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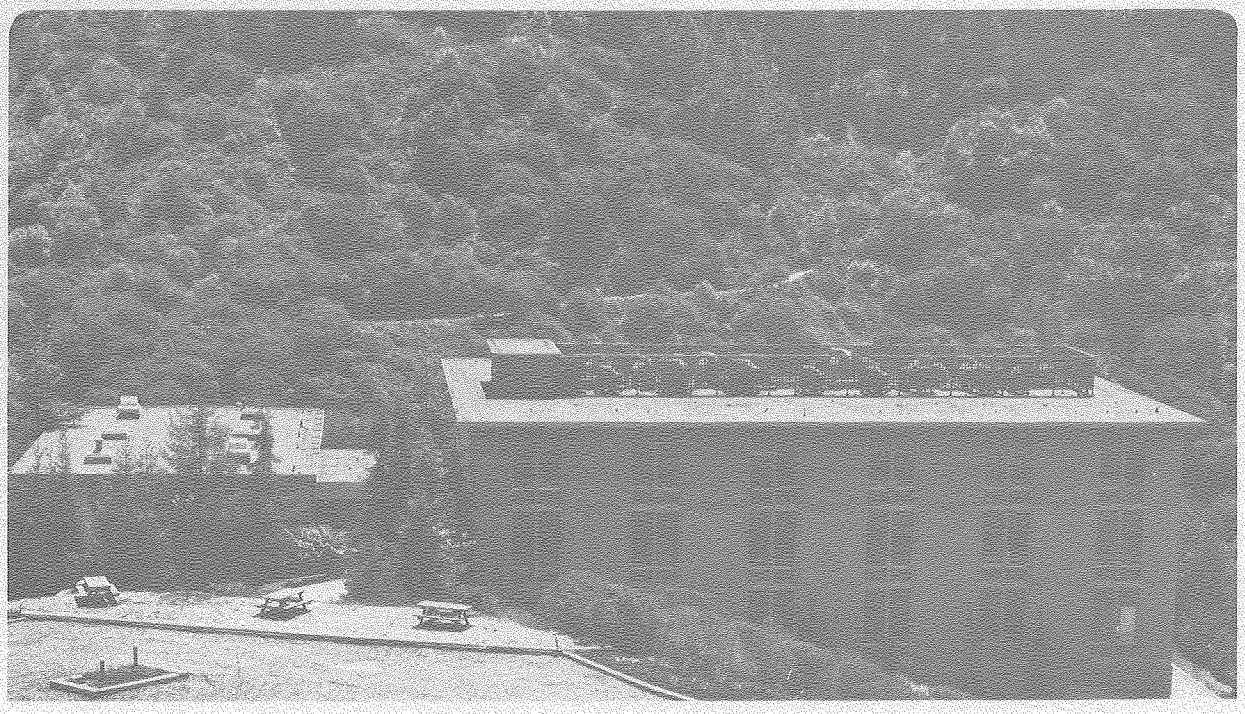

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The Materials of Fast Breeder Reactors

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The most difficult problems affecting commercialization of the liquid-metal fast breeder reactor (LMFBR) concern the behavior of its materials of construction in the thermal and irradiation environment in which the device must operate. The responses which these conditions provoke in the fuel, cladding, and core structures depend upon complex interactions of many physical and chemical processes.

At the heart of the fast breeder reactor, and of the debate it has generated, is the nuclear fuel which powers the fission process. The material which comprises the core of a fast breeder reactor must function in the most inhospitable environment that any technological device has ever had to withstand. Temperatures high enough to melt all but a few substances and intense bombardment by high-energy nuclear particles require that nuclear fuel elements be constructed of the most refractory materials and be designed with Spartan simplicity. The nuclear fuel is a ceramic oxide containing the uranium and plutonium needed for the nuclear reactions; plutonium-239 fissions upon neutron capture to produce the heat which is ultimately converted into electricity (Fig. 1). The uranium-238 in the fuel serves to regenerate plutonium by neutron-absorption and subsequent radioactive decay. These two heavy metals are combined chemically with oxygen because the oxide has a sufficiently high melting point and structural stability to survive the designated lifetime in the reactor. The ceramic fuel is contained, or clad, in a close-fitting stainless steel tube. These two components, ceramic fuel pellets and a metal cladding tube, constitute a nuclear fuel element.

The fuel elements for the present generation of light-water reactors differ from those used in fast breeder reactors in the materials of construction; the fuel in a light-water reactor is pure uranium dioxide (enriched in fissionable ^{235}U) and the cladding is an alloy containing primarily zirconium. The principal neutronic difference between a fast breeder reactor and a light-water reactor is the energy of the neutrons which cause fission in the fuel. The average energy of the neutrons in a light-water reactor is about 0.1 eV, whereas the neutrons in a fast breeder reactor retain nearly the energy they had on emerging from the fission process, or about 1 MeV. The efficiency

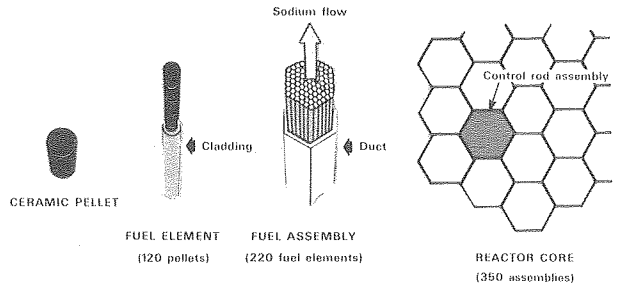

Fig. 1. The basic units of a fast breeder reactor core are pellets of ceramic oxide about 6 mm in diameter and 10 mm in height. Half of the pellets are pure $^{238}UO_2$ which captures neutrons and breeds ^{239}Pu. The remainder of the pellets contain approximately 15% plutonium oxide and 85% uranium oxide ("mixed oxide"). The pellets are stacked into a thin-walled (0.4 mm) stainless steel tube which is then welded closed under an atmosphere of dry helium. These fuel elements are loaded into a long hexagonal duct to produce a unit called a fuel assembly. The assemblies are arranged in a honeycomb pattern to form the core of the reactor, which has a diameter of several meters and contains 25 tons of the heavy-metal oxides. Special assemblies are fitted with control rods containing neutron-absorbing boron carbide instead of fuel. Axial movement of the control assemblies into and out of the core provides the nuclear reactivity changes needed for startup, power changes, and shutdown. Liquid sodium coolant flows upwards through each assembly, rising in temperature from ~300 to ~500 °C at the outlet. The hot sodium from the core is piped to an intermediate heat exchanger where it gives up some of its energy to a secondary sodium loop which then circulates through a steam generator. The reactor, intermediate heat exchanger, steam generator and the primary and secondary sodium loops constitute the nuclear steam supply for the electrical generating plant

of thermal neutron fission of the actinide elements is over 100 times larger than it is for high-energy neutrons. Consequently, in order to sustain the same fission rate per unit volume of fuel (or power density), the neutron flux must be one hundred times larger in a fast reactor than in a thermal reactor, yet for economical operation, the fuel elements in both reactor types should remain in the reactor core for about the same length of time (one to two years). This requirement automatically insures that the structural metals in a fast reactor will be subject to far more severe radiation damage than the materials in a light-water reactor; whatever the ultimate effect of radiation on metal, be it hardening, void formation, or helium embrittlement, the magnitude of the property change or the severity of the radiation effect is a strong function of the fast neutron fluence, which is the product of the flux and the time. Moreover, the production of economically competitive electric power from a fast breeder reactor requires that the power density in the fuel be four or five times greater than that characteristic of light-water reactors. This requirement can be met only by the use of fuel elements in fast reactors which are roughly one half of the diameter of light-water reactor fuel elements and by operating some of the fuel at temperatures close to its 2800 °C melting point. The high temperatures and large temperature gradients implied by this type of operation, when superimposed upon the fission process, produce novel phenomena such as gross restructuring of the ceramic, radial redistribution of its component chemical elements and substantial swelling due primarily to precipitation of insoluble fission gases. Economics also demands a fuel burnup (fraction of heavy metal fissioned) approaching 10% for fast breeder reactors, compared to the 2% burnup characteristic of light-water reactors.

The principal result of the fundamental difference in the nuclear physics of the thermal and fast neutron reactors and the higher operating temperatures and greater fuel burnup in the latter is an array of materials and compatibility problems which are an order of magnitude more difficult to rectify by design or by materials selection. In addition, the high fuel power density and the compact core of a fast breeder reactor place severe restrictions on the type of coolant which can be utilized to extract the nuclear heat. Although some research is being conducted on fast breeder reactors cooled by high-pressure helium, those under development in all highly industrialized countries of the world employ liquid sodium as the coolant, whence the name liquid-metal fast breeder reactor (LMFBR).

Despite the simplicity of a nuclear fuel element, both in design and in materials, long irradiation at high temperature produces a bewildering variety of physical and chemical changes. Because these alterations have a profound impact on the integrity of the fuel element, a great deal of money and effort has been invested in attempting to understand the phenomena involved and to develop methods for predicting the changes induced by irradiation. LMFBR funding is the largest single item in the US Department of Energy budget, and a substantial portion of this money is devoted to irradiation testing and mathematical modeling of reactor fuel elements. The primary purpose of the only fast breeder reactor under construction in the United States, the Fast Test Reactor at the Hanford Engineering Development Laboratory, is to conduct irradiation experiments on fast breeder fuel elements (Fig. 2) and other core structural components. Fully instrumented tests are very costly, and because of the complexity and number of physical and chemical processes which occur, a complete un-

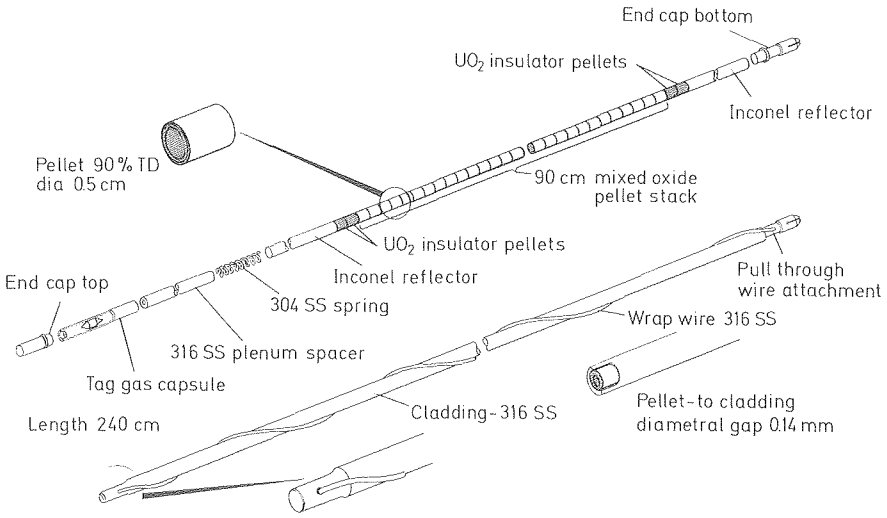

Fig. 2. Fuel rod for the Fast Test Reactor currently under construction at the Hanford Engineering Development Laboratory near Richland, Washington

derstanding of the evolution of a fuel element subject to irradiation at high temperatures cannot be achieved by experiments alone. Consequently, the same processes are simulated mathematically by large computer programs called fuel modeling codes. These codes utilize analytical models of each of the basic irradiation-induced processes which are derived from the fundamental physics and chemistry of the phenomena involved. The conditions which the analysis must be capable of treating range from average steady-state operation (with intermittent thermal cycling due to reactor shutdowns or power changes) for periods of up to two years to millisecond time scale transients preceding the hypothetical core-disruptive accident (HCDA). Fuel temperatures from 300 °C to the critical point (> 5000 °C) can occur; stresses ranging from zero to the yield stress of the cladding and to the fracture stress of the fuel will be encountered; the corresponding strain rates range over six orders of magnitude. The fuel modeling code permits all relevant processes to take place simultaneously (as they do in a real fuel element) in an effort to properly simulate their interaction. The calculation attempts to translate the cumulative effects of the many individual processes into gross property changes in the fuel element, such as pressure buildup due to fission-gas release, and stressing or straining of the cladding. Finally, the code provides a prediction of cladding failure.

It might appear that the condition for failure should be straightforward. Large structures such as bridges are considered to fail if at any instant the stress in any one of its members exceeds the stress which causes irreversible plastic deformation (yielding). However, a breach of the cladding of a fuel element is most likely to be the result of a different mechanical process, namely creep. Creep is slow plastic deformation which can occur at stresses well below that required for instantaneous yielding. Creep is important for high-temperature operation (above approximately one half of the melting point of the metal). Creep rupture is the principal failure mechanism in high-temperature turbines, jet aircraft engines, and nuclear reactor fuel elements. An unambiguous criterion for fuel element failure by creep has not yet been devised. Earlier analyses utilized a simple strain limit, which restricts the fractional change in the diameter of the cladding to a specified amount, say 0.3%. A more realistic criterion is based upon what is termed a damage summation rule. The concept is based upon the established fact that a metal held at fixed stress and temperature fails in a well-defined time. This lifetime is reduced by larger stresses or higher temperatures. In a reactor fuel element, the temperature and stress are changing with time and fast neutron bombardment is continuously altering the mechanical properties of the cladding. To account for these changes, the operating history of a fuel element is divided into short intervals over which stress and temperature conditions and the mechanical state of the metal are approximately constant. In each of these time segments, a calculable fraction of the total rupture lifetime is consumed. These fractions of a lifetime are added up as irradiation proceeds, and when the cumulative fraction reaches unity, failure is considered to occur. Thus the name damage summation rule, or alternatively, cumulative life fraction rule.

Still other failure criteria may be more relevant to the cladding of a reactor fuel element than the damage summation rule. Failure of a stressed metal can occur prematurely in the presence of particular chemical substances which accelerate the growth of microcracks at the surface. This phenomenon, called stress corrosion cracking, has been implicated in recent instances of cracking of external piping in several U.S. light-water reactors. In these cases, the active chemical ingredients are dissolved ions in the cooling water which the pipes contain. Stress corrosion cracking of fuel element cladding, however, occurs on the inside of the tube and is initiated by one or more fission products which have been released from the hot fuel. Fission product iodine has been singled out as the most likely corrosive agent which, when acting in concert with stress in the cladding due to internal fission-gas pressure or to contact with the swelling fuel, produces pinhole perforations in the metal tube.

Whether based upon experimental tests or analytical models, successful fuel element design ultimately reduces to predictability – of being able to affirm with quantitative confidence that a potential design will remain intact for the reactor exposure to which it is to be subjected. The object of the analysis is to determine *when* the fuel element fails under normal steady-state operation and *if* it fails in one of several transient situations specified by regulatory guidelines. Failure is manifest by rupture of the cladding, or in the more severe hypothetical accident transients, by melting of both fuel and cladding.

Companion analyses to fuel element modeling are concerned with the stresses and deformations in the structural components of the core, principally the fuel assemblies. These components can elongate or bow in a manner which adds nuclear reactivity to the core or impedes removal or insertion of control assemblies.

Fuel

All LMFBR's under construction utilize uranium-plutonium oxide as fuel. Advanced reactor concepts

envisage using the carbo-nitride of uranium and plutonium because the higher thermal conductivity and greater heavy-metal density of this type of fuel permits higher power density at considerably lower fuel temperatures than obtainable from oxides. Because of the low thermal conductivity of the heavy-metal oxides (UO_2 is a better insulator than alumina), removal of fission heat requires central temperatures in the most highly rated fuel pins which approach the melting point of 2800 °C. In fact, avoidance of fuel melting is the limiting condition of LMFBR fuel element performance.

The high power density of the fuel, in conjunction with the small radius of the fuel rod and the efficient convective heat removal by the flowing coolant, produces the largest temperature gradients and highest temperature levels ever sustained by a commercial technological device. The temperature gradient in the fuel, which can approach 10000 °C/cm, causes rapid migration of the original porosity in the as-fabricated fuel radially inward (Fig. 3). The mechanism of pore motion is simple vapor transport; the solid on the hot face of a pore in a temperature gradient exerts a higher vapor pressure than does the solid of the cold face. The oxide vapors diffuse from the hot face through the inert gas trapped in the pore (the helium filling gas or the fission gases xenon and krypton). This radially outward flow of matter causes the pore to move in the opposite direction. The net result of pore migration is the appearance (probably in a matter of hours after reactor startup) of a central hole or void in the fuel (Fig. 4).

Although migration of the original porosity up the temperature gradient occurs in a time short compared with the fuel residence time in the core, the microstructure is not static after the initial restructuring. The mixed oxide is plastic at temperatures above ~1400 °C, so that the hot central portion of the fuel deforms rather easily in response to the compressive stresses built up by fuel-cladding mechanical contact. The outer, cool part of the fuel is still brittle, however, and develops extensive network of radial cracks. Because of power cycling, the crack pattern is frequently altered, and cracks in the hotter zones slowly heal. During closure, the cracks may spawn pores which then move up the temperature gradient by the same mechanism as did the as-fabricated porosity. Although the outer annulus of fuel is at too low a temperature for thermal creep, it is, like the cladding, subject to irradiation creep. By effectively softening the outer rim of fuel, irradiation creep significantly alters the stress distribution in the fuel, and thereby affects the intensity of mechanical interaction between fuel and cladding.

Independently of the temperature or the temperature gradient, the fission process continually modifies the chemical makeup of the fuel. Nuclear transmutations ultimately convert 10% of the uranium and plutonium atoms into fission-product elements that span the periodic table. Some fission products are gaseous and either escape from the fuel to cause pressure buildup in the cladding or precipitate into bubbles which swell the fuel and bring it into mechanical contact with the cladding.

The gaseous fission products generated in the inner columnar grain zone are swept up by the temperature gradient in the same manner as the pores. This region of the fuel is thereby continually cleansed of fission gases, which ultimately appear in the plenum of the fuel pin and in the fuel-cladding gap. The released fission gases are responsible for the pressure buildup

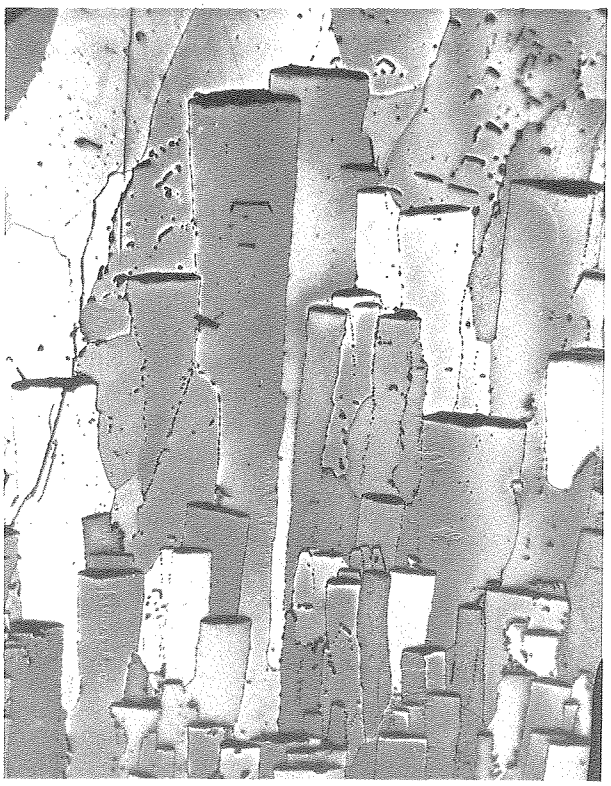

Fig. 3. Pores migrating up a temperature gradient in mixed-oxide fuel are revealed in this photomicrograph of a cut and polished cross section of fuel which had been irradiated in a reactor for a short time. Whatever the original shape of the void space in the as-fabricated fuel, they are transformed by motion into lens-shaped objects which are termed lenticular pores. Pore movement occurs by vaporization of the heavy-metal oxides from the hot faces (upper surfaces) and condensation on the cold faces (lower surfaces). The vapor condenses as a dense single crystal and so forms distinct grain boundaries with the zones outside of the cylindrical region swept out by the moving pore. The variation in shading is due to the chemical process used to prepare the surface, which etches grains of different crystallographic orientation to different extents. Courtesy G. Lundeen, General Electric Co.

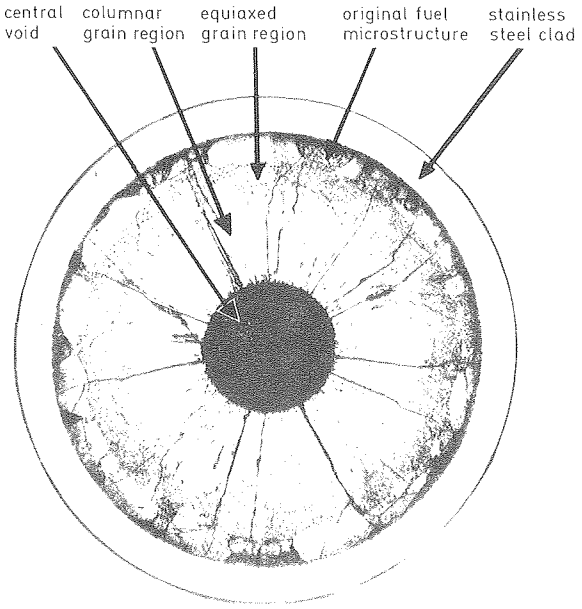

Fig. 4. Extensive restructuring occurs as a result of the steep temperature gradient in a fast breeder fuel element. The initial ceramic fuel is 90% of its theoretical (maximum) density, the remaining 10% being voids or pores about 50 μm in diameter distributed uniformly throughout the solid. Within a few hours after startup of the reactor, the pores migrate up the temperature gradient to form a central hole which is surrounded by an annular zone of high-density solid. The fuel in this zone is laced with radial streaks, which are the trails of the pores which have migrated through the zone. Beyond this columnar grain zone is an annulus consisting of large equiaxed grains produced by growth of the original grain structure of the fuel. The outer rim of the fuel is at too low a temperature to permit any observable alteration of the microstructure

which is a major source of stress in the cladding late in fuel life. The released gases also degrade the thermal conductance of the fuel-cladding gap by replacing the original helium with lower-thermal-conductivity xenon and krypton. As a result, the center fuel temperature slowly increases with burnup. Because of the nearly complete gas release from the columnar grain zone of the fuel, swelling in this region is minimal.

In the zone of equiaxed grain growth, the temperature is not high enough to provide sufficient mobility to fission-gas bubbles to permit them to move up the temperature gradient and the gas in this region coalesces into large bubbles on grain boundaries (Fig. 5).

In the peripheral rim of cool fuel, the fission-gas atoms are not mobile enough even to precipitate into bubbles. In this region, the gas remains atomically dispersed in the fuel matrix, and both gas release and swelling are low.

The gaseous fission products xenon and krypton, although contributing the lion's share to fuel swelling,

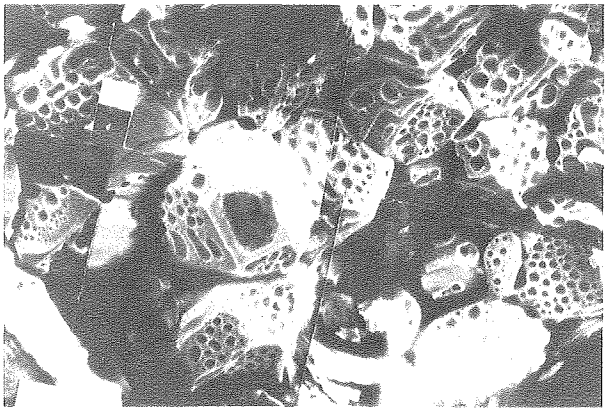

Fig. 5. A montage of photomicrographs of the fracture surfaces of irradiated UO_2 shows the development of fission-gas bubbles on grain faces. These bubbles eventually link up to form tunnels along the grain edges, and after about 8 volume percent swelling, the interlinkage is so extensive that the gas is vented from the fuel. The grain-edge tunnels and grain-face cavities then slowly close by sintering and the process repeats itself. The swelling due to the fission-gas bubbles trapped in the ceramic oxide causes the fuel to press against the cladding tube, which can rupture if the mechanical loading is too severe. Escape of fission gas by the venting process reduces fuel swelling but pressurizes the inside of the fuel element, which also mechanically stresses the cladding. Courtesy J.A. Turnbull, Central Electricity Generating Board, U.K.

constitute only 12–15% of the total quantity of fission products. The remainder of the fission products are solids, and either dissolve in the fuel matrix (Zr, rare earths) or precipitate (Fig. 6) into separate solid phases (Tc, Pd, Ba, Sr).

It is often stated that fission is an oxidizing process. This unlikely attribution of a chemical consequence to a nuclear phenomenon has a sound basis in fact. When a uranium or plutonium atom in an oxide nuclear fuel fissions, two changes occur. First, two fission-product atoms appear in place of the heavy-metal atom, and second, the two oxygen atoms that were associated with the original heavy-metal atom (in the solid UO_2 or PuO_2) are liberated. The fission products can be any one of the 30-odd elements in the middle of the periodic table. If one of the two products of fission has a strong affinity for oxygen (zirconium, for example), this element can re-immobilize the liberated oxygen. However, if neither of the fission products form stable oxides (ruthenium or iodine, as examples), the liberated oxygen is free to leave the fuel and to chemically attack the cladding. The collection of fission products arising from the plutonium component of the mixed-oxide fast breeder fuel $(U,Pu)O_2$, exhibits just this tendency. At high burnup, the fuel of a fast breeder reactor releases sufficient oxygen to thermodynamically favor corrosion of the cladding. In order to postpone the inevitable release of oxygen from the irradiated solid, the

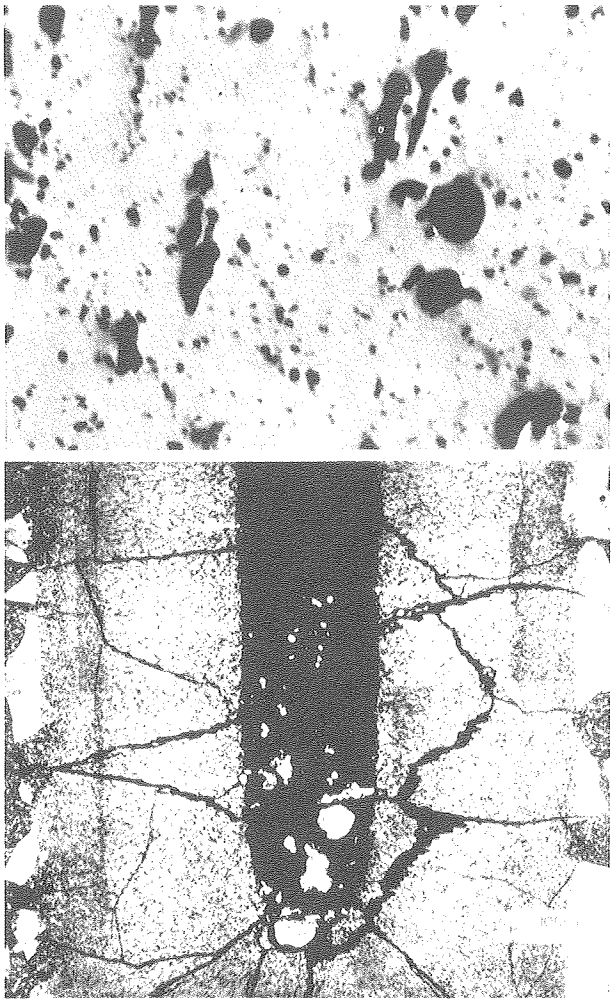

Fig. 6. Precipitates of noble metal fission products (an alloy of rhodium, ruthenium, technetium and palladium) in irradiated mixed-oxide fuel appear as small white inclusions in the top photomicrograph. The large black objects are pores. By some as yet unknown mechanism, the inclusions migrate to the hot center of the fuel where they agglomerate into large globules (some nearly a millimeter in diameter) and fall or drip to the bottom of the central void (bottom photograph of a longitudinal section through a fuel element)

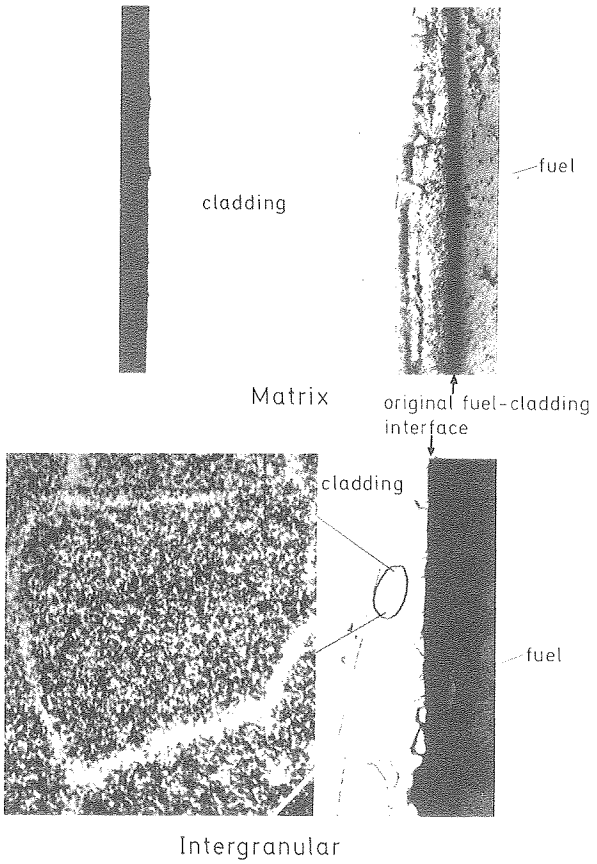

Fig. 7. Two types of corrosive attack of stainless steel cladding by mixed-oxide fuel in a fast breeder reactor fuel element. The top photograph shows a uniform attack of the matrix of the cladding, which affects about 10% of its thickness. In the bottom photograph, corrosion has proceeded selectively along grain boundaries of the metal. The enlarged view of a single grain shown at the bottom left was obtained with a scanning electron microscope which has the capability of detecting individual elements. This method reveals the concentration of fission product cesium in the grain boundaries of the metal. The grain boundary type of chemical attack can cause the weakened grains of the metal to fall out or can initiate further cracking of the intact outer zone of the cladding tube

initial fuel is purposely fabricated with a deficiency of oxygen (oxygen-to-metal ratio of ~ 1.95 instead of 2.00).

Superimposed on the chemical evolution of the fuel during irradiation, the temperature gradient causes most of the chemical species in the fuel to migrate radially; plutonium moves towards the center of the rod, thereby degrading heat transport by placing the fissile heat source further from the heat sink (the liquid sodium coolant) than in the as-fabricated fuel; oxygen is transported to the cladding, thereby enhancing corrosion; some fission products move up the thermal gradient while others, such as cesium, tellurium and iodine, move down the gradient to the cladding where they act to accelerate corrosion. This attack can produce uniform but shallow corrosion or deep and therefore more worrysome intergranular corrosion (Fig. 7).

Cladding

The cladding must exhibit acceptably low neutron absorption if the fuel element is to fulfill its primary role of sustaining the nuclear chain reaction. This requirement discourages utilization of many struc-

tural metals (such as high-nickel alloys or molybdenum) which otherwise possess desirable material properties. Use of thick-walled cladding would solve many of the structural integrity concerns, but is unacceptable because of the poor neutronic performance of a core containing the large fraction of neutron-absorbing metal which such a remedy implies. The fuel element must have a coolable geometry, which requires a small diameter to prevent central melting of the oxide. Yet large aspect ratio tubing is susceptible to bending due to interaction with the stiff duct which houses some 200 fuel pins. The cladding must also possess adequate creep strength at ~650 °C and resist corrosion by liquid sodium on the outside and by the fuel-fission product mixture on the inside. The high temperature strength requirement precludes use of ferritic alloys (ordinary steels) even though they are more resistant to the troublesome radiation effects such as void swelling and helium embrittlement than the austenitic alloys (stainless steel). The structural metal of the fuel rod must be cheap and easily fabricable, since the ultimate product of the reactor, electrical power, must be economically competitive with alternative generation methods. The material chosen for the demonstration LMFBR in the USA is type 316 stainless steel. This alloy contains 65% iron, 13% nickel (to stabilize the austenitic or face-centered cubic crystal lattice), 17% chromium (for corrosion resistance) and a number of minor elements, including carbon for strength. The stainless steel is 20% cold-worked, which means that its cross sectional area has been reduced by this amount in a rolling operation. Cold-working improves the creep strength and resistance to void swelling of the steel.

The high-energy neutrons which permeate the core of a fast reactor interact with the atoms of the structural metals in three principal ways, which affect reactor performance differently. Absorption reactions such as

$$n + {}^{59}Co \rightarrow {}^{60}Co$$

lead to radioactive products (^{60}Co in this case) which decay by emission of an energetic gamma ray. Small quantities of radioactive atoms are leached from the metal by the flowing sodium and are deposited in other parts of the coolant circuit, thereby making plant maintenance difficult. Neutron activation of the core structures will also pose as yet undetermined problems in dismantling the reactor after its 30- or 40-year lifetime.

Very energetic neutrons can produce reactions of the type

$$n + {}^{58}Ni \rightarrow {}^{55}Fe + {}^{4}He$$

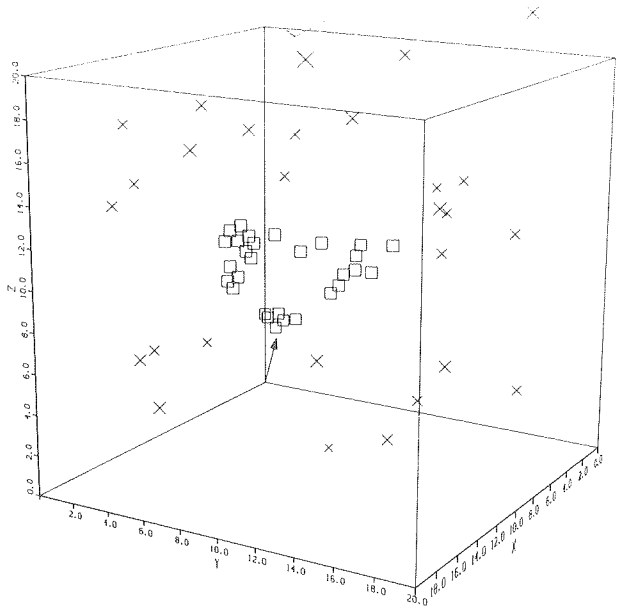

Fig. 8. Computer simulation of the microscopic damage resulting from a single collision of a fast neutron with an atom in copper shows the development of the collision cascade. The fast neutron collided with a lattice atom located on the lower rear corner of the cube and sent it off in the direction of the arrow with an energy of 10 keV. Subsequent collisions of this primary knockon atom (PKA) with other lattice atoms created the defects shown within the cube. The squares represent vacancies and the crosses are interstitial atoms (the atoms in normal lattice positions are not shown). Extensive clustering of like defects is seen. The scale on the edges of the cube represent distances in units of the lattice parameter of the copper crystal (about 3.61 Å) so that the damaged region is contained in a cubical volume approximately 75 Å on a side. Courtesy H.L. Heinisch, Hanford Engineering Development Laboratory

where the reaction product of concern is helium. This inert gas is insoluble in the metal lattice, but rather than escape at free surfaces, it agglomerates into bubbles in the metal. Many of the helium bubbles form at or migrate to grain boundaries and by growing under tensile stresses, contribute greatly to the high temperature embrittlement of the metal.

However, the principal fast neutron reaction with the metal atoms is scattering. The neutron leaves such a nuclear event having transferred a small fraction of its initial energy to the struck atom, which is known as a primary knockon atom or PKA. The average PKA energy in a fast reactor is 15–20 keV, which is more than sufficient to dislodge it from its lattice site. The PKA then strikes neighboring atoms and they go on to collide with others. The cascade of damage which results from this chain of events is over in a picosecond and produces a large number of pairs of vacant lattice sites (vacancies) and atoms lodged in interstitial positions in the crystal structure (interstitials) (Fig. 8). Vacancies and interstitials are antiparti-

cles like those of nuclear physics. The chance encounter of these two point defects in the solid results in the disappearance of both and the release of energy. The quantity of energy released by recombination is usually negligible. However, when a large number of point defects are built up by low-temperature irradiation and then become mobile by virtue of an inadvertent temperature rise, the heat released by their mutual annihilation can be sizeable. Just this process was responsible for the overheating of the British graphite-moderated reactor at Windscale in 1957, which resulted in the release of significant radioactivity to the environment. This phenomenon, which is called Wigner release after the physicist E.P. Wigner who earlier had predicted its potential effects, cannot occur in fast breeder reactor structures because the quantity of thermal energy contained in the hot metal is far larger than that released by point-defect recombination.

Within a microsecond after the production of a collision cascade by neutron collision with a lattice atom in a fast breeder structure, recombination of nearby vacancies and interstitials removes many of the original point-defect pairs, others join with their own kind to form vacancy or interstitial clusters and a few are able to migrate away from the scene of the original neutron collision. The clusters of interstitials are the nuclei of interstitial loops, and the vacancy clusters may eventually grow into voids (Fig. 9). The free single interstitials and vacancies migrate about the metal and disappear by mutual annihilation or are removed by various types of sinks which include the native structural defects in the metal (dislocation lines and grain boundaries) and the radiation-produced voids and loops. Only a small fraction of the vacancies produced by neutron interactions with the lattice (less than 0.1%) survive in the form of voids. Correspondingly, an equal number of interstitials appear in the microstructure as loops, which are circular disks of atoms squeezed between normal atomic planes of the crystal lattice (Fig. 10).

Void formation in metals bombarded by fast neutrons was a totally unexpected occurrence when C. Cawthorne and E. Fulton first observed them in 1967 on transmission micrographs of specimens removed from the Dounreay fast reactor. Voids are nearly spherical empty holes embedded in the irradiated metal, which acquires the structure of swiss cheese. They had never been found in metals of light-water reactors because the fast neutron fluence (or dose) is 100 times smaller than in LMFBRs. The principal gross effect of void formation is swelling of the metal. The volume increase of steel irradiated in an LMFBR can be as large as 10%, which means that the linear

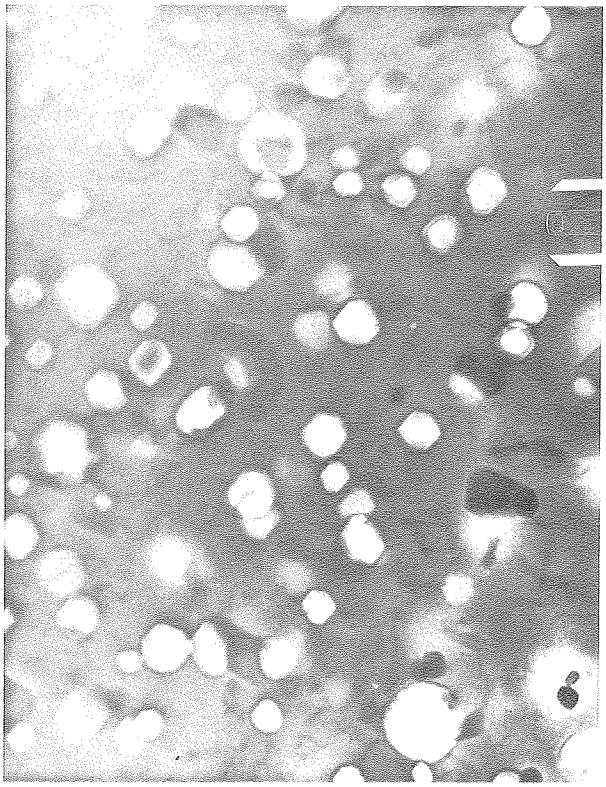

Fig. 9. Voids in fast-neutron-irradiated stainless steel are revealed in this transmission electron micrograph. They are typically several hundred angstroms in diameter and each represents the agglomeration of over a million vacancies. These cavities are roughly spherical in shape but often exhibit faceted surfaces which have been identified as the stable close-packed planes of the crystal lattice of the metal. This feature of voids in metals is analogous to the growth habits of naturally occurring crystalline minerals, which mimic on a large scale the crystal symmetries of the atomic structure of the solid

dimensions of the specimen can increase by about 3%. Although a dimensional change of this order is acceptable in the cladding of the fuel element, it presents severe structural design problems when it occurs in other components of an LMFBR core, particularly in the ducts which house the fuel elements. Unless sufficient clearance between adjacent ducts in the core is allowed, removal or insertion of the fuel assemblies during refueling can be impeded. A more serious structural effect of void swelling arises from the variation of temperature and neutron flux over the core of a fast breeder reactor. Such variations can cause opposing faces of the ducts to swell by different amounts, which results in bowing of the fuel assemblies. The core of a fast breeder reactor would open up like a flower in the morning sun unless void swelling is suppressed or the core is clamped to prevent the outward movement. For a machine which is designed and constructed with high precision, the large and nonuniform swelling due to voids in

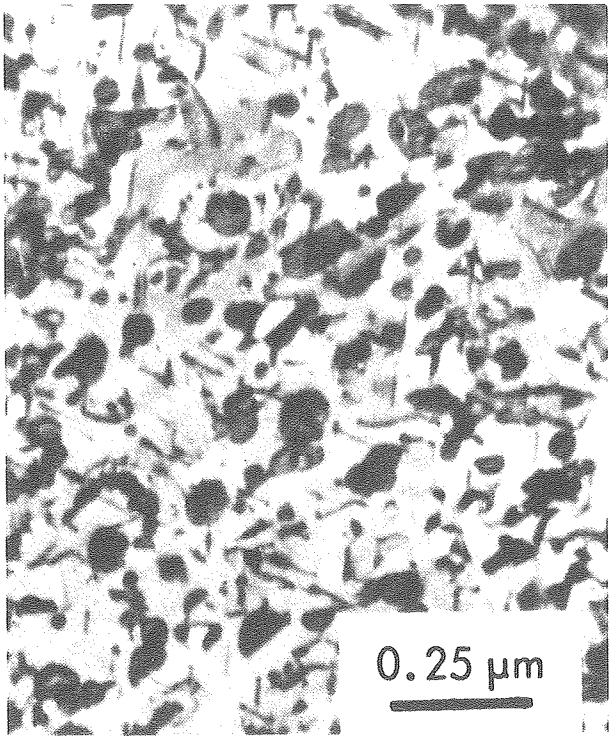

Fig. 10. Dislocation loops in fast-neutron-irradiated stainless steel appear as dark ovals in this transmission electron micrograph. The loops are circular platelets of interstitial atoms which have collected in between the close-packed planes of the crystal structure. These disks are about 500 – 1000 Å in diameter and contain roughly 100 000 interstitial atoms each. The periphery of each disk has the same atomic structure as an ordinary edge dislocation so the disks are called dislocation loops. The worm-like lines in the picture are normal dislocations in the metal

the metal represents a veritable bull in a china shop. For successful operation of fast breeder reactors, this effect must either be suppressed (by operating at lower temperatures or for shorter times or by using a void-swelling-resistant metal) or be properly accounted for in design, which requires very accurate knowledge of the influence of temperature, neutron fluence, and metallurgical state of the metal on the dynamics of void growth.

Void formation represents a delicate balance between competing phenomena. First, the process is active only in a restricted range of temperature. At low temperatures (in general, less than $^1/_3$ of the melting point in °K for fast-neutron irradiation), the vacancies are not sufficiently mobile in the metal to be capable of coalescing into voids. At high temperatures (greater than about $^1/_2$ of the melting point of the metal), voids are unstable and dissipate like a morning fog in the noonday heat. Second, the rate at which voids grow in a neutron-irradiated metal is intimately related to the properties and number of a natural defect in the crystal structure of metals, the dislocation line (see Fig. 11). No solid, even a laboratory-grown single crystal, is free of dislocation lines. They possess the remarkable property of acting as unsaturable sinks for point defects. That is, if a vacancy or interstitial, in the course of its migration about the crystal structure, encounters a dislocation line, the point defect is absorbed and vanishes. The only effect on the dislocation line is a slight displacement. The process is a two-dimensional analog of the disappearance of a droplet of water falling into a puddle. Void formation is possible only because dislocation lines have a slight preference for absorbing interstitials rather than vacancies. Consequently, the dislocation density must be just right for voids to grow. Too few dislocations, and their bias cannot be exploited to form voids; too many dislocations suppress the concentration of both types of point defects, and void nucleation and growth are prevented. The severe distortion of a metal in a cold working operation increases the density of dislocations by a factor of 10 to 100. Cold-working has been a common metallurgical process for many years because the increased dislocation density it produces serves to harden the metal. For LMFBR use, however, the hardening effect is secondary; the primary objective of cold-working is to increase the number of dislocations so that their intrinsic appetite for point defects can be exploited to suppress the growth of voids during irradiation.

The slow, permanent distortion which occurs in hot metals under stress is a familiar phenomenon to mechanical designers. This type of deformation, which eventually leads to rupture, is called thermal creep because it is significant only at temperatures above about one-half on the metal's melting point. On a microscopic level, the need for high temperature can be explained in terms of dislocation motion. Stressing a metal acts to move dislocations, and when many have traversed the specimen, visible, permanent deformation occurs. Dislocations cannot move freely through a metal, however. They can be blocked by obstacles such as impurity aggregates, grain boundaries, or other dislocation lines. At low temperatures, application of a sufficiently large stress enables the dislocation to cut through the obstacles in its path and go on to deform the metal. The stress at which dislocation liberation occurs is known as the yield stress of the solid, the name evoking the sudden loss of rigidity which characterizes the demarcation between elastic and plastic behavior. At elevated temperatures, another mechanism becomes available to the dislocation to circumvent the obstacles in their way. High temperatures generate a natural population of vacancies in solids, in much the same way that hot water supports a well-defined pressure of steam

with which the liquid is in equilibrium. In a hot, stressed metal, these thermally generated vacancies are absorbed by the dislocation lines which are pressed against obstacles but are unable to cut through them because the applied stress is insufficient. The vacancy-absorption process causes the dislocation lines to move at right angles to the direction that the stress pushes them, and as a result, the dislocations literally climb over the obstacles which are impeding their motion. Once past the obstacle, the stress pushes the dislocations to new obstacles, where the same escape process is repeated. This sequence of jerky climb-glide motions enables the dislocations to proceed through the metal without ever having to cut through the obstacles to their natural motion. The net effect is macroscopic distortion of the solid, albeit very slow by comparison to the essentially instantaneous deformation which occurs when the stress is above the yield point. If the yielding process can be likened to an automobile moving at high speed on the open road, the vehicular analog of creep is driving along a busy city street with the traffic lights staggered the wrong way.

Radiation enhances creep in metals by obviating the need for high temperatures. Instead of thermally generated vacancies providing the means of dislocation climb around obstacles, irradiation-produced vacancies perform the same function (Fig. 12). A stressed metallic structure which would normally be dimensionally stable at low temperature exhibits significant creep distortion when placed in an irradiation field such as the energetic neutron flux of a fast breeder reactor. All structural components of a nuclear reactor core are susceptible to irradiation-induced creep. This new form of creep is undesirable because it produces permanent distortion of components and requires data and theory on a new radiation-sensitive mechanical property in order to circumvent by proper design. On the other hand, the normally rigid structures in the core of a fast reactor become compliant because of irradiation creep, and thus are capable of relieving stresses which might otherwise have built up to the fracture point.

In addition to the irradiation-produced physical phenomena of void swelling and creep, several chemical

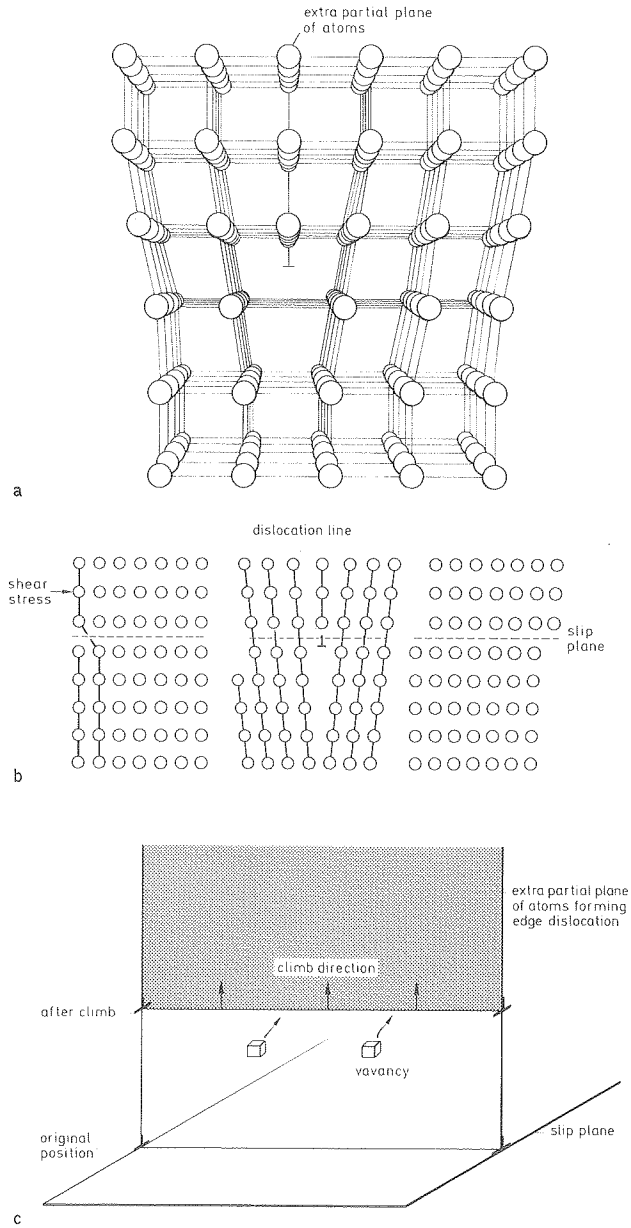

Fig. 11. (a) The line defect in solids known as an edge dislocation is the termination of an extra partial plane of atoms inserted in the crystal lattice. (b) The diagram shows how a shear stress applied to a face of the crystal produces an edge dislocation and how the dislocation moves through the solid along a particular plane (the slip or glide plane) to cause permanent deformation. The shear stress causes the column of atoms on the extreme left above the slip plane to shift its alignment to the second column of atoms below the slip plane. As this realignment process proceeds along the slip plane, the dislocation moves to the right in much the same way that a wrinkle in a sheet is smoothed out. Eventually, the dislocation reaches the opposite face of the crystal from which it was formed and the net effect is a realignment of all atomic planes above and below the slip plane by one interatomic distance. When a very large number of dislocations have passed through the crystal in this manner, visible shear deformation occurs. (c) The diagram shows a different form of motion of an edge dislocation caused by absorption of vacancies on the bottom of the extra partial plane of atoms. Originally, the dislocation, represented by the line joining the two upside-down tees, rests on the slip plane in the diagram. Vacancy absorption chews away at the bottom of the extra partial plane of atoms causing it to move upward, or to climb. If the original slip plane is regarded as a stage, climb is equivalent to raising the curtain. If the dislocation line absorbs more interstitials than vacancies, it climbs in the opposite direction from the one shown

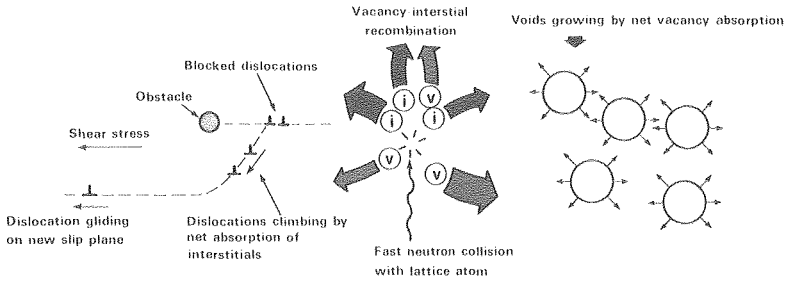

Fig. 12. The preference of edge dislocations for absorbing interstitial atoms rather than vacancies is responsible for the twin phenomena of void growth and irradiation creep. The cascade initiated by collision of a fast neutron with a lattice atom creates equal numbers of vacancies and interstitials. A fraction of those recombine to reform the perfect lattice structure (top). Of the remainder, a slightly larger number of interstitials than vacancies flow to edge dislocations (left) and to make things come out even, more vacancies than interstitials must be absorbed at the only other sink, the voids (right). Consequently, the voids enlarge and the dislocations climb over obstacles in the path. Or, the metal simultaneously swells and creeps (if it is stressed). Were it not for the inherent bias of dislocations for interstitials, both types of point defects would flow to both types of extended defects in equal numbers, and neither void growth nor irradiation creep would occur

phenomena affect cladding performance by altering the properties of the metal. The propensity of the mixed-oxide fuel of the fast breeder reactor to corrode the inside wall of the cladding has already been mentioned. In addition, depending upon the carbon content of the sodium coolant, the cladding may be decarburized or carburized. If the liquid sodium has a low carbon concentration, it dissolves carbon from the steel, which makes the metal weaker. If the carbon content of the liquid metal is high, carbon is absorbed by the steel and becomes embrittled. Inadequate control of the oxygen content of the sodium can result in catastrophic corrosion of the outer surface of the stainless steel cladding tube. If an oxygen level in the sodium of less than 1 ppm is maintained, however, the loss of metal is less than 0.02 mm annually. Operation of the cladding at temperatures greater than 600 °C can desensitize the steel, or cause precipitation of the carbide phase $Cr_{23}C_6$. Desensitization lowers the corrosion resistance of stainless steel and also reduces its strength by immobilizing the two essential components (chromium and carbon) into the ineffective carbide form.

Conclusion

Had the vast array of chemical, physical and metallurgical phenomena which occur in irradiated fast breeder fuel and cladding been anticipated twenty years ago, the engineering task of structuring a reliable power-producing machine might have appeared insurmountable. However, the large-scale demonstration units in France and the United Kingdom have demonstrated that the materials problems are manageable; in all likelihood, liquid-metal fast breeder reactors can achieve their potential of a 60-fold improvement in uranium-resource utilization and power-plant thermal efficiencies greater than 40% compared to 33% for plants with light-water reactors as the steam supply.

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