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Overview of Studies in the United Kingdom on Neutron Irradiation Embrittlement of Pressure Vessel Steels

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ABSTRACT: The paper describes work on irradiation embrittlement that has been undertaken in the United Kingdom since preparation of the Second Marshall Report on the Integrity of pressure water reactor (PWR) pressure vessels. Results from research programs concentrate specifically on work on steels for Magnox and PWR pressure vessel (PV) applications.

Models have been developed that describe the influence of copper precipitation on the yield stress changes in Magnox PV steels. Results from strength and microscopic examination confirm the significance of copper precipitation after long-term aging. The results provide support to the expression in the model to describe the time dependence of the copper precipitate contribution to the increase in yield strength.

Accelerated irradiation and surveillance data are present for both the Magnox and PWR PV steels. Modeling of data includes variations on neutron energy spectrum, neutron fluence, flux intensity, and irradiation temperature. The data for PWR PV materials have been used to examine the applicability of a recent draft revision of the U.S. Nuclear Regulatory Commission's Regulatory Guide 1.99 for estimating irradiation shifts on modern materials.

A study of mechanisms of embrittlement is reported including developments in positron annihilation studies and small-angle neutron scattering (both with and without the application of magnetic fields).

KEY WORDS: radiation effects, irradiation embrittlement, pressure vessel steels, pressure water reactors, neutrons, mechanical property changes, Charpy tests, yield stress, copper, phosphorus, mechanisms, modeling

In this paper, an attempt is made to present an overview of studies in the United Kingdom on neutron irradiation embrittlement of pressure vessels steels. It would be a difficult task to provide a comprehensive review of work in the United Kingdom. This paper is therefore essentially a partial view. We have

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restricted the paper to an outline of the findings, in this area, of a major recent study, and we go on to consider the implications of those on the current scene in the United Kingdom. Attention is drawn to a recent draft revision to the U.S. Nuclear Regulatory Commission's (NRC) Regulatory Guide 1.99. Surveillance and Materials Test Reactor (MTR) data are considered in this context, and it is shown that the proposal is unrealistically pessimistic for welds typical of modern practice. We then proceed to provide a partial view of some recent developments in studies of Magnox steels because of their relevance to the general area of pressure water reactor pressure vessel (PWR PV) studies. Finally, we produce some closing comments that indicate areas of further study.

PWR Studies

Light Water Reactor Study Group

A Light Water Reactor Study Group (LWRSG) on the assessment of PWR PV integrity has existed in the United Kingdom since 1973. The first LWRSG report was published in 1976 [*1*]. The Study Group undertook the task of updating its first report in December 1979 and completed the task by December 1981. The second report [2] provided an evaluation of modern PWR PV technology and a comprehensive fracture assessment of a typical pressure vessel. The Study Group recommendations were classified as either "essential" or "for improved understanding," the former being regarded as necessary for the safe operation of PWRs installed in the United Kingdom. A pre-construction safety report (PCSR) for a PWR station at Sizewell in Suffolk has been prepared by the Central Electricity Generating Board (CEGB) and the National Nuclear Corporation (NNC) and submitted to H. M. Nuclear Installations Inspectorate (NII). The proposal to build the Sizewell 'B' PWR is currently the subject of a Public Inquiry. Deliberations at the Inquiry should be completed by the end of 1984 and the Inspector is expected to present his report to the Government at a later date.

Section 3 of the second Study Group report [*2*] discusses the mechanical and fracture toughness properties of pressure vessel materials.

In the context of this overview, three mechanisms were identified by the Study Group that could lead to a degradation of material properties during service life. These were: irradiation embrittlement, thermal aging, and strain aging. The

TABLE 1—*Specification for ASME SA508 Class 3 chemical composition (percent by weight).*

Carbon	0.25 max
Manganese	1.20 to 1.50
Phosphorus	0.025 max
Sulfur	0.025 max
Silicon	0.15 to 0.40
Nickel	0.40 to 1.00
Chromium	0.25 max
Molybdenum	0.45 to 0.60
Vanadium	0.05 max

TABLE 2—*Additional compositional requirements (percent by weight, maximum) for the Sizewell 'B' reactor vessel.*

	Forgings	Weld Metal
Carbon	0.20	0.15
Phosphorus	0.008	0.01
Sulfur	0.008	0.01
Copper	0.09	0.07
Vanadium	0.01	0.01
Aluminum	0.045	...
Antimony	0.008	0.008
Arsenic	0.015	0.015
Cobalt	0.02	0.02
Hydrogen	1 ppm (product)	...
Nickel	0.85	0.85
Tin	0.01	0.01
Silicon	0.3	...
Chromium	0.15	0.15

degree of embrittlement accruing from these mechanisms can be minimized by specifying pressure vessel materials having closely controlled chemical compositions that are readily achievable using current commercial practice.

A United Kingdom PWR PV will be fabricated using A508 Class 3 ring forgings. The American Society of Mechanical Engineers (ASME) specification for A508 Class 3 steel and the additional compositional requirements that have subsequently been imposed for the Sizewell 'B' pressure vessel are shown in Tables 1 and 2.

The principal elements that enhance the irradiation sensitivity of SA508 Class 3 steel and associated welds are copper and phosphorus. The upper limits on these elements have been restricted to Cu = 0.09% and P = 0.008% by weight for forgings and Cu = 0.07% and P = 0.010% by weight for welds. It can be seen from Table 3 that the maximum end-of-life neutron dose at the beltline of a four-loop PWR PV will be 2.3×10^{19} n/cm^2 ($E > 1$ MeV), whereas that for the weld closest to the reactor core will be only 5.8×10^{18} n/cm^2 ($E > 1$ MeV). The mechanical properties would not be expected to be changed significantly by the lower total dose experienced at the girth weld.

An analysis of materials test reactor data indicated that modern materials typical

TABLE 3—*End-of-life neutron doses in a four-loop PWR pressure vessel (40 year life).*

Position	Neutron Dose, n/cm^2 ($E > 1$ MeV)
Belt Line	
(i) Inner wall	2.3×10^{19}
(ii) ¼ T	1.3×10^{19}
(iii) ¾ T	2.7×10^{18}
Girth Weld	5.8×10^{18}

of current practice irradiated to a neutron dose of 3×10^{19} n/cm^2 ($E > 1$ MeV) would experience changes in transition temperature not exceeding 30°C [*2*]. Few data were available to assess changes in upper-shelf toughness when modern materials are irradiated but the limited evidence suggested that effects of irradiation, up to dose levels of interest, would not be significant. In view of the scarcity of data, the Study Group concluded that the effect of irradiation on upper-shelf toughness should be determined directly for relevant materials. The "essential" recommendation from the Study Group concerning embrittlement was:

> All analytical and mechanical tests used in US quality control procedure trials and irradiation surveillance should be included in the requirements for a vessel to be installed in the United Kingdom. In addition fracture toughness tests, for example on 12.5 mm thick compact tension specimens, or thicker as appropriate, should be included in the United Kingdom surveillance programme.

Precipitation, both within grains and on grain boundaries, and segregation of certain impurity elements, which may occur during long-term aging at 300°C, could contribute to an increase in the brittle-ductile transition temperature. The Study Group acknowledged that both chemical composition and fabrication procedures should be adequately controllable so that these thermal aging effects could prove to be insignificant. It was considered prudent, however, to include some allowance in fracture assessments for an increase in transition temperature, in nonirradiated regions, which would allow for any effect of thermal aging.

Strain aging promotes an increase in the brittle-ductile transition temperature and a reduction in both upper and lower-shelf toughness. The Study Group [*2*] noted that the strain aging phenomena cannot be isolated from the effect of warm prestressing, and that warm prestressing will result in a beneficial effect on the transition temperature—so that the combined effect of strain aging and warm prestressing on the transition temperature shift could be either positive or negative.

The Study Group concluded that there was insufficient knowledge concerning the extent to which the changes in transition temperature due to irradiation embrittlement, thermal aging, and strain aging would be additive. Taking into account the improved response of modern materials to such phenomena, the Study Group recommended that applying a transition temperature shift of 30°C to *all* regions of the vessel should encompass any toughness degradation during the operational life of a modern PWR PV.

Aspects relating to factors that may affect in-service material properties are covered in the following "Recommendations for Improved Understanding:"

> The materials surveillance programme should include specimens typical of the various components and welds of the vessels to evaluate the effect of long-term ageing, strain ageing and combined effects with neutron irradiation where appropriate at temperatures of about 300°C. It is expected that the effects on the transition temperature and the toughness values in modern steels and welds will be very small but sufficient

testing should be undertaken to show that this is the case for actual materials used for a particular vessel.

The inservice material surveillance programme should be supplemented by a programme to evaluate the effect of long-term thermal ageing, strain ageing, and combined effects with neutron irradiation where appropriate) at about 300°C on the fracture toughness of base materials and weld metal and also the temperature variation of sensitivity to these effects between about 250°C and 360°C.

Studies Related to Sizewell 'B'

Although the Study Group report was a generic study and did not consider the specific case of the Sizewell 'B' design, in developing their Safety case, the CEGB and NNC have taken account of the Study Group recommendations.

When considering the effect of irradiation on the shift in the brittle-ductile transition temperature, the case for Sizewell 'B' involved a more cautious approach by assuming that the shift will be not more than 50°C. This higher shift is based on both surveillance and MTR data for relevant materials and also evidence that the neutron fluxes shown in Table 3 could vary by ±20%. These results are summarized in Fig. 1. (Some of the data in this figure relate to higher phosphorus contents than the maximum level of 0.010% by weight for Sizewell 'B').

The effects of thermal and strain aging are considered to be minimal for the materials to be used in Sizewell 'B', but will be allowed for by assuming a 30°C shift in transition temperature for those parts of the vessel outside the core region.

Exact details of the surveillance program that would be used for Sizewell 'B' have yet to be decided. The Pre-Construction Safety Report and evidence to the Sizewell 'B' Power Station Public Inquiry indicate that the program will use standard Westinghouse-designed capsules. These will be located in guide baskets welded to the outside of the neutron shield pads and positioned directly opposite the center portion of the core. The program will use six capsules. A typical Westinghouse capsule would contain tension, Charpy V-notch impact, and fracture toughness specimens for base metals and welds in the proportions shown in Table 4. However, in view of the considerably reduced significance of the response to irradiation of girth welds and associated heat affected zones fabricated to the Sizewell 'B' specification (see Table 3), the exact numbers of specimens of a given material may well vary from the basic Westinghouse package.

The fracture toughness samples are likely to be 12.5-mm-thick compact specimens. These particular samples will be used to monitor the effect of irradiation on upper-shelf toughness. Where appropriate, the surveillance program will, as a minimum, conform with ASTM Practice for Conducting Surveillance Tests for Light Water-Cooled Nuclear Power Reactor Vessels (E 185-82). Details of the dosimeters that will be used to evaluate the neutron doses accumulated by the samples and the vessel wall have still to be finalized.

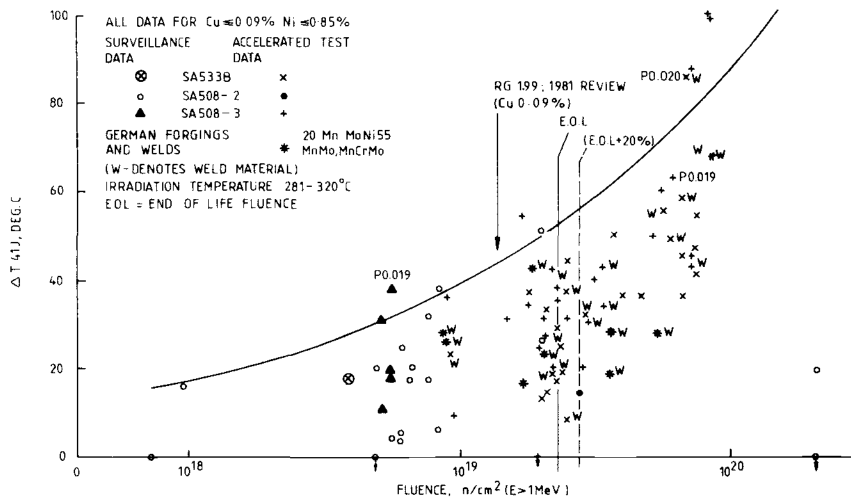

FIG. 1—*Reactor surveillance and accelerated test results on PWR PV steels with ≤0.09% by weight copper and ≤0.85% by weight nickel (Phosphorus contents are generally within 0.010 to 0.012% by weight, but certain high phosphorus points are identified.)* [49].

The U.S. Regulatory Position Concerning the Prediction of Neutron Damage

In the United States, estimates of neutron radiation damage are achieved using trend curves that are embodied in both Design Codes [*3,4*] and the U.S. Code of Federal Regulations [*5*]. Current regulations invoke the NRC Regulatory Guide 1.99 Revision 1 1977 [*6*]. The increase in transition temperature due to neutron damage is calculated using the expression:

$$A = T_{41}\mathrm{J} = \frac{5}{9}\,[40 + 1000(\%\mathrm{Cu} - 0.08) + 5000(\%\mathrm{P} - 0.008)] \times \left[\frac{f}{10^{19}}\right]^{1/2} {}^{\circ}\mathrm{C} \quad (1)$$

TABLE 4—*Contents of typical Westinghouse surveillance capsule.*

Material	Charpy Specimens	Tension Specimens	Fracture Toughness Specimens
Forging:			
Hoop direction	15	3	4
Axial direction	15	3	4
Weld metal	15	3	4
Heat affected zone	15	...	...

where

T_{41}J = the transition temperature shift in °C indexed at 41-J Charpy V-notch impact energy,
f = fluence in n/cm^2 ($E > 1$ MeV), and
%Cu, %P = the amounts of copper and phosphorus in percent by weight.

The relationship is valid for $A > 27.8$°C and $f < 6 \times 10^{19}$ n/cm^2 ($E > 1$ MeV). For Sizewell 'B', the material specification will call for copper and phosphorus contents of, respectively, 0.09 max and 0.008 max for base metals and 0.07 max and 0.010 max for weld metals. Inserting these values into Eq 1 provides the same limiting trend curve for both materials that reduces to

$$T_{41}\text{J} = 27.78 \left[\frac{f}{10^{19}}\right]^{1/2} \text{°C} \tag{2}$$

Regulatory Guide 1.99 Rev. 1 is thought to be overly conservative at high fluences and the NRC is actively pursuing methods to revise the curves and thus reduce the conservatism. The first tentative steps towards revision were described at the 1981 International Atomic Energy Agency (IAEA) Specialists Meeting [*7*] where separate trend curves were presented for materials of low and high (>0.5% by weight) nickel content. The tentative trend curves for high nickel materials, (which would be relevant to Sizewell 'B') are given by

$$\Delta T_{41}\text{J} = \frac{5}{9}\,[30 + 1000(\%\text{Cu} - 0.05)] \left[\frac{f}{10^{19}}\right]^{0.35} \text{°C} \tag{3}$$

Limiting curves for Sizewell 'B' materials would be for base materials

$$\Delta T_{41}\text{J} = 38.89 \left[\frac{f}{10^{19}}\right]^{0.35} \text{°C} \tag{4}$$

and for weld metals

$$\Delta T_{41}\text{J} = 27.78 \left[\frac{f}{10^{19}}\right]^{0.35} \text{°C} \tag{5}$$

The trend curves defined by Eqs 2, 4, and 5 are compared in Fig. 2. The end-of-life (EOL) fluences shown in the figures are those given in Table 3.

The 1981 Trend Curves have been superseded by a recent draft proposed revision to Regulatory Guide 1.99 [*8*]. These current NRC proposals were produced by combining correlation functions for surveillance data that had been

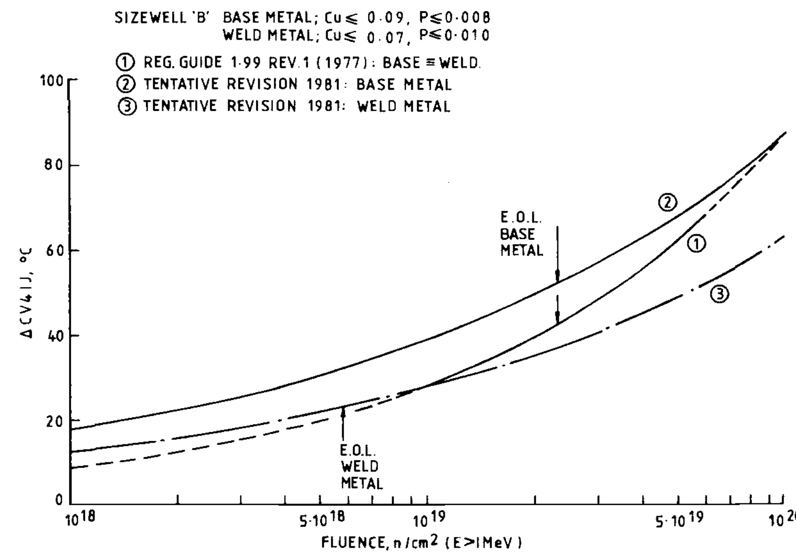

FIG. 2—*Comparison of predicted shifts in transition temperature for Sizewell 'B' materials using Regulatory Guide 1.99 Rev. 1 (1977) and tentative revisions made in 1981* [7].

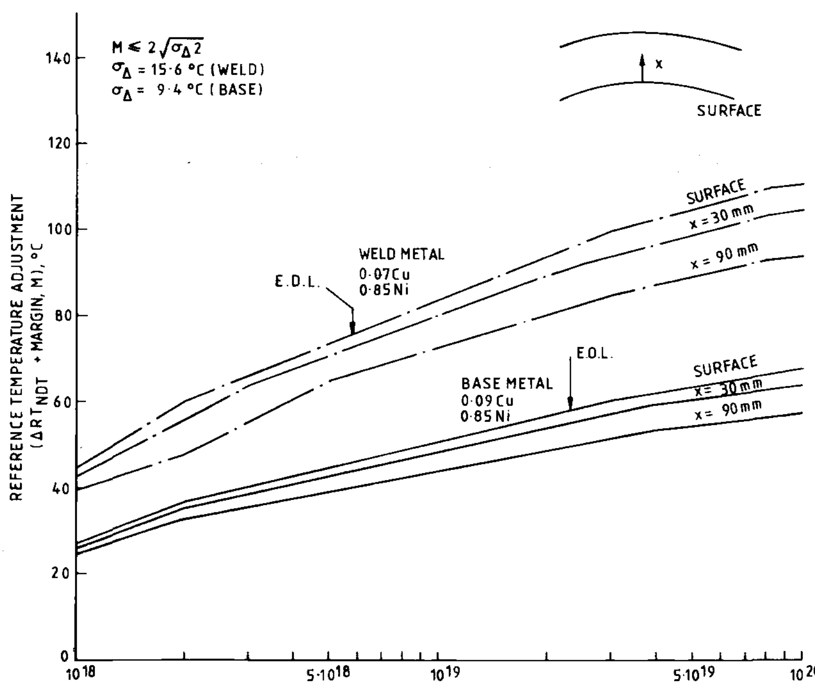

FIG. 3—*Predicted upper limits on shifts in transition temperature from draft revision of Regulatory Guide 1.99* [8].

derived independently by Guthrie [9] and Odette [10]. The mean irradiation-induced shift is given in the new proposal by

$$\Delta RT_{\mathrm{NDF}} = \frac{5}{9}\,[\mathrm{C.F.}] \cdot f^{(0.28-0.10\,\log f)}\ ^\circ\mathrm{C} \tag{6}$$

where

C.F. = a chemistry factor based on the copper and nickel contents that is tabulated separately for base and weld materials, and
f = fluence normalized to 1×10^{19} n/cm^2 ($E > 1$ MeV).

Conservative adjusted reference temperatures (ART) are defined by

$$\mathrm{ART} = \text{Initial } RT_{\mathrm{NDT}} + \Delta RT_{\mathrm{NDT}} + \text{Margin} \tag{7}$$

where

$$\text{Margin} = 2 \cdot \sqrt{\sigma_I^2 + \sigma_\Delta^2},$$

where

σ_I, σ_Δ = standard deviations for Initial RT_{NDT} and RT_{NDT},
σ_I = 0 if initial RT_{NDT} values are measured, and
σ_Δ = 9.4°C for base metals and 15.6°C for weld metals or 0.5, RT_{NDT} whichever is the smaller.

The proposal also allows credit to be taken of the attenuation of neutron embrittlement with increasing penetration from the inner-wall surface of the pressure vessel. This attenuation factor is given by

$$\Delta RT_{\mathrm{NDT}}(x) = \Delta RT_{\mathrm{NDT}}(\mathrm{Surface}) \times e^{-0.067x} \tag{8}$$

where x is measured in inches.

Limiting trend curves for Sizewell 'B' base metals and weld metals, derived using Eqs 6, 7, and 8, are presented in Fig. 3. The trend curves corresponding to positions 30 and 90 mm from the inner wall merely illustrate the allowance that can be taken for damage attenuation and have no specific significance with regard to PV integrity. The most striking feature in Fig. 3 is the significant increase in predicted shifts in reference temperature for the weld metal compared to those for the base metal. (It should be noted that the situation would be further aggravated for RPVs fabricated from plates where welds would be subject to neutron damage at peak flux.) The prediction of high shifts in reference temperature for weld metals is directly related to the inclusion of a higher chemistry factor for welds, the relatively high upper limit on nickel content (0.85), and inclusion of a margin, M, which is high (typically 100%) relative to the predicted mean shift, ΔRT_{NDT}. In practical terms, the curves suggest that the girth weld, although well removed from the core region and receiving only minimal neutron flux (5.75×10^9 n/cm^2/s assuming 32 full-power years operation, 40 year life), would be a critical location on the basis of this Revision, for design assessments, and could be critical for in-service fracture assessments. Such a situation is at variance with our understanding of the sensitivity to irradiation embrittlement of modern PWR PV materials. A recent IAEA Coordinated Research Programme (CRP) [*11*] has demonstrated that, for materials having closely controlled chemical compositions, there is no significant difference between the irradiation-induced shift in transition temperature for base metals and associated welds. Relevant results from this program are shown in Fig. 4. Those results and the results shown in Fig. 1 indicate similar response from base materials and welds. Thus, available data for modern materials suggest that the current draft proposal for the Regulatory Guide revision will be unduly conservative when used to predict transition shifts for modern low-copper welds. One motivation behind the proposed revisions has been to reduce the degree of conservatism when applying Regulatory Guide 1.99 Rev. 1 in assessments of over-cooling transients. Such transients are of concern mainly in older RPVs fabricated from materials having less carefully specified chemical compositions. Recognition of possible problems of this nature at the design state has allowed them to be largely eliminated, for example, by using materials having closely controlled chemical compositions and incorporating specific design features.

Therefore, it would appear to be essential to give further detailed consideration to the current proposals for revising Regulatory Guide 1.99 to preclude the possibility of introducing unrealistically high predictions of irradiation-induced transition temperature shifts in modern materials, particularly weld metals fabricated using state-of-the-art welding technology.

Some Current Studies on the Embrittlement of PWR PV Steels

Druce et al [*12–15*] at Harwell are investigating the effects of thermal aging treatments in the temperature range of 300 to 550°C for times up to 20 000 h on a variety of steels that include A533 B Class 1 plate and A508 Class 3 forging materials (both as base materials and as simulated heat affected zone material) and also weldments. These studies show that base metal steels typical of modern practice are highly resistant to aging in the temperature range of 300 to 500°C. Typical increases in ductile-brittle transition temperature over the temperature range of their study were observed to be generally 40°C, while most specimens aged at 300°C showed no effects. Work on model alloys and simulated heat affected zone by Druce [*12*] and Brear and King [*16*] show the deleterious effects of large grain sizes (~200 μm) and impurity content. Phosphorus and antimony are significant in their effect and arsenic and tin can also promote embrittlement. Brear and King have suggested limits on arsenic, antimony, and tin of 0.048, 0.011, and 0.015% by weight, respectively, to produce effects equivalent to the maximum phosphorus content permitted by ASME for the PV beltline material

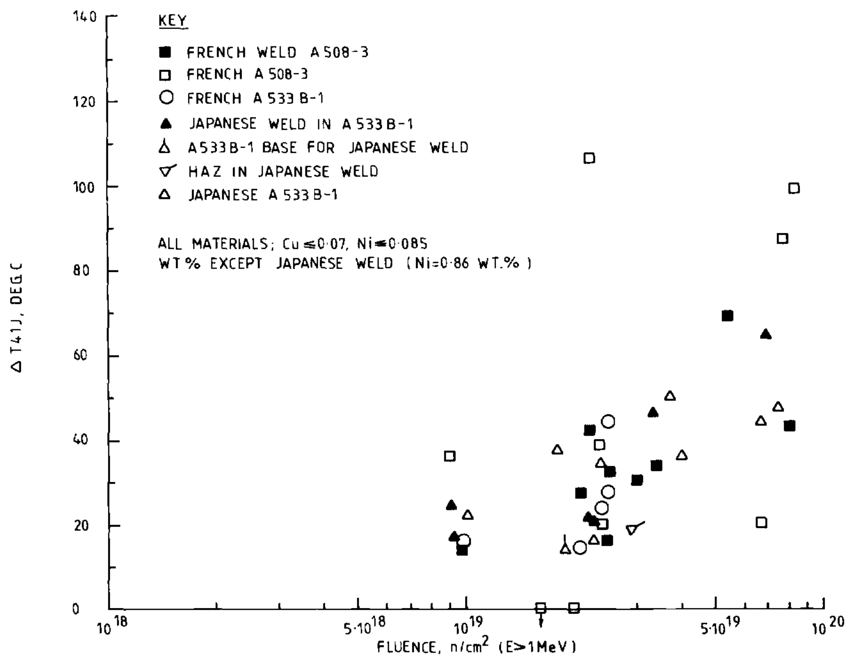

FIG. 4—*Comparison of Charpy 41-J transition shifts for weld metals and associated base metals tested in the IAEA CRP (after Ref* 11).

(0.012% by weight). Generally, lower austenitization temperature showed lower embrittlement on aging but coarser structures characteristic of higher austenitization temperatures promoted a greater degree of embrittlement on aging at the higher temperatures. However, aging at 300°C showed little effect.

Little [*17*] has investigated the combined effects of strain aging and irradiation at a temperature of about 290°C to a fluence of 3.1×10^{19} n/cm^2 ($E > 1$ MeV) on A533 B Class 1 and A508 Class 3 steel containing free nitrogen levels of 14 and 8 ppm, respectively. Some of the specimens were given a tensile prestrain of 5%. Some specimens were also thermally aged at 290°C. He found that C_v shifts in transition temperature due to strain aging were not additive to the corresponding shift from neutron irradiation because the irradiated prestrain and nonprestrained results were essentially coincident and because irradiation appears to suppress the C_v transition shift from strain aging.

For Phase 3 of the IAEA Coordinated Research Programme on optimizing RPV surveillance programs and their analyses, the UK contribution contains, as a cornerstone of its proposal [*18*], an investigation of the combined effects of irradiation embrittlement and strain age embrittlement on the fracture toughness temperature transition using 12.5-mm-thick compact specimens some of which will have been prestrained at ambient temperature prior to irradiation to an equivalent K_I value of 120 MPa$\sqrt{\text{m}}$. Results will be compared with those from Charpy tests and tension tests in the unirradiated, unirradiated and aged, and also

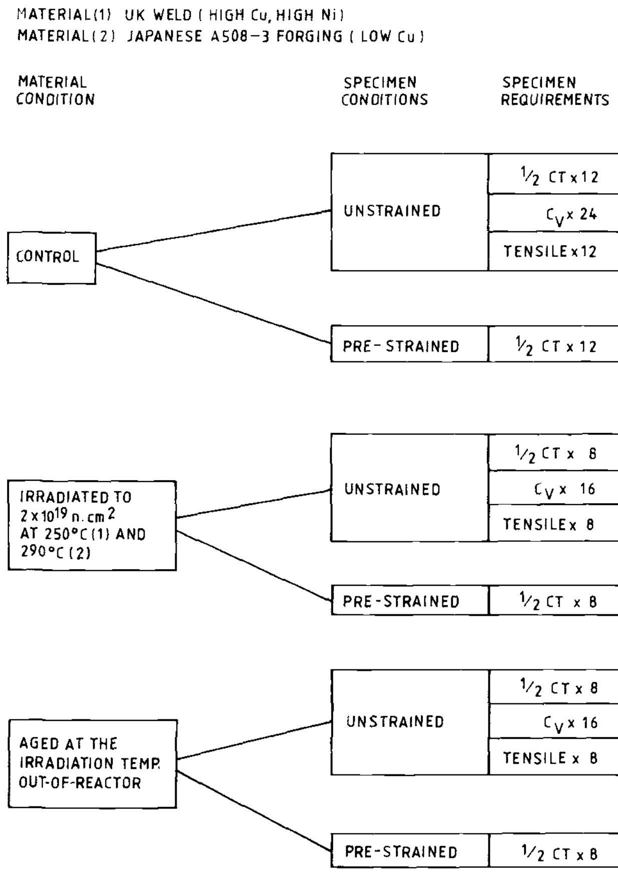

FIG. 5—*Deployment of specimens for the strain-aging experiment within the UK contribution to the IAEA-CRP* [18].

in the irradiated condition. The deployment of specimens for that part of the proposal is shown schematically in Fig. 5.

Small-angle neutron scattering techniques have been employed in the investigation of several irradiated pressure vessel steels [*17,19*] that had shown significant changes in mechanical property on irradiation but had shown little change in microstructure during electron microscope examination [*20*]. Enhanced scattering in PWR PV steels was related to the copper content but was affected by the presence of nickel and other elements. Figure 6 is from Jones and Buswell [*19*] and includes steels from Phase 2 [*20*] of the IAEA CRP (steels JW, Japanese weld containing 0.04% by weight copper; FF, French forging containing 0.06% by weight copper and the radiation sensitive UKW 0.2% by weight copper, 1.57% by weight nickel; Steels A and B were model alloys containing 0.01 and 0.22% by weight copper, respectively). An increase in the scattering intensity with increasing Varsik and Byrne Chemistry Relationship [*21*] can be seen in Fig. 6. If the results were plotted against copper content only, then the results on UKW are located nearer the *x* axis in such a graph and evidence for a continuous enhanced scattering with both increasing copper and nickel is lost. Little [*17*] also confirms the additional scattering associated with increasing both nickel and phosphorus content.

Small-angle neutron scattering studies involving the determination of the ratio of the degree of scattering perpendicular to and parallel to magnetic fields applied to specimens have been carried out [*19*] to identify the nature of the scattering centers. (It will be remembered that Frisius et al [*22*] calculated ratios of 11.5 for pure copper precipitate and 1.3 for voids, and their measurements on model binary alloys gave ratios of 11.0.) Jones and Buswells [*19*] work provided ratios of about 2.5 for irradiated PWR steels and the UK weld used for Phase 2 of the IAEA [*20*] CRP that was very sensitive to irradiation. This work again provides a clear indication that real PWR PV steels behave in a manner more complex during neutron irradiation than model alloys. Such experimental and interpretive work is continuing [*23*].

Highton [*24*] has reported on the results from his positron annihilation studies where he found an increasing signal as a function of copper or chemistry relationship in unirradiated steels (He also reported on his observations on an increase in hardness as a function of heat treatment temperature in unirradiated PV steels.) Those studies are continuing [*25*] and preliminary results on model alloys indicate

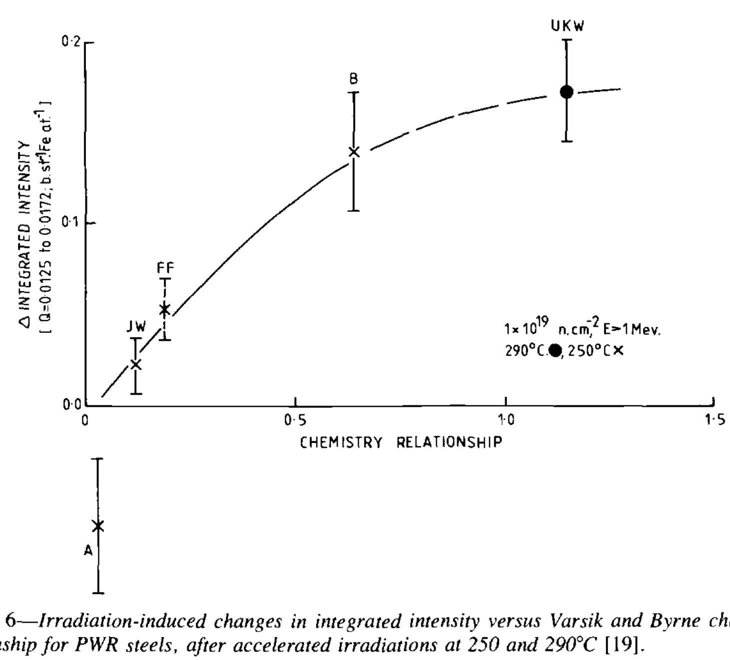

FIG. 6—*Irradiation-induced changes in integrated intensity versus Varsik and Byrne chemistry relationship for PWR steels, after accelerated irradiations at 250 and 290°C* [19].

an increasing signal (S value) as a function of decreasing irradiation temperature for the same composition.

Recent Magnox PV Steel Studies of Relevance to PWR PV Studies

Current assessments of Magnox pressure vessel integrity depend upon the original proof test using analyses based on fracture mechanics. Thus, values of fracture toughness are required that are relevant to the state of the vessel materials both at the time of the initial proof test and after a period of operation. For the proof test condition, values have been obtained from recent tests on material cut from the original reactor vessels at the time of construction. To obtain fracture toughness appropriate to material that has been in service and has thus been subjected to neutron irradiation, a number of approaches have been adopted:

(*a*) Techniques have been developed to allow the original monitoring scheme Charpy specimens to be used. Instead of impact testing, specimens are fatigue pre-cracked, side-grooved, and subjected to three-point bend loading. Papers outlining the analytical background to this work, along with analysis procedures, have been published by Neale [*26,27*]. The implementation of the techniques to hot-cell fracture toughness testing have been given by Neale and Priest [*28–30*] and a description of the CEGB's shielded fracture toughness testing facility was reported by Neale and Priest [*31*].

(*b*) The original monitoring schemes have been supplemented with standard fracture toughness specimens. Clearly, it will be some time before these will have experienced sufficient exposure to make testing worthwhile.

(*c*) Accelerated irradiation experiments have been undertaken. These have included tension, impact, and fracture toughness tests using both pre-cracked Charpy specimens and compact specimens. Data from these tests have been discussed by Priest, Charnock, and Stewart [*32*] and Priest, Charnock, and Neale [*33*].

In a recent series of papers Fisher et al [*34–36*] have presented a simple model to explain the influence of copper precipitation on the yield strength changes observed in Magnox PV steels. In terms of neutron irradiation embrittlement, the composition of older non-nickel modified PWR PV steels is generally similar to Magnox PV steels (see, for example, Table 5, which is reproduced in simplified form from Ref *34*). The main differences are the generally higher copper, molybdenum, and carbon content of the PWR PV steels, and the lower nickel content in the Magnox steels. The differences in heat treatment and composition of Magnox and PWR PV steels are manifested in a difference in microstructure. The Magnox plate metallographic structure is ferrite/pearlite compared with tempered bainite/ferrite structures in PWR PV steels.

Fisher et al [*34*] have surveyed relevant studies on irradiation embrittlement. Their survey includes the period covering the results from the earlier work of

TABLE 5—*Typical analyses of the plate steels exposed in the low-temperature Magnox locations* [34].

Station	C	Si	Mn	S	P	Cu	Ni	Sn	Cr	Mo	Co	Ti	Al	Nb
1	0.19[a]	0.3	1.22	0.027	0.015	0.12	0.11	0[b]	0.1	0.03	0	0	0.01[c]	0
2	0.09	0.37	1.05	0.02	0.015	0.06	0.06	0.01	0.04	0	0.03	0.005[c]	0.008	0.01[c]
3	0.11	0.13	1.1	0.02	0.015	0.43	0.07	0.01	0.07	0.23	0.03	0.005[c]	0.018	0.01[c]
4	0.15	0.16	1.3	0.035	0.02	0.09	0.06	0.01	0.01	0.01	0.02	0.01[c]	0.051	0.01[c]
5	0.17	0.25	1.1	0.034	0.016	0.15	0.08	0.02[c]	0.07	0.02[c]	0.02[c]	0.02[c]	0.02[c]	0.02[c]

[a] Values are in percent by weight.
[b] 0 indicates not determined.
[c] Indicates upper limit.

Potapovs and Hawthorne [*37*] that identified the detrimental effect of copper, through to the recent Field Ion Microscope studies [*38–40*] where Lott et al [*40*] confirmed the presence of copper-rich clusters in PWR steel irradiated to a neutron fluence of about 2 × 10^{19} n/cm^2 at 290°C. The small-angle neutron scattering studies of Frisius et al [*22*] led to the conclusion that the neutron irradiation embrittlement of the PV steels containing small quantities of copper is caused, at least partially, by radiation-induced precipitation of copper. Hawthorne's [*41*] observation, that PWR PV steels having high sulfur content reduced the detrimental effect of copper, is seen as confirming the observation of the presence of a copper-containing compound $Cu_{1.8}S$ [*42*] seen to be present in some Magnox steels. (The presence of this compound could provide misleading estimates of the amount of copper in solid solution in the steels because chemical analysis gives total copper content. Hence, bulk copper content may not be the most appropriate parameter to use in differentiating between steels of different sensitivity.)

In the studies on Magnox steels that support the model, copper precipitates 140 Å in. diameter were observed on aging for 20 000 h at 350°C and the measured increase in strength arising from the precipitation of copper was consistent with the value predicted from the theory of Russell and Brown [*43*]. The increase in yield strength detected in some surveillance specimens containing 0.4% by weight copper irradiated at 390°C derived solely from the precipitation of copper. (For a fuller description of the surveillance data and the thermal aging work, the reader is referred to Refs *34, 35,* and *36*.) The contribution to the change in yield strength from damage clusters is negligible at these irradiation temperatures (350 to 390°C) and the experimental data already available on model steels and ferritic steels show that copper precipitation will occur rapidly even without any enhancement from neutron irradiation, at these temperatures. For steels operated at lower temperatures (about 170 to 225°C), an increase in yield strength from copper precipitation would be expected from these alloys, but an enhancement of the precipitation process is expected during irradiation.

One input into the model assumes that the strengthening from dislocation loops on neutron irradiation is described by and, derived from the following equation, which is usually used to describe the change in yield strength on irradiation (after making allowances for composition and neutron spectra)

$$\Delta\sigma_{\mathrm{damage}} = A(\phi t)^{1/2}$$

where A is a function of the irradiation temperature and aluminum or silicon content.

The copper in solution in the steels prior to irradiation is assumed to precipitate during irradiation, and the rate of precipitation is dependent on the rate of copper diffusion and, in turn, of the mobility and concentration of vacancies. (During irradiation, there is a supersaturation of vacancies so that the precipitation rate is enhanced to a temperature above the corresponding thermal temperature.)

Interestingly, the estimated size of copper precipitates is such that they would remain below the limit of resolution of electron microscope examination throughout the lifetime of Magnox stations.

Under irradiation at a temperature (T_{irr}), a reduced time to develop peak strength (tp') is calculated from the relationship

$$tp' = \left[\frac{C_v \text{ (thermal)}}{C_v \text{ (irradiated)}}\right] tp$$

where

C_v (thermal) = the thermal equilibrium concentration of vacancies;

C_v (irradiated) = the calculated vacancy concentration under irradiation at T_{irr}; and

tp = time to peak strength, unirradiated, but at T_{irr}, from existing data.

The maximum increase in yield strength from a particular copper concentration has been derived from iron-copper alloys and is described by the following equation

$$\Delta\sigma_{max} = [3.6 \times 10^3(f)^{1/2} - 60] \text{ MN/m}^2$$

where f is the volume fraction of copper.

Having thus evaluated the separate contribution to yield strength of the damage loops, Fisher et al evaluate the overall superposition of the effects by the following summation

$$\Delta\sigma_{total}(t) = \Delta\sigma_{Cu}(t) + \Delta\sigma_{dam}(t)$$

Overall, this study has allowed the successful interpretation of yield stress measurements on plate steel monitoring specimens in low-temperature Magnox reactors.

In the small-angle neutron scattering experiments, mentioned earlier in this paper, by Jones and Buswell [*19*], results were given of a comparison between Magnox surveillance specimens irradiated at 220°C and accelerated tests at 180°C. Their results are reproduced, for convenience, as Fig. 7 where it can be seen that the results support the Fisher et al model that a fine distribution of copper particles is precipitated during long-term irradiation but that the process is less advanced in accelerated irradiations.

Buswell [*44*] has also compared results of irradiation-induced changes of proof stress and ductile–brittle transition temperatures from PWR surveillance data from Electric Power Research Institute (EPRI) [*45*] and MTR data from other published sources. He concluded that it was generally acceptable to consider the changes

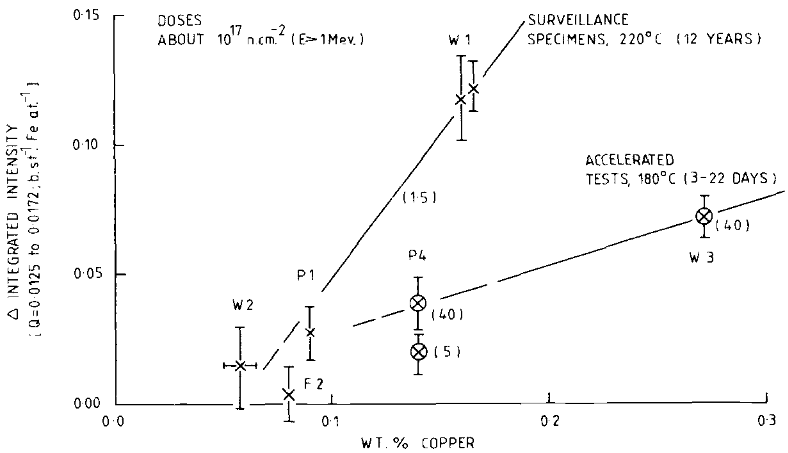

FIG. 7—*Irradiation-induced changes in integrated intensity versus copper content for Magnox steels, after long-term surveillance and short-term accelerated irradiations* [19].

in terms of copper content alone but the general agreement between surveillance and MTR data was lacking. The significant difference was that MTR data tended to show larger effects on steels typical of modern practice with copper contents less than about 0.1% by weight after irradiation to neutron doses of 1 to 2 $\times$ 10^{19} n/cm^2 ($E > 1$ MeV) with the implication that results from MTR tests on such materials will be conservative for design application.

Buswell [*44*] also comments on the model produced by Odette [*46*], which is somewhat similar to that development by Fisher et al [*36*], but was developed to describe and interpret the EPRI data base on PWR PV surveillance results [*45*].

Closing Comments

Knowledge of the features underlying the strengthening processes in PWR PV steels during neutron irradiation has grown markedly in the past few years. In this paper, we have drawn attention to some of the recent developments in the United Kingdom, in particular, the encouraging agreement obtained between observed and predicted changes in yield stress of Magnox surveillance steels irradiated over a range of temperatures, fluxes, and spectra. Certainly, the initial application of such modeling techniques to PWR steels is showing promising signs of a capability to account for the changes found in both MTR and surveillance irradiations. However, there are additional features that need to be investigated further and the following are some examples: (1) there is evidence [*40*] for Mo_2C precipitates in PWR PV test specimens during irradiation, and this feature may provide an additional hardening source term for modeling mechanisms; (2) the role of nickel in aiding embrittlement when copper is present to significant levels needs to be understood for the analysis of older PWR PVs; (3)

the role of phosphorus requires to be understood; (4) recovery processes during annealing have been explained [*47*] by mechanisms different from those responsible for strengthening, and the mechanisms involved in strengthening and recovery should be consistent; (5) the increase in *S* value in positron annihilation studies with increase in both copper content and decreasing irradiation temperature [*35*] would indicate an increasing vacancy concentration that seems to contradict the mechanisms assumed in current models; etc.

There is a continued need to develop data bases on materials typical of current practices and from various sources of supply. In order to do this, there is a requirement for the continued development and standardization of test methods and techniques for both surveillance and post-irradiation testing processes. Many of these problems are being addressed in various national programs and in the phases of the IAEA Coordinated Research Programme and other international programs, such as the Surveillance Dosimetry Improvement Programme [*48*], but much work still remains to be done to understand irradiation embrittlement.

References

[*1*] Marshall, W. (chairman), "An Assessment of the Integrity of PWR Pressure Vessels," United Kingdom Atomic Energy Agency, 1976.

[*2*] Marshall, W. (chairman), "An Assessment of the Integrity of PWR Pressure Vessels," Second Report, United Kingdom Atomic Energy Agency, 1981.

[*3*] *ASME III Appendix G*, American Society of Mechanical Engineers, 1983.

[*4*] *ASME XI Appendix A*, American Society of Mechanical Engineers, 1983.

[*5*] 10 CFR 50, U.S. Code of Federal Regulations, 1980 (Fracture Toughness and Surveillance Program requirements).

[*6*] "Effects of Residual Elements on Predicted Radiation Damage to Reactor Vessel Materials," Regulatory Guide 1.99, Revision 1, U.S. Nuclear Regulatory Commission, Washington, DC, April 1977.

[*7*] Randall, P. N., "The Status of Trend Curves and Surveillance Results in US NRC Regulatory Activities," IAEA Specialists Meeting, Vienna, International Atomic Energy Agency, Oct. 1981.

[*8*] Randall, P. N., private communication to T. Ingham of "Draft Proposed Revision to Reg Guide 1.99," 14 June 84.

[*9*] Guthrie, G. L., "Charpy Trend Curves Based on 177 PWR Data Points," LWR PV SDIP, Quarterly Progress Report April 1983–June 1983, NUREG/CR-3391, Vol. 2, HEDL-TME 83-22, U.S. Nuclear Regulatory Commission, Washington, DC.

[*10*] Odette, G. R. and Lombrozo, P. M., "Physically Based Regression Correlations of Embrittlement Data from Reactor Vessel Surveillance Programmes," EPRI NP-3319, Final Report, Electric Power Research Institute, Jan. 1984.

[*11*] Steele, L. E., Davies, L. M., Ingham, T., and Brumovsky, M. in *Effects of Radiation on Materials: Twelfth Conference, ASTM STP 870*, F. A. Garner and J. S. Perrin, Eds., American Society for Testing and Materials, Philadelphia, 1986, pp. 863–899.

[*12*] Druce, S. G. and Edwards, B. C., *Nuclear Technology*, Vol. 55, Nov. 1981.

[*13*] Druce, S. G., AERE Report R11043, AERE Harwell, Nov. 1983.

[*14*] Druce, S. G., Gage, G., Jordan, G., and Hudson, J. A., "Effect of Thermal Aging on the Mechanical Properties of PWR Pressure Vessel Steels and Weldments," United Kingdom Atomic Energy Authority Report No. AERE-R 11460, April 1985.

[*15*] Druce, S. G. and Edwards, B. C., *Proceedings*, ANS Conference on Materials Performance in Steam Generators, Florida, 1980, American Nuclear Society.

[*16*] Brear, J. M. and King, B. L., CEGB CERL Report, LM/MATS 348, Central Electricity Generating Board, 1980.

[17] Little, E. A. in *Effects of Radiation on Materials: Twelfth Conference, ASTM STP 870*, F. A. Garner and J. S. Perrin, Eds., American Society for Testing and Materials, Philadelphia, 1986, pp. 1089–1126.
[18] Outline proposal for UK contribution to IAEA CRP, submitted July 1984.
[19] Jones, R. B. and Buswell, J. T., *Proceedings*, Brighton Conference, British Nuclear Energy Society, Vol. 2, April 1984.
[20] Davies, L. M., et al, "Analysis of the Behaviour of Advanced RPV Steels under Neutron Irradiation—The UK Programme," United Kingdom Atomic Energy Agency, April 1983.
[21] Varsik, J. D. and Byrne, S. T. in *Effects of Radiation on Structural Materials (9th Conference), ASTM STP 683*, J. A. Sprague and D. Kramer, Eds., American Society for Testing and Materials, Philadelphia, 1979, pp. 252–266.
[22] Frisius, F., Kampmann, P. A., Beavan, P. A., Bunemann, D., and Wagner, R., "Small Angle Neutron Scattering Analysis of Irradiation Induced Microstructural Changes in Steel and Fe-Cu Alloys," 1981, Gesellschaft fuer Kernenergieverwertung in Schiffbau und Schiffahrt (atomic power) (Germany) (GKSS), 81/E/58.
[23] R. B. Jones, and J. T. Buswell, private communication from R. B. Jones, Sept. 1984.
[24] Highton, J. P. in *Radiation Embrittlement and Surveillance of Nuclear Reactor Pressure Vessels: An International Study, ASTM STP 819*, L. E. Steele, Ed., American Society for Testing and Materials, Philadelphia, 1983, pp. 117–129.
[25] Highton, J. P., private communication, Sept. 1984.
[26] Neale, B. K., *International Journal of Pressure Vessels & Piping*, Vol. 10, 1982, pp. 375–398.
[27] Neale, B. K., *International Journal of Pressure Vessels & Piping*, Vol. 12, 1983, pp. 207–227.
[28] Neale, B. K. and Priest, R. H., *Proceedings*, CSNI Workshop on Ductile Fracture Test Methods, Paris, 1982, pp. 464–476.
[29] Neale, B. K. and Priest, R. H., in *Elastic-Plastic Fracture Test Methods: The User's Experience, ASTM STP 856*, American Society for Testing and Materials, Philadelphia, 1983, pp. 375–393.
[30] Neale, B. K. and Priest, R. H., *Engineering Fracture Mechanics*, Vol. 19, No. 3, 1984, pp. 441–448.
[31] Neale, B. K. and Priest, R. H., *Proceedings*, International Conference on Fracture Toughness Testing, London, Paper 2, 1982.
[32] Priest, R. H., Charnock, W., and Stewart, A. T., in *Effects of Radiation on Materials—11th International Symposium, ASTM STP 782*, H. R. Brager and J. S. Perrin, Eds., American Society for Testing and Materials, Philadelphia, 1982, pp. 475–491.
[33] Priest, R. H., Charnock, W., and Neale, B. K., in *Effects of Radiation on Materials: Twelfth Conference, ASTM STP 870*, F. A. Garner and J. S. Perrin, Eds., American Society for Testing and Materials, Philadelphia, 1986, pp. 1150–1162.
[34] Fisher, S. B., Harbottle, J. E., and Aldridge, N., "Copper Precipitation in Magnox Pressure Vessel Steels: Part 1 Introduction," CEGB Report TPRD/B/0396/N84, Central Electricity Generating Board, Jan. 1984.
[35] Fisher, S. B., Harbottle, J. E., and Aldridge, N., "Copper Precipitation in Magnox Pressure Vessel Steels: Part 2: The Examination of Long Term Aged Steels," CEGB Report TPRD/B/0397/N84, Jan. 1984.
[36] Fisher, S. B., Harbottle, J. E., and Aldridge, N., "Copper Precipitation in Magnox Pressure Vessel Steels: Part 3, A Detailed Description of the Model and Its Applications," CEGB Report TPRD/B/0398/N84, Central Electricity Generating Board, Jan. 1984.
[37] Potapovs, U. and Hawthorne, J. R., *Nuclear Applications*, Vol. 6, 1969, p. 27.
[38] Brenner, S. S., Wagner, R., and Spitznagel, J. A., *Metallurgical Transactions A*, Vol. 9A, 1978, p. 1961.
[39] Goodman, S. R., Brenner, S. S., and Low, J. R., *Metallurgical Transactions*, Vol. 4, No. 10, 1973, p. 2371.
[40] Lott, R. G., Brenner, S. S., Miller, M. K., and Wolfenden, A., *Transactions*, American Nuclear Society, Vol. 38, 1981, p. 303.
[41] Hawthorne, J. R., in *Irradiation Effects on Structural Alloys for Nuclear Reactor Applications, ASTM STP 484*, American Society for Testing and Materials, Philadelphia, 1971, pp. 96–127.

[42] Fisher, S. B., Harbottle, J. E., and Aldridge, N., CEGB Report NO TPRD/B/0108/N82, Central Electricity Generating Board, 1982.
[43] Russell, K. C. and Brown, L. M., *Acta Metallurgica*, Vol. 20, 1972, p. 969.
[44] Buswell, J. T., CEGB Report TPRD/B/0351/N83, Central Electricity Generating Board, Oct. 1983.
[45] "Irradiated Nuclear PV Steel Data Base," EPRI NR 2428, Electric Power Research Institute, 1982.
[46] Odette, G. R., *Scripta Metallurgica*, Vol. 17, 1983, pp. 1183–1188.
[47] Pachur, D., *Arch. Eisenhuttenwesen*, Vol. 47, 1976, p. 501.
[48] Serpan, C. Z., *Nuclear Safety*, Vol. 22, No. 1, 1981.
[49] CEGB Evidence to the Sizewell B Public Inquiry, CEGB P12 Add 5, Central Electricity Generating Board, 1984.