The effects of fast-neutron irradiation on the mechanical properties of austenitic stainless steel*

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SYNOPSIS

The paper reviews the effects of fast-neutron irradiation on the tensile properties of austenitic stainless steels at irradiation temperatures of less than 400°C, using, as an example, work carried out at Pelindaba on an AISI 316 type steel produced in South Africa. Damage produced in these steels at higher irradiation temperatures and fluences is also briefly discussed.

The paper concludes with a discussion of some methods of overcoming or decreasing the effects of irradiation damage.

SAMEVATTING

'n Opsomming word gegee oor die invloed van snelneutronbestraling op die trekeienskappe van austenitiese roesvrystaal by bestralingstemperature onder 400°C, waar werk wat te Pelindaba op 'n Suid-Afrikaansevervaardigde tipe AISI 316 staal onderneem is, as voorbeeld te gebruik. Die skade wat in hierdie staalsoorte teen hoër bestralings temperature en tydvloede geproduseer word, word ook kortliks bespreek.

Ter afsluiting word sommige metodes vir die oorkoming of vermindering van die invloed van stralingskade bespreek.

Introduction

With the introduction of nuclear-power reactors in South Africa, the Atomic Energy Board, in compliance with the Government's local-content policy, decided to study the effects of neutron radiation on the properties of locally produced stainless steels, with a view to the possible incorporation of these steels in the component parts of future generations of power reactors and reactor cores.

The problems associated with the use of austenitic stainless steels in reactor environments are well known. Much research effort has been expended on the reasons behind the phenomena observed, and on finding answers to overcome these effects.

The purpose of this paper is to provide an understanding of the problems involved by showing the effects produced in these steels by irradiation at lower temperatures, using, as examples, some of the work carried out at Pelindaba. This work has utilized irradiation temperatures below 400°C, this being the temperature range in which South African power reactors will operate. It is then proposed to take a brief look at the effects of increasing irradiation temperature and fluence, which is of interest in the case of hot spots within the cores of low-temperature reactors, and in the study of materials for use in high-temperature/high-flux reactors such as fast reactor systems.

Irradiation Effects on Austenitic Stainless Steels

Irradiation hardening and embrittlement in austenitic stainless steels exposed to neutron radiation over extended periods is well known and documented[1–6]. The irradiation-induced structural changes affecting tensile properties can be divided into three forms that are a function of the irradiation temperature and the neutron fluence.

1. 'Classical' irradiation hardening, occurring at low irradiation temperatures (less than 500°C) and moderate fluence levels.
2. Formation of voids, occurring at higher temperatures but within a fairly narrow range (400 to 650°C) and after fast-neutron irradiation to higher doses.
3. Formation of helium bubbles found at high temperatures (greater than 600°C) and in both thermal- and fast-flux irradiations.

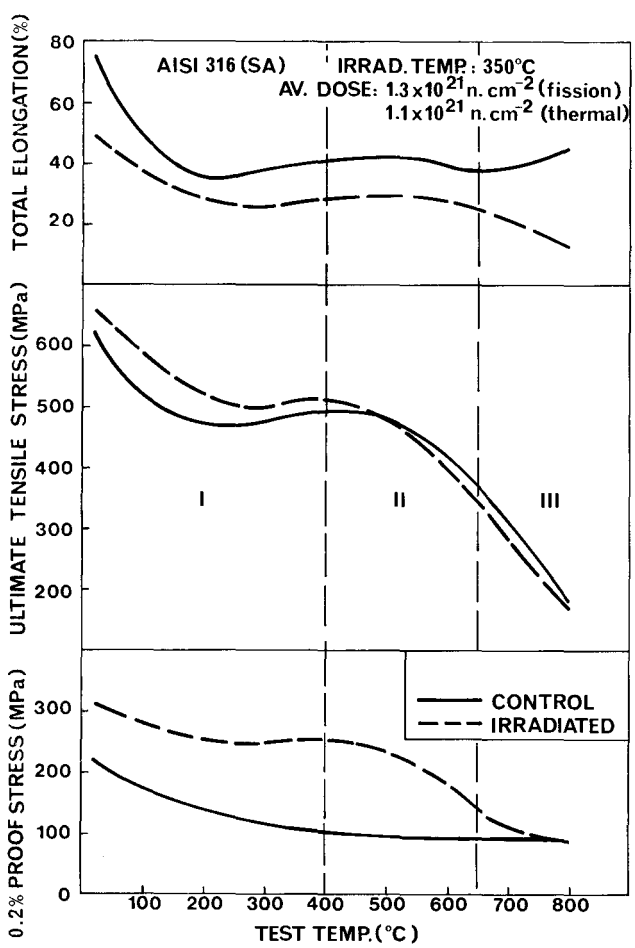


Fig. 1—Tensile properties versus test temperature for AISI 316 stainless steel produced in South Africa and irradiated in Safari 1 to 1,3 × 10^{21}n.cm^{-2} (fission) and 1,1 × 10^{21}n.cm^{-2} (thermal) at 350°C

*A paper given at the S.A. Institute of Physics symposium on 'The Deterioration of Materials in Service', which was held in Cape Town from 5th to 7th December, 1977.

†Atomic Energy Board, Pretoria.

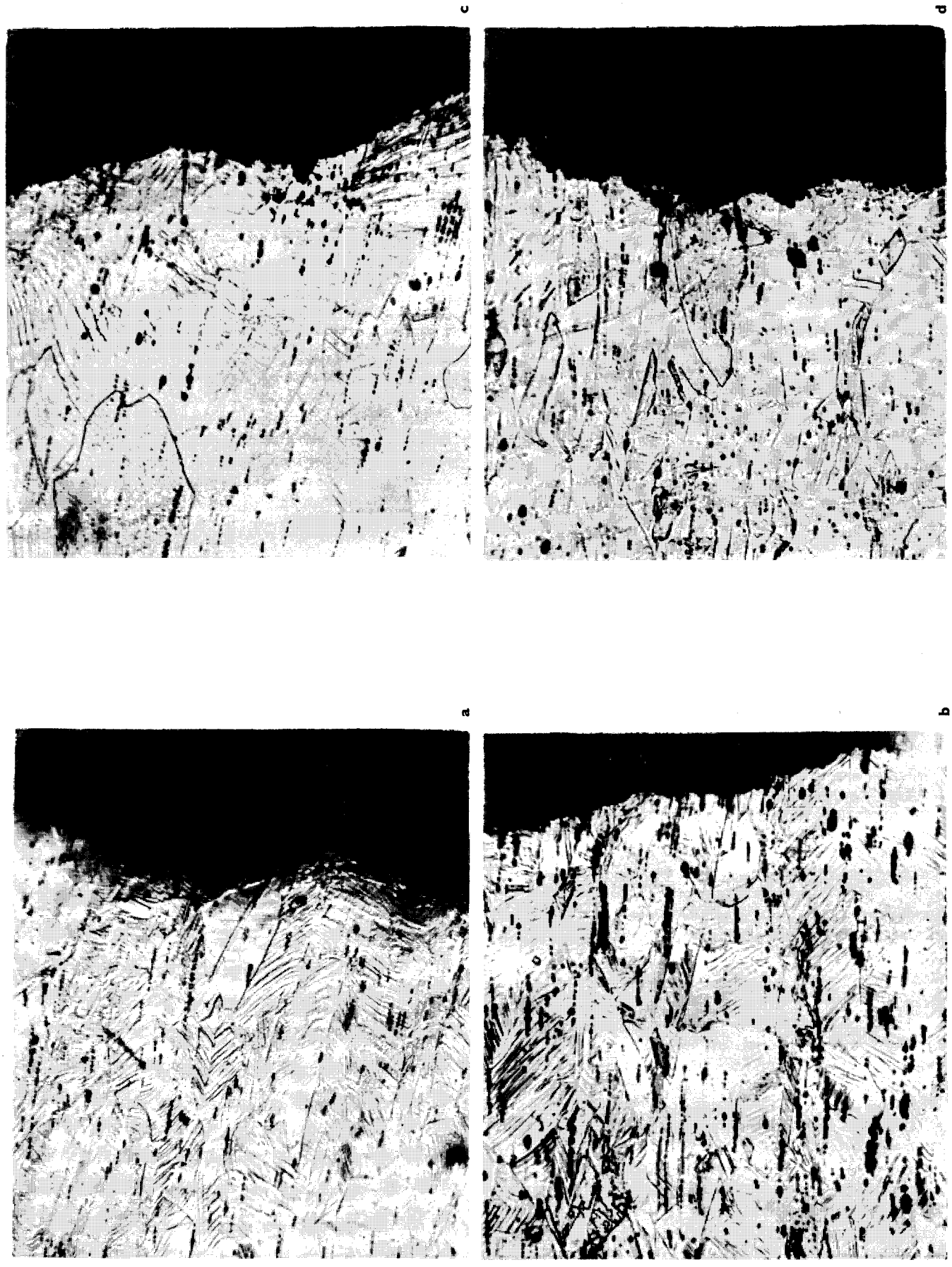


Fig. 2—Fractures in irradiated AISI 316 stainless steel (X250) tested at
a room temperature b 150°C c 300°C d 400°C

Low-temperature Irradiations (Below 500 °C)

Fig. 1 shows a plot of mechanical properties versus testing temperature for a series of post-irradiated and non-irradiated control tensile tests carried out on an AISI 316 type stainless steel produced in South Africa. The testing range was room temperature to 800 °C. These specimens were irradiated at an average temperature of 350 °C to an average dose of $1{,}3 \times 10^{21}$ n . cm^{-2} (fission) and $1{,}1 \times 10^{21}$ n . cm^{-2} (thermal). This graph shows that the behaviour falls typically into three distinct zones.

The first of these zones, occurring between room temperature and approximately 400 °C, is characterized by a marked enhancement of the proof-stress values, a lesser increase in the ultimate tensile stress, and an associated decrease in ductility as measured by total elongation of the irradiated material compared with the control material. Another feature is that the ductility values go through a minimum at around 300 °C, the temperature at which South African power reactors will operate.

Optical metallography carried out on specimens tested within this zone show appreciable evidence of deformation and a transgranular fracture mode. The amount of deformation decreases as the test temperature is increased (Fig. 2). Transmission electron microscopy reveals that, at the lower test temperatures, there exist heavy concentrations of what appears to be 'black dot' precipitation (Fig. 3). This has been shown, in fact, not to be precipitates but to be lattice defects in the form of barely resolvable prismatic loops[7], or larger interstitial loops and smaller vacancy clusters[8]. Subsequent annealing at progressively higher temperatures causes these defects to grow in size.

These defects in the structure are a result of the elastic scattering of fast neutrons within the lattice, resulting in primary displacements of atoms from their lattice sites. Displacement 'spikes' in the form of an array of interstitial and vacancy clusters result from these primary displacements and subsequent 'knock-on' events[9]. The defects thus formed aggregate and coalesce to form, throughout the lattice, vacancy and interstitial loops of varying size and complexity. The interaction between these defects and dislocations permits the explanation of the observed features of irradiation hardening. These features include an increase in the yield and flow stress, a decrease in the strain-hardening coefficient, and the subsequent early onset of plastic instability, which results in reduced uniform elongation[10].

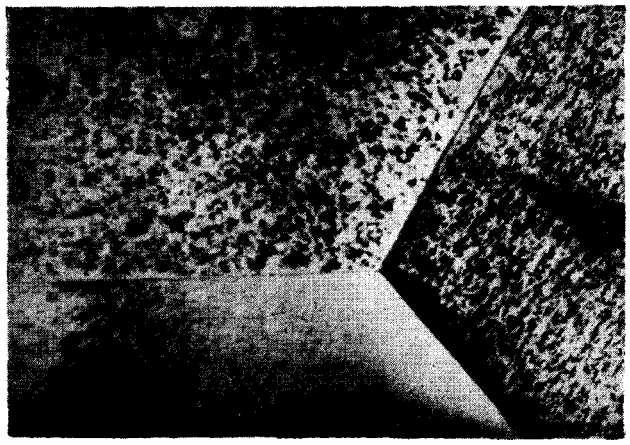


Fig. 3—'Black dot' type of low-temperature irradiation damage (Cawthorne and Fulton[39])

It has been found[11] that the amount of hardening is dependent on the irradiation temperature. The concentration of neutron damage that is effective in irradiation hardening depends largely on the relative rates of production and removal by annealing; that is, the higher the irradiation temperature, the less the increase in the mechanical properties, and hence the predomination of this effect at low irradiation temperatures. In fact, post-irradiation properties of steels irradiated at temperatures between 400 and 600 °C at low fluences show quite a change from those at lower irradiation temperatures. At test temperatures lower than 600 °C, no significant change in tensile properties due to irradiation occurs[4]. This lack of any neutron-induced alteration of behaviour under these conditions is attributed to the very rapid annealing, which keeps irradiation-induced damage at insignificant levels.

The number of defects produced by neutron irradiation increases with increasing neutron energy[12]; consequently, the magnitudes of the changes in properties at the low test temperatures increase with increasing fast-neutron dose.

The second of the zones lies between the testing temperatures of approximately 400 °C and 650 °C (Fig. 1). In this zone there is a rapid decrease in the proof stress of the irradiated material. This 'annealing-out' from the irradiation-hardening zone is due to the accelerated migration rates at these temperatures promoting an enlargement of and wider separation between the residual defect structure, and rendering it less effective in hardening the matrix by dislocation pinning and impedance.

Optical metallographic structures are nearly or completely strain free within this zone, failure still occurring in the trans-crystalline mode (Fig. 4).

The third zone, occurring at test temperatures above about 650 °C, shows strength properties comparatively unaffected by irradiation, but ductility severely reduced as compared with the control values. This severe loss in ductility is due to the curtailment of the non-uniform portion of the stress/strain curve (Fig. 5), which occurs during the necking down of the specimen.

Optical metallography carried out on specimens tested within this zone typically show little necking down, intergranular fracture, and cavitation between grains in areas removed from the fracture zone (Fig. 6). Electron-microscope investigation of these specimens reveals the presence of small bubbles or pores at the grain boundary (Fig. 7). It is known that inelastic interaction of high-energy neutrons with the constituent elements of stainless steel, notably (n,α) and (n,p) reactions, result in the formation of significant quantities of gaseous helium and hydrogen. At test temperatures above 650 °C, these gases are able to migrate rapidly to the grain boundaries, where they coalesce to form small spherical pores. Annealing experiments, supported by chemical analysis, suggest that these small pores would be filled with

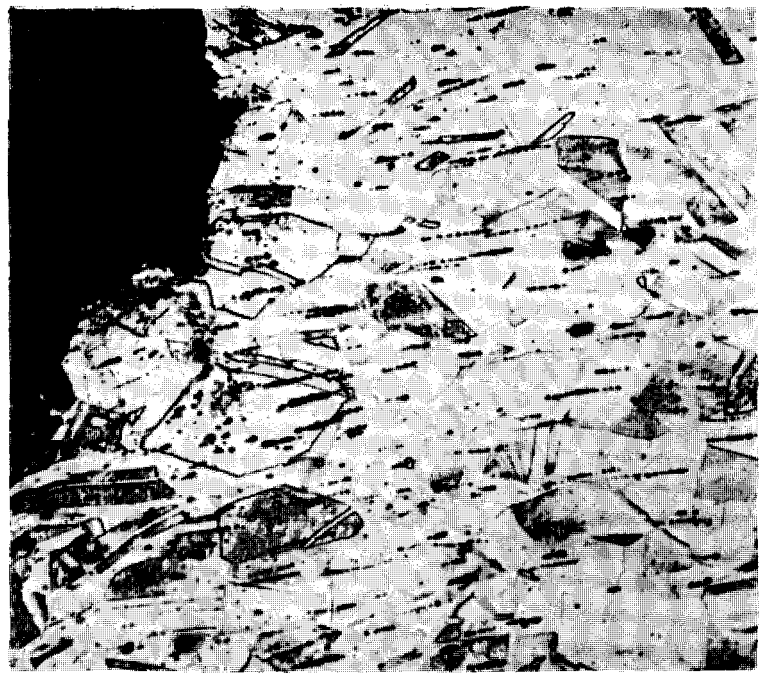


Fig. 4—Fracture of irradiated AISI 316 steel tested at 600°C (X250)

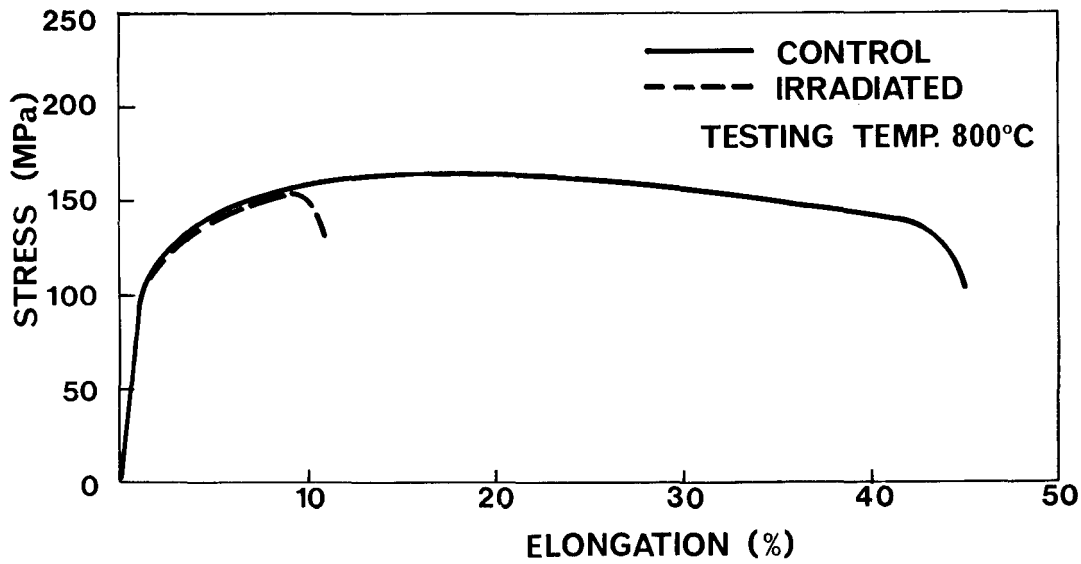
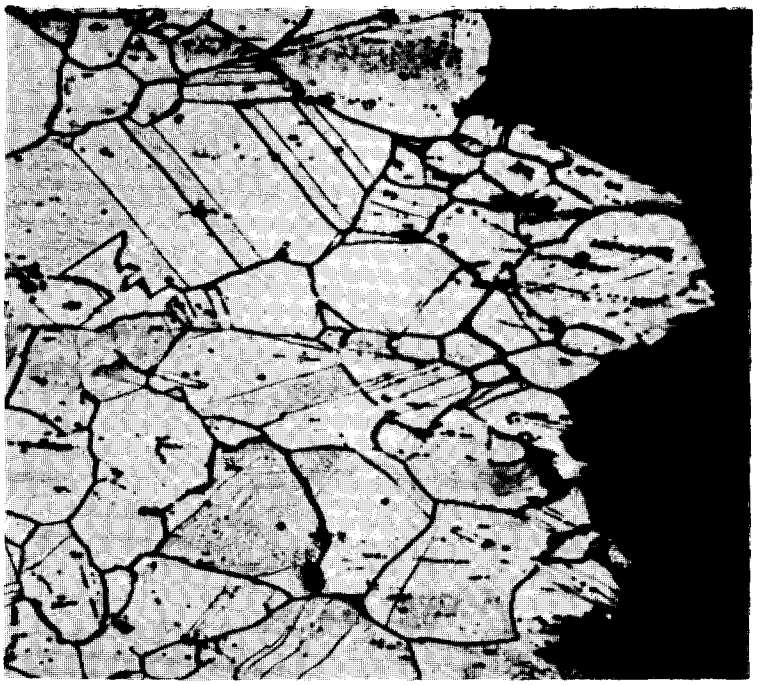


Fig. 5—Comparison between irradiated and control stress/strain curves for AISI 316 stainless steel tested at 800°C

helium rather than hydrogen, which, unlike helium, has a high solubility in the steel; they also suggest that the 10B(n,α)7Li reaction was the main contributor[3].

Barnes[1] used an embrittlement model developed by Hyam and Sumner[13] to explain the marked reduction in ductility and the change in the failure mechanism. It is argued that, since helium has a low solubility in the metal lattice, it would precipitate to form bubbles. Bubbles would be located at the grain boundaries because, at these temperatures, they are energetically preferred nucleation sites and because grain-boundary bubbles would be able to grow more rapidly than any that may be precipitated in the matrix as a result of more rapid vacancy production in the boundaries. Under the action of an applied stress, these bubbles will grow by acquiring vacancies. These grain-boundary bubbles are said to control the properties of the material because stresses can be concentrated at locations along the boundary. The loss of cohesion between the grains is ascribed to the interlinking of these bubbles under the influence of stress and temperature.

Rowcliffe *et al.*[14], on the other hand, although they demonstrated bubble growth, could not demonstrate that direct linking of these bubbles caused fracture. They ascribed failure as being due to a simple cavitation mechanism, since the bubbles they observed after fracture had occurred were below the critical size necessary for linking to have taken place. This theory was given some support by Waddington and Lofthouse[15, 16], who proposed that failures were of a wedge type, with the lower



a

b

Fig. 6—Fractures in irradiated AISI 316 stainless steels (X250) tested at
a 700°C
b 800°C

Fig. 7—Small helium bubbles at a grain boundary (Kramer *et al.*[40])

ductility resulting from an increase in the rate of crack propagation. This point of view was also given support by Summerling and Rhodes[17], although, in all these cases, the reason for the increase in the rate of crack propagation was obscure. Fracture at the grain boundaries in this zone, therefore, would appear to involve a combination of mechanisms[6]. Cracks may be initiated by the stress-induced growth of cavities, with the helium bubbles nucleating these cavities. Actual fracture does not occur due to cavity coalescence; instead, the boundary cracks, initiated by the growth of cavities, propagate and cause failure before extensive cavitation can occur.

Formation of Voids (400 to 650 °C)

Irradiation of austenitic stainless steels in a high fast-neutron flux in the temperature range 400 to 650 °C results in the formation of voids and associated Frank dislocation loops throughout the lattice (Fig. 8). These large defects result in a hardening of the material by the interaction of dislocations with the dislocation loops and by cavity hardening[18, 19], and cause swelling of the material[20, 21].

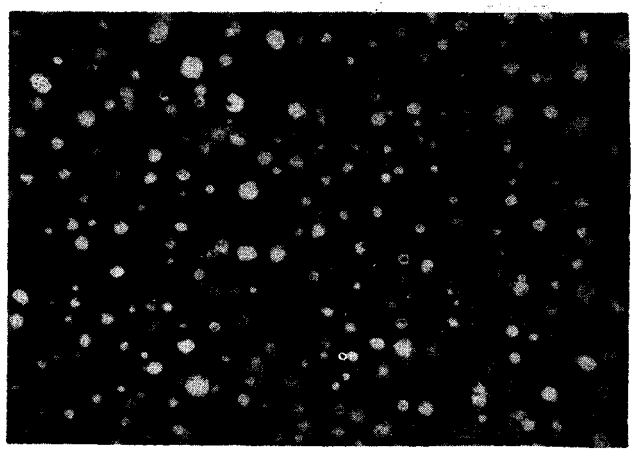


Fig. 8—Voids and dislocation loops in irradiated AISI 316 stainless steel (X 83,500) (Cawthorne and Fulton[39])

The stress/test-temperature graph for these steels is similar to that for lower-temperature irradiation, except that hardening in the lower test-temperature range is less pronounced[5, 22]. However, the hardening persists to higher temperatures owing to the voids and their associated loops remaining relatively stable at higher temperatures as compared with the low-temperature damage, which 'anneals-out' fairly readily[22]. The rapid fall-off in ductility at higher test temperatures (more than 650 °C) is similar to that found in materials irradiated at low temperatures and involves the same mechanisms.

The formation of voids in stainless steels is a phenomenon that was observed relatively recently[23]. The generally accepted mechanism is that voids evolve through a condensation of vacancies formed by displacement reactions on stable nuclei, which are preferably clusters[24] of helium atoms, the helium being derived from (n,α) reactions. Vacancies and interstitials are produced in equal numbers by displacement reactions, and would normally be eliminated by recombination or diffusion to neutral sinks such as grain boundaries. The appearance of voids, which implies a net excess of vacancy concentration, can be accounted for by dislocation loops and tangles proving to be effective interstitial sinks.

Voids appearing in the 300-series steels are essentially octahedral in shape, bounded by {111} planes. Void diameters vary between the smallest visible (about 30 Å) to 1200 Å, population density being of the order 10^{15} . cm^{-3}. They are randomly dispersed within grains, but not at grain boundaries, and are invariably associated with a large number of interstitial loops, predominantly faulted in the range 400 to 500 °C and becoming fault free at 550 to 600 °C, and dense dislocation tangles developing at temperatures above 550 °C. There is a consistent increase in void diameter as the irradiation temperature rises[25], and voids cease to form at about 750 °C. At approximately 630 °C, voids are seen attached to carbide particles of the $M_{23}C_6$ type, and it is believed that accelerated swelling can occur at high temperatures if thermal or irradiation processes lead to the appearance of coarse precipitates. Owing to the importance that a change of dimensions may have on fuel cladding or in core support members, the prime consideration in the use of material that falls in the void-formation range is, of course, the associated swelling of the material.

High-temperature Irradiations (Above 600 °C)

In irradiations at higher temperatures, embrittlement has generally been attributed to helium arising from 10B(n,α)7Li and fast (n,α) reactions, the principal elemental constituents of the steel forming bubbles at the grain boundaries, as was described for the high-temperature embrittlement of material irradiated at low temperatures. In this case, however, the precipitation of $M_{23}C_6$ carbides during irradiation must be taken into consideration. The rôle of changes in precipitate distribution[26, 27] and solid transmutation products[28] have been studied, and it would seem likely that these make some contribution to the irradiation embrittle-

ment; however, it seems likely that the primary mechanism is due to the helium at the grain boundaries.

Conclusions

One might well ask whether there is anything that can be done to at least minimize the debilitating effects of irradiation on the properties of austenitic stainless steels.

In the case of hardening as a result of low-temperature irradiation, something can be done. Some samples of a 'reactor-grade' AISI 316 type of steel were irradiated in the same rig, and therefore under the same conditions, as the South African steel discussed earlier. It can be seen from Fig. 9, in which the two steels are compared, that the effects of irradiation were not so severe on the reactor-grade steel; that is, irradiation hardening was reduced, and the ductility was improved at low test temperatures. The reason for this is that reactor-grade steel has a much tighter specification range than those of ordinary production steels, and is double-vacuum remelted and cast. One of the aims of the irradiation programme at Pelindaba is to make similar specifications for the nuclear-grade steels produced in South Africa.

Considerable effort is now being directed towards finding alloys with improved voidage-swelling characteristics. It has, for instance, been found that minor changes in the composition of stainless steels, such as the addition of small amounts of carbide-stabilizing elements like titanium[29, 30] or niobium[21], materially improve resistance to swelling. Brammen[21] also found some indications that a simple reduction in the carbide content of a stainless steel may be beneficial. Lauritzen[31] found that deliberate coarsening of carbide precipitates before irradiation resulted in increased swelling. The most dramatic reduction in the formation of voids has been found in the case of Nimonic alloy PE16, which has been attributed to the fine dispersion of γ' precipitates[32].

In the case of helium embrittlement, it has been suggested[33] that solutions to this problem can be related to alterations to the unirradiated alloy that affect the process of intergranular fracture and to modification of the alloy that reduces the amount of helium located at the grain boundaries of the irradiated alloy. One approach is the desegregation of boron, and the production of helium sinks within the matrix of the grains. Titanium additions are believed to form complex metal borides dispersed homogeneously within the matrix, and the precipitate matrix serves as a depository for helium[34]. Bagley found that niobium carbide precipitates acted as trapping sites for helium in cold-worked stainless steel, and prevented agglomeration and growth of grain-boundary bubbles in solution-treated steel[35]. Votinov[36] also found that niobium addtions caused a stainless steel to suffer little irradiation embrittlement up to a temperature of 750 °C. Finally, there are indications that ferritic steels are not embrittled at high temperatures[37, 38] in conditions under which austenitic steels showed severe embrittlement.

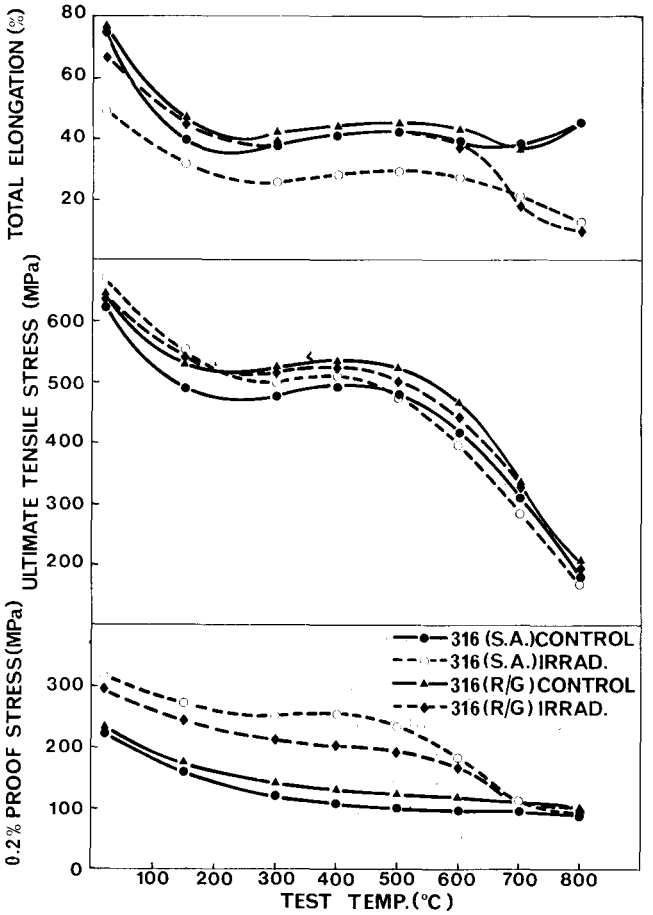


Fig. 9—Comparison of the tensile properties of a steel produced in South Africa with those of a nuclear-grade AISI 316 stainless steel

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Semiconductor technology

A Symposium on Semiconductor Technology is to be held in Pretoria on 30th November and 1st December, 1978. The Symposium, which is being held under the auspices of the South African Institute of Physics (SAIP), is organized by the SAIP's Subcommittee for Solid State Physics and Materials Science, assisted by the Solid State Electronics Division of the National Electrical Engineering Research Institute (NEERI) of the CSIR.

The major aim of the Symposium is to provide a forum for scientists and engineers from universities, research laboratories, and industry to meet and exchange ideas on semiconductor technology. Overseas speakers will also be invited. Both ordered and disordered material will be covered.

All correspondence should be addressed to The Organizers, Symposium 'Semiconductor Technology', c/o Solid State Electronics Division, NEERI, CSIR, P.O. Box 395, PRETORIA 0001.

Seminars on mining and metallurgy

During the 1978-79 academic year, McGill University will once again be offering professional development seminars in various aspects of mineral engineering, mineral management, and mineral economics. These seminars have been designed to meet the practical needs of the Canadian mineral industry and of mining organizations in developing countries. Lectures, discussion groups, case studies, and workshops are conducted by the staff of McGill University with the assistance and co-operation of recognized authorities from government agencies, mining companies, independent consultants, and other universities. The United Nations Centre for Natural Resources, Energy and Transport and the Canadian International Development Agency help in arranging the participation of personnel from developing countries.

The following seminars are planned:

- Support of underground workings — principles. Seminar Leader J. E. Udd. 16th-20th Oct., 1978.
- Support of underground workings — practices. Seminar Leader J. E. Udd. 23rd-27th Oct., 1978.
- Slope stability. Seminar Leader J. E. Udd. 30th Oct.-3rd Nov., 1978.
- Blasting systems. Seminar Leader R. R. MacLachlan. 6th-10th Nov., 1978.
- Health and safety aspects of the mine environment. Seminar Leader D. A. Trotter. 13th-17th Nov., 1978.
- Engineering aspects of the mine environment. Seminar Leader R. R. MacLachlan. 20th-24th Nov., 1978.
- Quality of working life in the mineral industry. Seminar Leader T. E. Hawkins. 27th Nov.-1st Dec., 1978.
- Environmental control. Seminar Leader R. R. MacLachlan. 4th-8th Dec., 1978.
- Economic analysis for mine operators. Seminar Leader B. W. MacKenzie. 11th-14th Dec., 1978.
- Union management–employee relations in the mineral industry. Seminar Leader T. E. Hawkins. 29th-31st Jan., 1979.
- Geostatistical or reserve estimation. Seminar Leaders M. L. Bilodeau and M. David. 5th-9th Feb., 1979.
- Mineral investment decision techniques. Seminar Leader B. W. MacKenzie. 12th-23rd Feb., 1979.
- Organizing for results in the mineral industry. Seminar Leader T. E. Hawkins. 26th Feb.—2nd Mar., 1979.
- The financing and implementation of mineral projects. Seminar Leader P. Glenshaw. 5th-9th Mar., 1979.
- The planning and execution of mineral development strategies. Seminar Leader P. M. T. White. 12th-16th Mar., 1979.
- Materials handling in mines. Seminar Leader D. A. Trotter. 19th-23rd Mar., 1979.
- Mineral processing systems. Seminar Leader J. A. Finch. 26th Mar.-6th Apr., 1979.

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