The Effect of Microstructure on Irradiation Sensitivity
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Abstract
Most embrittlement trend curves do not directly account for the effects of the initial microstructure on the neutron irradiation sensitivity of reactor pressure vessel steels for light water reactors. It is intuitive, however, that as more and more hardening obstacles are introduced into the microstructure during irradiation then there must be an interaction with the pre-existing obstacles such as grain boundaries and carbides. The effect of microstructure, as reflected in initial tensile properties has been studied using data available in the open literature. The IVAR experiment included capsule VSR which contained a number of steels which had been given a range of post-weld stress relief treatments sufficient to give a reasonable variation in unirradiated strengths. Taken overall, increased unirradiated yield strengths reduced irradiation sensitivity and this was most marked in the more sensitive steels, for example with higher copper and nickel levels. The effects were considered to be independent of any differences in matrix copper contents induced by the heat treatment variations. Surveillance data have also been examined and show the same trend in both low copper and high copper steels, albeit with much greater degrees of scatter due to the heat to heat variations.

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1 Introduction

During service the Reactor Pressure Vessel (RPV) is bombarded by neutrons escaping from the core. Fast neutrons, typically with energies above 1MeV, collide with iron atoms, displacing them and causing further damage within the displacement cascade. Most of the Frenkel pairs, vacancies and self-interstitial atoms, immediately recombine but some survive and are highly mobile. Vacancies can accelerate the diffusion of solute atoms such as copper, allowing them to more rapidly cluster and precipitate whilst other defects such as dislocation loops and vacancy clusters may also form, known as matrix damage. These irradiation induced features act as barriers to dislocation movement causing an increase in strength and hardness to the detriment of ductility and toughness.

The factors affecting the magnitude of the change in properties are the irradiation exposure conditions; including the irradiation temperature, the accumulated dose and the dose rate; and the material. For the material, most Dose Damage Relationships (DDR) incorporate some compositional factors, including Cu, Ni, and P, and can also include a so-called product form variable or a different formulation to account for differences in behaviour between, for example, welds and forgings. However, the initial microstructure or mechanical properties are not explicitly included.

Many shift prediction models include product form terms. The table below shows these for ASTM E900 and the EONY model, also known as 10CFR50.61a, Reference 1. The EONY model uses different multipliers for Stable Matrix Damage (SMD) and Copper Rich Precipitation (CRP) in its product form terms. These product form terms, normalised with respect to forgings, are as follows:

Product Form	ASTM E900 CRP	EONY SMD	EONY CRP
Forgings	1.0	1.0	1.0
Plates	1.22	1.37	1.0
Welds	1.83	1.24	1.52

With respect to microstructure, grain boundaries act as sinks for diffusing species but at the nano-scale of irradiation defects the effect of grain size on irradiation damage is minimal. However, grain morphology (ferrite, bainite, martensite) does (Reference 2) although this is probably through their association with different levels of carbide precipitation. Most low alloy steels used for RPVs are bainitic, probably with different amounts of ferrite, so that DDRs have historically been fitted to this class.

The measured hardening introduced by a given population of radiation-induced defects is dependent on the hardness of the original material, and whether it contains strong obstacles to slip (the proportion of hard precipitates and inclusions to weaker obstacles such as dislocations and grain boundaries). As a result, model alloys will harden significantly more than RPV steels, and there may be differences between different classes of steel and product forms.

Some of these material effects may be reflected in the different DDRs developed in different countries for different classes of steel and different product forms.

Start of life microstructure has not been included in engineering DDRs partly because microstructural information is usually not available and partly because it is difficult to quantify microstructural variation. However, differences in microstructure result in variation in the initial unirradiated properties such as strength and hardness which are routinely measured. Improvements to DDRs may be possible, therefore, through incorporation of initial property functions.

In view of these issues it was considered pertinent to review microstructural effects and examine any relevant data in the light of current understanding. In this context, two potential sources of data have been revealed that have not previously been tested in this context. They are described below.

a) The Irradiation VARiables (IVAR) Materials Test Reactor (MTR) experiment included a capsule of materials that had received a range of stress relief treatments of sufficient magnitude to give significant differences in unirradiated strengths. The test materials covered a range of copper, nickel and phosphorus contents.

b) Surveillance data have been published that include tensile property data.

This paper summarises the examination of these data.

2 IVAR Data

2.1 Background

The IVAR MTR experiment was organised and reported by University of California at Santa Barbara (UCSB) utilising the Advanced Test Reactor at Idaho National Laboratories. A wide range of materials were irradiated at nominally three doses, three dose rates and three temperatures. All the test specimens were in the form of tensile test pieces punched from thin strips.

Four capsules, T14, T4, PB and VSR, included the CM-Series of split-melt steels. Most materials were in the as-tempered condition but capsule VSR also included CM-series samples that had been given a range of additional stress relief heat treatments after tempering. The material, test conditions and property measurements have been reported as Reference 3.

2.2 Material Compositions

The IVAR capsules reported in Reference 3 contained both model steels, the ‘CM’ series, and other model alloys. Only the CM steels were given alternative heat treatments. The compositions of the CM steels are given in the reference and Table 1. They are typical of RPV steels similar to SA508 Grade 3 forgings with around 0.85% Ni, 1.6% Mn and 0.5% Mo but the compositions have been systematically varied to demonstrate a number of irradiation damage effects.
CM3 and CM5 compare high and low P at low Cu, CM18, CM19 and CM20 show the effects of Ni at high Cu, whilst CM3, CM13, CM16, CM11, CM19 and CM12 have increasing copper contents with all other elements fixed.

2.3 Irradiation Conditions

The reported irradiation conditions for the four capsules are given below:

Capsule	Flux, $n/cm^2/s$	Fluence, n/cm^2	Temperature, °C
T4	0.97×10^{12}	0.85×10^{19}	290
T14	0.32×10^{12}	0.48×10^{19}	290
VSR	0.97×10^{12}	0.85×10^{19}	290
PB	0.7×10^{12}	1.0×10^{19}	288

2.4 Heat Treatments

CM alloys were given a base treatment consisting of an austenitisation at 900 °C for 0.5h followed by a salt bath quench at 450 °C for 10 minutes, air cooling and then tempering at 660 °C for 4h and air cooling. The normal IVAR stress relief cycle, as applied to CM alloys in capsules T4, T14 and PB was 607 °C for 4h followed by a slow air cool at 8 °C/h to 300 °C and subsequent air cooling but capsules.

For capsule VSR ('variable stress relief') the following stress relief treatments were given followed by slow air cool at 8 °C/h to 300 °C and subsequent air cooling:

Temperatures, °C	**Times, h**
590	24, 48, 96
607	12, 24, 48, 96
624	12, 24, 48
641	6, 12, 24

2.5 Variable Stress Relief Treatments

The series of three heats at ~0.42% Cu and with Ni varying from 0.02% to 1.69% are plotted in Figure 1a for the increase in yield stress against initial unirradiated yield stress. Similar results for the copper variation at 0.85%Ni are plotted in Figure 1b. Examination of these figures shows that for the higher shifts (uppermost lines) there is a marked negative gradient which decreases as shift decreases such that the lowest shifting data (CM18 in Figure 1a and CM3 in Figure 1b) show very little, if any, trend with initial unirradiated yield stress.
There are a number of other interesting observations. Firstly, it is apparent that at the higher copper levels the data start to overlap. Secondly, the gradients of the linear fits are steepest for the materials with the greatest shifts and almost flat for the lowest shifts.

A cursory consideration of these plots might lead to the conclusion that there is a microstructural effect due to the interaction of SOL strong carbide distributions (reflected in the unirradiated strengths) with the moderate strength copper precipitates. However, this is complicated by the amount of copper that may have precipitated.

The increased irradiation sensitivity with increasing copper content and with increasing nickel content is well established and captured by most DDRs. The absence of a continually increasing shift with increasing copper content – an apparent copper saturation effect - has also been observed previously. The EONY DDR, Reference 1, uses minimum and maximum copper contents of 0.072% and 0.301%, respectively. On this basis the shifts in CM11, CM19 and CM12 would be expected to be same, all other things being equal. This is broadly the case although the CM11 points tend to lie a little below the CM19 points.

The explanation of these copper limits is the variation of copper solubility with temperature. The minimum value is the solubility of copper at the reactor operating temperature (typically 290 °C in civil plants to which the EONY model was fitted) whilst the maximum value is the copper solubility limit at the final heat treatment temperature; this would normally be the stress relief temperature. For materials with greater than 0.301% Cu, during the final stress relief copper will precipitate to leave around 0.301% Cu, assuming equilibrium has been achieved, in solution in the matrix. This is then available for precipitation during the irradiation process. Precipitation of copper occurs during irradiation until the matrix copper content has fallen to 0.072%, again assuming equilibrium has been achieved. Copper precipitated at high temperatures during heat treatment is in the form of coarse (>50nm diameter) particles which contribute to the unirradiated strength in a similar manner to carbides although they are much fewer in number. The presence of the lower solubility limit is observed in practice in high dose irradiations where the copper induced hardening plateaus (ie the hardening saturates).

In the context of the present work, the upper copper limit is sensitive to the heat treatment. The amount of copper available for precipitation, which ultimately affects the degree of hardening, is dependent on the matrix copper content at the start of irradiation and is, consequently, related to the final heat treatment temperature. Higher stress relief temperatures will result in more copper in solution in the matrix which can then be precipitated to give greater hardening, dose for dose.

For the data shown in Figure 1, the apparent trends have been produced, in part, by heat treating at different temperatures. For materials with high copper contents, above the maximum copper limit, the samples stress relieved at the higher temperatures would be expected to have more copper in solution (assuming there is sufficient excess copper) which would result in greater hardening. The higher temperature stress relief treatments would also result in lower unirradiated SOL strengths. Thus the observed behaviour of increased hardening with lower unirradiated strengths may be due to the amount of copper precipitation and not a SOL microstructure effect. Similarly, if the precipitation of copper at high temperatures is not rapid then longer stress relief times will permit more precipitation during heat treatment leaving less copper in solution, reducing shift during irradiation.

The effect of copper on unirradiated yield strength and strengthening is demonstrated in Figure 2. The effect of copper on irradiation sensitivity is shown in Figure 2a where for very high copper steels the increase appears much lower than might be anticipated from the behaviour of the lower copper steels. This difference may be explained by consideration of the unirradiated strengths, Figure 2b. It may be seen that the additional copper precipitation with increasing copper contents markedly increases the unirradiated strengths. If these data were examined in isolation then the lower hardening behaviour at 0.8% Cu would appear anomalous. However, when viewed together it is clear that the lower than expected shifts in the high copper steel CM12 is because of its higher unirradiated strength and that it fits in well with the overall trend.

An objection to these observations is that heat treating at different temperatures will result in differing amounts of copper in solution so that, potentially, the irradiation sensitivity may be responding to this and not the unirradiated strength. To clarify this, the 0.85% Ni steels with more than 0.4% copper were replotted after dividing into temperature groups. The results are shown in Figure 3. In this case the initial strength sensitivity may be seen at each heat treatment temperature. In addition, the difference between the temperatures is small so that time-dependent effects are probably also small.

It should be remembered that the above observations are for steels which might be considered to have an ‘excess’ of copper so that the copper contents in solution in the matrix after stress relieving heat treatments are variable. As an alternative approach, Figure 4 plots strengthening increases against unirradiated strengths, sub-divided by temperature for the lower copper steels CM13, 0.11% Cu, and CM16, 0.22% Cu. Although there is more scatter in these steels, due in part to the generally lower irradiation sensitivity, it may be seen that, at a given stress relief temperature, there as a significant relationship between strengthening and unirradiated strength and that this is independent of stress relief temperature. In these steels it is supposed that for all heat treatment temperatures the matrix copper contents do not change since they are limited by the bulk copper content and not the copper solubility, ie the copper contents are below the solubility limit so that the heat treatments will only affect carbide distributions.

For completeness, similar plots for the low copper, medium nickel steels are shown in Figure 5. The CM5 steel, which is high phosphorus and gives the greater irradiation sensitivity, shows a similar trend with respect to strengthening and unirradiated strength. There is only a weak trend in CM3 and one of the 607 °C data points appears to be in error.

3 French and US Surveillance Data

3.1 Background

Tensile property data on unirradiated materials are not widely published. However, one such set exists for early French PWR surveillance data published in 1992, Reference 4. This gives tensile data, both before and after irradiation, on base metals, similar to SA508 Class 3, and welds. Unfortunately, the tensile data is from tests at 300 °C so that the data are not directly comparable to the ambient tests described above. However, it is generally considered that the relationship between yield strength and test temperature is similar for a wide range of materials and is only displaced to higher strengths due to irradiation. This allows comparisons of trends between ambient and elevated temperature tests.

In all, there are data for 10 French power plants with two to five reactors per plant. For each RPV there may be one or two forgings and one weld. The surveillance data for a given material have been tested in up to three capsules which have experienced different doses. Most reactors are reported to operate at 286 °C (except Chooz A but this has only high copper materials).

The US Power Reactor database has been published but the number of datasets with both unirradiated and irradiated yield strengths are small in relation to the number of Charpy shifts. Tensile data are from ambient measurements

3.2 Data Analysis

In order to combine all the French materials, the data have been divided into base metals and welds and for only low copper (<=0.08% Cu) since there are too few higher copper steels to make a sensible analysis. The irradiation sensitivity was estimated using the ratio of yield strength (measured at 300 °C) increase divided by the square root of dose. The results are plotted in Figure 6a against the unirradiated strengths. It is apparent that, despite the scatter, the gradients of the best fit lines are both negative in a similar manner to the previous results.

For the US data, these have also been normalised using the yield stress change divided by square root of dose. With so few results they have been divided into two copper groups, low copper with less than 0.1% and high copper with above 0.15%. They are plotted in Figure 6b. The US data are clearly similar to both the French data and the IVAR VSR results.

4 Discussion

The IVAR data suggest that as strength is reduced by applying additional heat treatments then irradiation sensitivity increases. This is most marked in high copper steels, which have the greatest degree of hardening, but has also been observed in CM5 at 0.02% Cu, Figure 5. It is possible that there are concurrent effects of changes in matrix copper concentrations due to heat treatment temperature effects but it has been demonstrated by both reference to medium copper level steels and high copper steels that the effect is independent of heat treatment temperature (Figure 3) so that the initial strength is more important.

The reason for this effect cannot be directly ascertained from the IVAR data but it is hypothesised that it is connected with the interaction of hardening objects, in particular, that there is an interaction between the radiation-induced hardening defects and the carbides dispersed through the microstructure. In the 'softened' steels with the additional stress relief treatments it is suggested that the hard carbide particles are fewer and further apart than in the strong steels so that the softer irradiation induced features have a greater effect.

One test for this would be to compare high and low carbon steels. Unfortunately this is not possible in the IVAR dataset because only two CM-series steels have other than ~0.15%C but these also have other compositional differences. Welds and base metals have significantly different carbon levels (nominally 0.05% and 0.2%, respectively) but these frequently have other compositional differences such as silicon and phosphorus.

Within surveillance data sets the measured variations in unirradiated properties are due to differences in both composition and heat treatment. Nevertheless, the results presented from the French and US databases show the same trends within their groupings as in the more controlled IVAR MTR data.

In general, it may be assumed that the addition of alloying elements to steel will increase their initial strength, for example through solid solution strengthening or additional second phase formation. At the same time, most additions to steel tend to increase their irradiation sensitivity. The only possible exception is molybdenum, by possibly reducing grain boundary segregation, but there is little variation within any of the data sets used in this paper. Thus, intuitively, increased strengths would be expected to worsen irradiation hardening. That the trends shown in this work have the opposite sense attests to the importance of the strength effect.

Take for example the effect of manganese. It is well accepted that increased manganese levels tend to increase irradiation sensitivity. However, manganese also increases initial yield strength which will tend to reduce sensitivity. It is clear that improved engineering shift models require both effects to be taken into account in order to produce more accurate, less uncertain models.

5 Conclusions

Analysis of data from the IVAR test irradiation has shown that variation in unirradiated strength affects the sensitivity to fast neutron induced irradiation strengthening of low alloy steels for light water reactor pressure vessels. Higher strengths reduce the amount of strengthening for given irradiation conditions. This has been attributed to the interaction of new, radiation induced hardening obstacles with the pre-existing obstacles.

The effect is most apparent in steels with greater sensitivity such as high copper, high nickel steels but has also been observed in less sensitive, low copper steels. It has been observed in both steels that have been given alternative heat treatments to change their unirradiated strengths and in two sets of surveillance data, although the scatter is much greater due to the heat to heat variation. The confounding effects of heat treatment temperature on copper solubility have been eliminated.

Improved shift models may be developed in the future if initial strength is included as an input variable.

6 References

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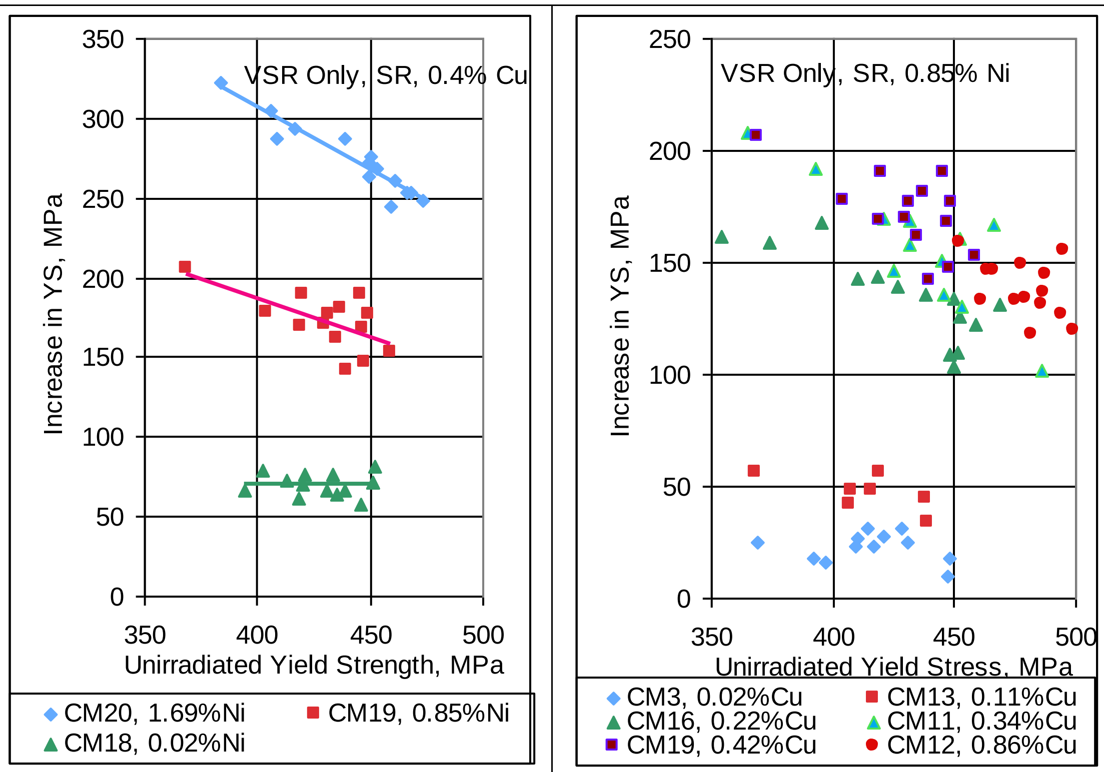
3. UCSB, "The Effects of Composition and Heat Treatment on Hardening and Embrittlement of Reactor Pressure Vessel Steels". US Nuclear Regulatory Commission Report NUREG/CR-6778, May, 2003.

4. Brillaud C and Hedin F, "In-Service Evaluation of French Pressurized Water Reactor Vessel Steel". ASTM STP 1125, 1992, pp 23-49.

Table 1   Composition of Selected IVAR VSR Capsule Steels

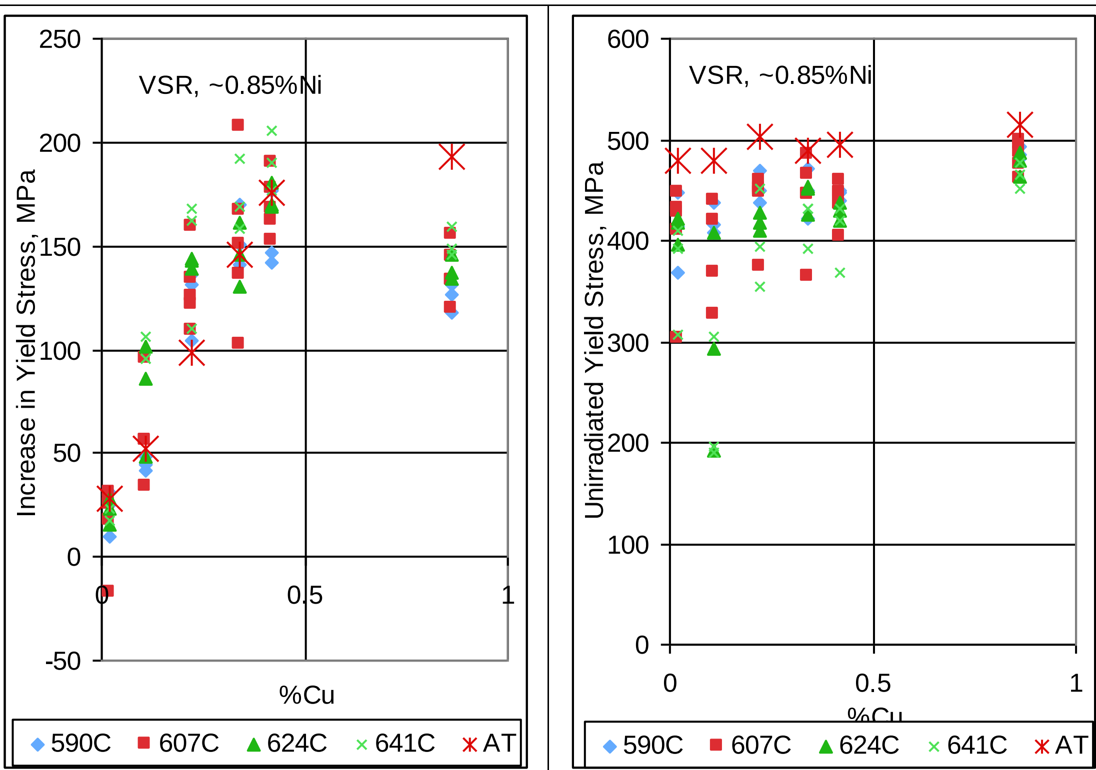
Alloy	Cu	Ni	Mn	Cr	Mo	P	C	Si	S
CM3	0.02	0.85	1.60	0.00	0.49	0.006	0.13	0.16	0.000
CM5	0.02	0.86	1.61	0.04	0.53	0.050	0.15	0.16	0.000
CM11	0.34	0.85	1.64	0.02	0.53	0.006	0.15	0.18	0.003
CM12	0.86	0.84	1.65	0.02	0.51	0.006	0.15	0.17	0.003
CM13	0.11	0.83	1.61	0.00	0.51	0.004	0.15	0.16	0.000
CM16	0.22	0.82	1.58	0.00	0.51	0.004	0.16	0.25	0.000
CM18	0.43	0.02	1.70	0.02	0.56	0.002	0.14	0.15	0.003
CM19	0.42	0.85	1.63	0.01	0.51	0.005	0.16	0.16	0.003
CM20	0.43	1.69	1.63	0.02	0.50	0.006	0.16	0.16	0.003



a) Effect of nickel in with high copper

b) Effect of copper with medium nickel

Figure 1 Effect of unirradiated yield strength on irradiation induced yield strength increases



b) Irradiated yield strength increase

b) Unirradiated yield strength

Figure 2 Effect of copper on unirradiated yield strength and increase in yield strength after irradiation in medium nickel steels (AT = As Tempered)

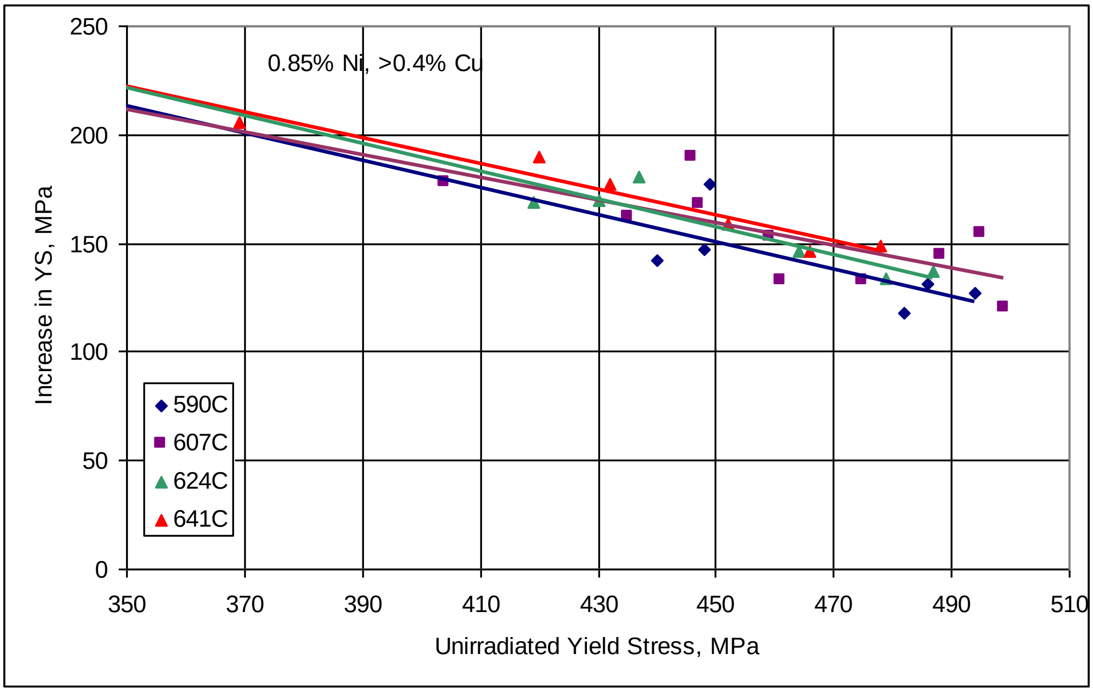


Figure 3 The effect of unirradiated strength on the irradiation sensitivity of high copper, medium nickel steels related to stress relief temperature

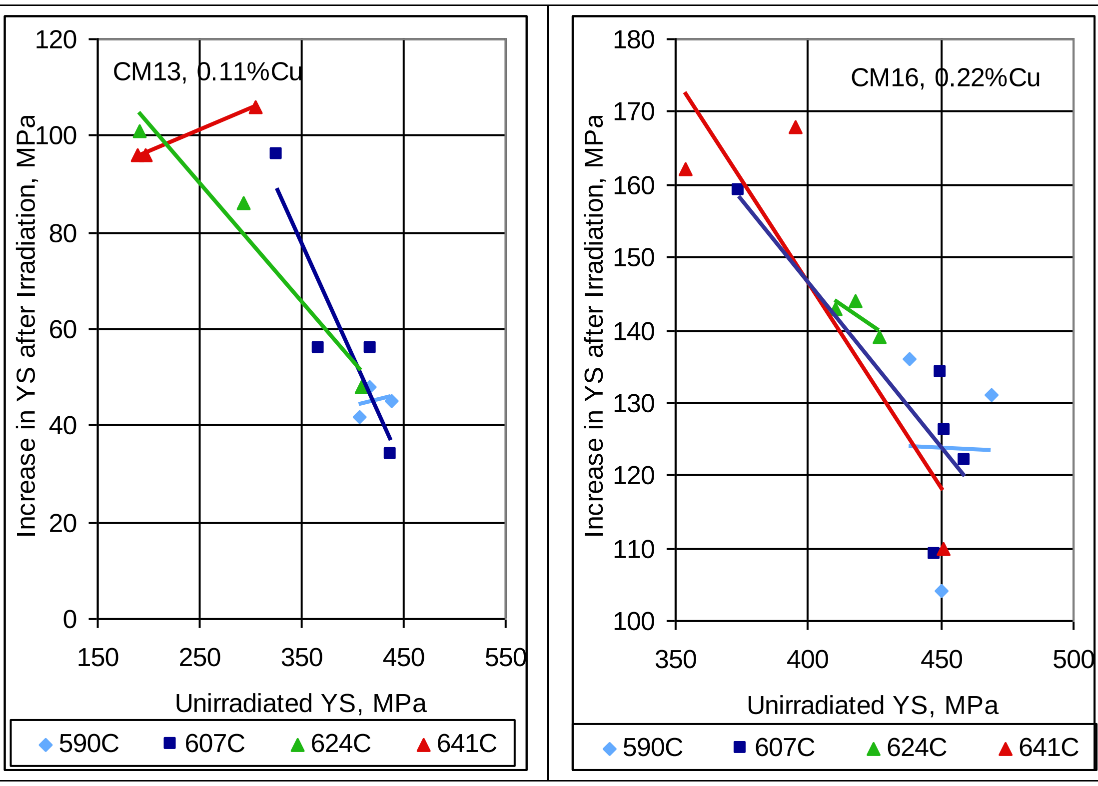


Figure 4 The effects of stress relief temperature on the irradiation sensitivity of medium copper, medium nickel steels CM13 and CM16 related to unirradiated strength

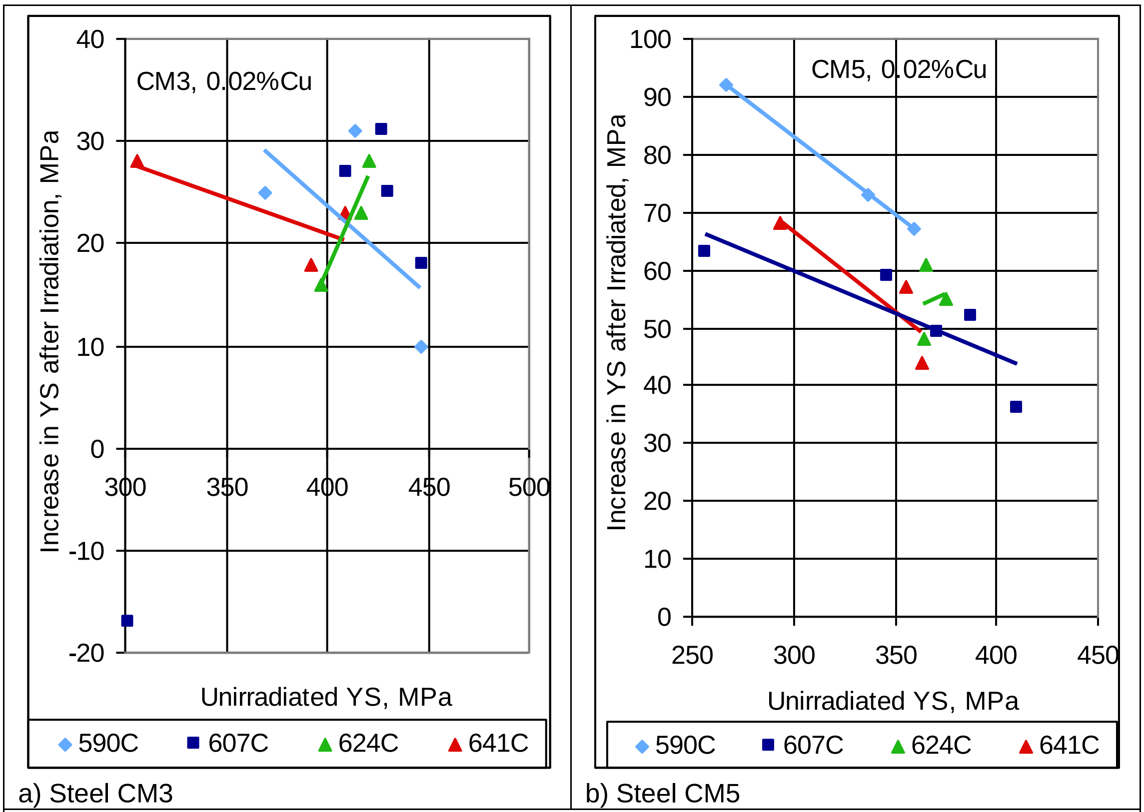


Figure 5 The effects of stress relief temperature on the irradiation sensitivity of low copper, medium nickel steels CM3 and CM5 related to unirradiated strength

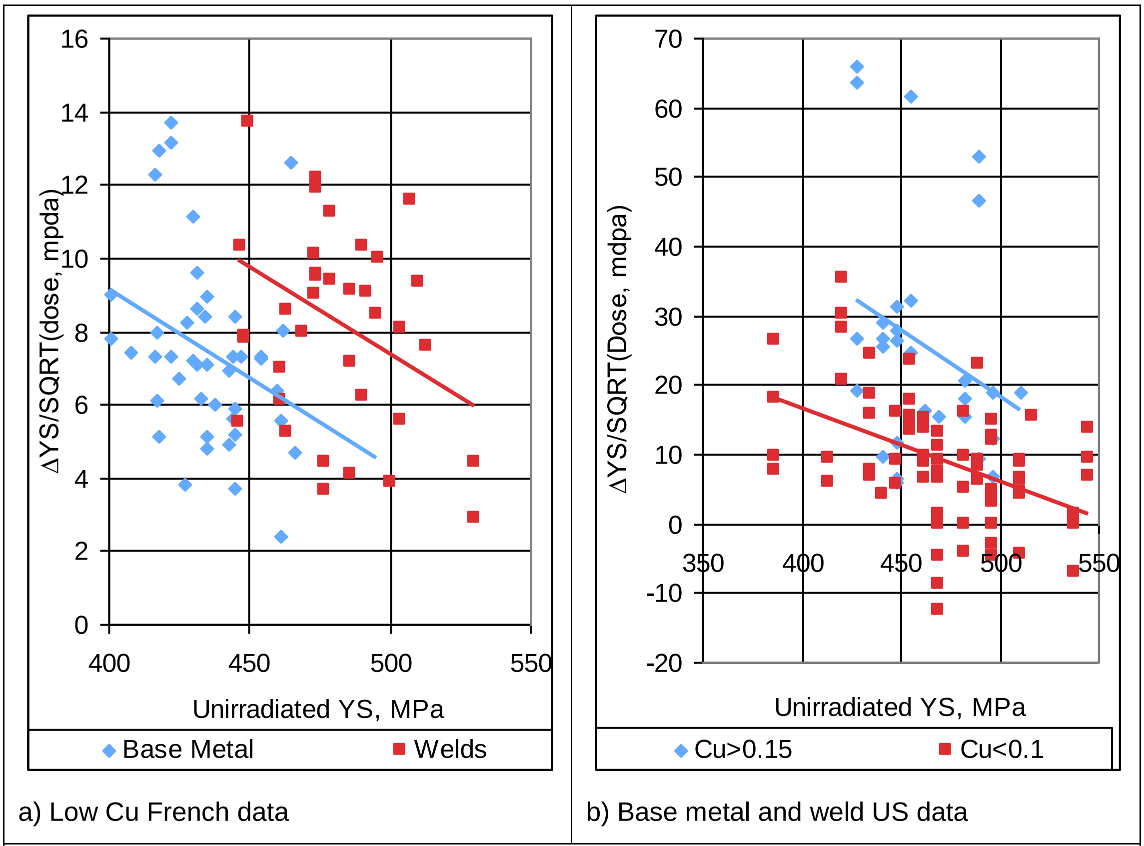


Figure 6 Normalised yield strength increases and unirradiated yield strength in French low Cu and US weld and base metal combined surveillance data