardening coefficient.

The irradiated structure is characterized by point defect clusters whose size depend on the irradiation temperature. These clusters are strong barriers to dislocation motion, thus causing the large increase in yield strength. Transmission electron micrographs show that once dislocations move, they sweep out the defect clusters in narrow channels. At this point, one has bands of very hard and very soft material. Further deformation proceeds by dislocations moving in these soft channels with a minimum of interaction. The result is highly localized deformation, very low macroscopic or uniform elongation and little work hardening.

At deformation temperatures greater than about 0.5 T_M the prevailing fracture mode is along grain boundaries. Irradiation in this temperature range permits the transmutation gases to collect in grain boundaries. These gas bubbles decrease the ductility by acting as crack nuclei and may act as sources of internal stress along the grain boundaries.

At temperatures $\lesssim 0.5\ T_M$, stresses less than the postirradiation yield stress and at neutron fluxes $\gtrsim 10^{17}$ n/m^2 sec (E > 0.1 MeV), metallic materials often exhibit enhanced deformation when tested during irradiation compared to either unirradiated or post-irradiation deformation. This radiation-enhanced deformation (irradiation-induced creep) concerns reactor designers not only because it may lead to different in-reactor material configurations but because the conditions under which it occurs are those where radiation-induced void formation is prevalent. The material swelling which accompanies void formation introduces stresses, in addition to those present due to thermal and mechanical conditions, which may be relaxed by irradiation creep. On the other hand, stress may influence the swelling process. Thus, it has been considered that the processes of irradiation-induced creep and void formation are closely inter-related.

Experimental Requirements for Use of Accelerators

General

Since the radiation effects problems of most immediate concern are associated with the fast breeder reactor, nickel ions are perhaps satisfactory for the production of displacement damage in stainless steels and nickel-base alloys. However, it must always be remembered that an imbalance is created in the total vacancy and interstitial content within the specimen. In a typical case where one extra interstitial is created for every 10^4 vacancy-interstitial pairs, the addition of the extra interstitial can be neglected if the temperature is low enough that significant recombination of the defects does not occur and if the irradiation-induced dislocation density is sufficiently high. These two conditions are generally met in experiments which attempt to simulate in-reactor behavior. The fact that the region of maximum displacement damage is slightly removed from the end of the ion range gives some relief toward upsetting the point defect balance.

The major disadvantage of nickel ion irradiations is the short ion range. Lighter ions can be used; longer range is gained at the expense of lower displacement rates and possibly significant changes in specimen composition or purity. For example, carbon ions have been used to irradiate steels but the carbon content is significantly altered at high fluences. Proton irradiation raises the question of possible effects of hydrogen on phase stability, defect nucleation, and mechanical properties. However, all attempts to detect residual hydrogen in proton irradiated stainless steel have failed, a result interpreted to mean that the hydrogen rapidly diffuses out of the sample.

During the next few years increasing consideration will be given to the use of refractory metals in fusion reactors. In anticipation of such studies, sources should be developed for such species as vanadium, niobium, zirconium, and molybdenum. Design of experimental arrangements which will allow the simultaneous accumulation of displacement damage and transmutation products in the same volume will be necessary. For example, a desired, but perhaps impossible, experiment for evaluation of the use of niobium in fusion reactors requires niobium, zirconium, helium, and hydrogen to be deposited in a fixed ratio in a given volume.

For nickel ion bombardment of nickel or stainless steel, beam currents of 0.5 to 2 μA/cm^2 at energies of 5 MeV or higher are required. Higher currents give problems due to heating and lower ones require excessive machine time. On the other hand, lower currents can be used for the study of the basic damage mechanisms (e.g., changes in resistivity by bombardment at 4.2°K) since the fluences needed usually are several orders of magnitude lower.

The time structure of the beam may be important and should be defined. Its possible importance arises from the interstitial to vacancy ratio achieved under different irradiation conditions. Fast reactors and some fusion reactors will have displacement rates of about 10^{-6} displacements per atom per second; whereas charged particle bombardment gives rates of the order of 10^{-4} to 10^{-2}. This presents a major problem in deciding just how accurate is the simulation of reactor irradiation which must also be considered when we evaluate the importance of the time variation of cyclotron beams (~ 10 MHz), the rocking of specimens to spread the damage over a thicker region, or a scan of an area by rastering the beam. There is experimental evidence that such rate effects cannot be ignored.[13]

If the time variations in beam energy and/or current are important, then experiments in support of the controlled thermonuclear reactor program must be planned to take account of the relaxations associated with the mode of reactor operation. The "steady-state" reactors such as the Tokamaks may operate on a cycle with a long "burn" period (100 to 1000 s) and a short "cool" period (~ 10 s), whereas the pulsed reactor θ-pinch may have a burn of 1 μs to 70 μs followed by approximately 1 to 10 s cooling time. Laser systems have displacement reactions lasting ~ 1 μs and those bursts are repeated up to 10 times per second. The instantaneous displacement rates in the first wall of the θ-pinch and some concepts of the laser fusion reactor thus will be of the order of 10^{-4} to 10^{-1} dpa/s. The time averaged neutron fluences for both systems are about the same.

In all experimental arrangements, special attention must be given to an analysis of the ion species in the beam and to measurements of current (preferably continuously at the specimen), energy, specimen temperature, specimen environment, and fluctuations of these parameters. In the case of refractory metals being studied at temperatures of 600 to 1000°C, pressures lower than 10^{-8} torr are required.

Mechanical Properties

There are at least two methods for the attempted simulation of irradiation creep. In one, the accelerator is used to produce both the damage structure and the displacements in a specimen held under stress at an elevated temperature.[14] Production of the damage structure in this case may give information on swelling under stress, an important investigation itself. A second approach is to produce the damage state by reactor irradiation and use the accelerator to produce displacements for a relatively short period. This method approximates "instantaneous" measurements of strain rate at various points in the irradiation history of the material.

In some experiments of this type, the cyclotron beam (besides producing the required defects) will be the main source for heating the sample to desired temperatures. Since the anticipated creep strains are of the same order of magnitude as the thermal expansion due to a temperature change of 1°C, the temperature of the sample must be controlled quite precisely. The necessary temperature stability may be achieved by passing an electric current through the sample. Since the ohmic heating is about 10% of the heat input from the cyclotron beam, beam current fluctions of at most 10% can be balanced by an internal control system. Furthermore, smaller beam fluctuations in the frequency range between 20 and 2 Hz will be difficult to compensate because of the time constants in the control system (fluctuations with frequencies > 20 Hz will be averaged out by the sample itself which has a thermal time constant of about 0.1 s). It therefore appears necessary to control the beam current within ± 1% in the frequency range between 2 and 20 Hz and within ± 5% for slower variations. Since a typical duration of creep experiments is expected to be 10 to 20 h, the beam stability should be maintainable over at least this period. These are cyclotron operating conditions that have not in the past been demanded by nuclear physicists, and they impose new requirements on the accelerators.

Surface Effects in Fusion Reactors

In addition to problems caused by the large amounts of transmutation-produced gases, that will be distributed relatively uniformly throughout the wall, problems may arise from gas atoms which reach the wall from the plasma. The D-T fusion reaction yields a 14.1-MeV neutron and a 3.52-MeV alpha-particle. Most Tokamak conceptual designs of fusion reactors include a divertor to prevent most of the alpha-particles as well as other constituents of the plasma from reaching the wall. However, some of the ions which leak from the plasma will not be collected by the divertor. The University of Wisconsin fusion reactor reference design (UWMAK-I)[10] estimates that 10% of such ions will reach the first wall. For a neutronic wall load of 1.25 Mw/m^2, the particle fluxes and energies are calculated to be:

	(kev)	(particles/cm²/sec)
Deuterium	23	6.4×10^{13}
Tritium	23	6.4×10^{13}
Helium	23	4.7×10^{12}
Helium	> 23 to 3500	1.7×10^{11}
Metal Ions	23	2.5×10^{12}

Since the ranges of such particles are a few thousand angstroms they will come to rest in a region near the inner surface of the wall. The presence of such particles may cause at least two phenomena which can lead to wall erosion and contamination of the plasma: sputtering and blistering. There is a significant body of literature on sputtering and some data on blistering. However, the complete range of conditions and materials have not been covered and some of the results are clearly erroneous due to contamination of the specimens by carbon, oxygen, and perhaps other impurities in the accelerator beam or specimen chamber. Accelerator probe techniques can be used to study the migration of implanted ions and to characterize their distribution. The re-emission of the gases by either diffusion to the plasma-wall surface or rupture of the blisters must be determined.

The size, density, shape, and critical fluence for the formation of blisters appear to be functions of: (1) energy of ions (i.e. range); (2) diffusivity of the injected ions; (3) solubility of the gases; (4) strength of the wall material; (5) temperature; (6) flux of ions; (7) fluence; (8) crystallographic orientation; and (9) metallurgical state of the metal. There is not now a comprehensive theory to predict the extent of erosion due to blistering.

Conclusions

Although there are a number of materials programs using accelerators in progress and there has been some success in relating effects due to neutron irradiation and acceleration irradiation, there is need for much more. Quantitative relationships must be developed and innovative experiments designed to answer the many remaining questions.

Some specific materials properties and the role and requirements for accelerator study are listed below. In general, beam currents of 0.5 to 2 $\mu A/cm^2$ are desired for a specimen area of 0.5 to 1.0 cm^2.

Property	Role of Accelerator	Accelerator Requirements
1. Swelling	Production of displacements; introduction of transmutation products	Simultaneous or sequential irradiation by self-ions and transmutation products; energy > 1 MeV
2. Mechanical Properties	Simulation of damage structure; simulation of irradiation field (i.e. displacements)	Stable beam; high energy particles (α's 40 to 60 MeV) instrumented facility
3. Structural and Composition Changes	Composition changes by ion implantation; displacements	Simultaneous injection; ion implantation—uniform and as deep as possible; high energy particles; backscattering experiments
4. Surface Effects	Implantation of gases near surface	D,T,α, self-ions $\sim$ 20 keV α to 3.5 MeV; backscattering experiments

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