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INVESTIGATION OF RADIATION INDUCED HIGH-TEMPERATURE EMBRITTLEMENT OF FERRITIC STEELS IN THE TEMPERATURE RANGE OF THE α-γ-TRANSFORMATION

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In connection with an investigation of the mechanism of high-temperature embrittlement the neutron irradiation induced embrittlement of three Cr-steels and one Mo-steel was studied in the range of the ferrite-austenite transformation-temperature, which was different due to the chemical compositions.

The specimens were irradiated at 50 °C up to a fluence of 5×10^{19} n/cm^2 (thermal). The results of the tensile test clearly indicate that a high-temperature embrittlement of the Cr-steels only occurs at temperatures above the α-γ-transformation. The Mo-steel, which shows a tendency towards intergranular fracture in the unirradiated condition already embrittles below the α-γ-transformation. No influence of the transformation on the embrittlement could be observed. The results are discussed.

Dans le cadre des études relatives au mécanisme de l'accroissement de la fragilité à hautes températures après l'irradiation aux neutrons, trois aciers au chrome ferritiques et un acier au molybdène furent irradiés (aux neutrons thermiques) à 50 °C jusqu'à un flux de 5×10^{19} n/cm^2 dans le FR 2 à Karlsruhe et ensuite examinés à l'aide d'un essai de traction. Les températures d'essai se sont situées dans la région d'intervention de la conversion α, γ des aciers qui furent différents conformément à la composition chimique des matériaux.

Il est apparu que la fragilité des aciers au chrome n'accroît qu'à une température plus élevée que la température de conversion α, γ et au moment où une partie de la matrice s'est transformée du ferrite en austénite. Par contre, l'acier au molybdène—qui est fortement susceptible d'une rupture intercristalline même en absence d'irradiation—fragilise déjà à l'état ferritique. Une influence de la température de conversion sur le comportement à la rupture ne peut être observée dans ce cas. Les résultats sont discutés.

Im Rahmen der Untersuchungen über den Mechanismus der Hochtemperaturversprödung nach Neutronenbestrahlung wurden 3 ferritische Chromstähle und ein Molybdänstahl im FR 2 in Karlsruhe bei 50 °C bis zu 5×10^{19} n/cm^2 (thermisch) bestrahlt und im Zugversuch geprüft. Die Versuchstemperaturen lagen im Bereich der α-γ-Umwandlung der Stähle, die entsprechend der chemischen Zusammensetzungen der Werkstoffe unterschiedlich waren.

Es zeigte sich, dass eine Versprödung bei den Chromstählen erst beobachtet wurde, wenn die α-γ-Umwandlungstemperatur überschritten und ein Teil der Matrix vom Ferrit zum Austenit umgewandelt ist. Im Gegensatz dazu versprödet der Molybdänstahl, der auch im unbestrahlten Zustand bereits eine starke Neigung zum interkristallinen Bruch besitzt, bereits im ferritischen Zustand. Ein Einfluss der Umwandlungstemperatur auf das Bruchverhalten ist hier nicht zu erkennen. Die Ergebnisse werden diskutiert.

1. Introduction

It is well known that the high-temperature embrittlement after neutron irradiation due to (n, α)-reactions can mainly be observed on nickel-base-alloys and austenitic stainless steels. These materials are fcc-metals and show an intergranular fracture in the unirradiated condition at elevated temperatures. Therefore it can be supposed that fcc-metals have a stronger tendency to intergranular fracture than bcc-alloys and that thereby the occurrence of high-temperature embrittlement of fcc-metals is especially favoured.

This assumption is supported by a number of published investigations on bcc-alloys with comparable boron contents, where no high-temperature embrittlement after neutron irradiation could be observed. Stress rupture and tensile-tests on iron-chromium-alluminium-yttrium-alloys show, that the ductility at temperatures above 600 °C is not affected by irradiation [1]). Irradiation-tests on a ferritic steel "type 410 SS" did also not reveal any influence of neutron irradiation on the elongation at elevated temperatures [2]). Tensile tests on vanadium-base-alloys, which in the unirradiated condition have a transgranular fracture even at 950 °C, indicate also no high-temperature embrittlement [3]).

The connection between the structure of the matrix and the occurrence of high-temperature embrittlement is most distinctly demonstrated by an investigation on a 12%-Cr-steel. The results show that the high-temperature embrittlement can only be observed if a α-γ-transformation has occurred [4]). Following this investigation the irradiation-behaviour of several ferritic steels was tested in the range of the α-γ-transformation.

2. Experimental

Steels with increasing Cr-contents between 11 and 17% were selected to study the influence of varying amount of transformed matrix and of different α-γ-transformation temperature on the irradiation behaviour. Furthermore the influence of heat-treatments, leading to different ferritic structures was investigated on one of the steels.

The chemical composition of the steels as well as the temperatures, where the α-γ-transformation starts, are given in table 1. Dilatometric measurements on the three Cr-steels reveal that the degree of transformation decreases with increasing Cr-content. A complete transformation is observed for example at the 11%-Cr-steel, while the 14 and 17%-Cr-steel change only 30 respectively 5% of their matrix into austenitic structure.

Specimens of the 11%-Cr-steel were irradiated in the following three heat-treatments:

a. Annealed at 1080 °C, quenched in oil and than annealed at 600 °C;
b. Annealed at 1080 °C, quenched in water;
c. Annealed at 1080 °C, quenched in water and than annealed during 20 h at 700 °C.

The other steels were tested as received.

Tensile-specimens with a diameter of 3 mm were irradiated at about 50 °C in the FR 2 at Karlsruhe up to a fluence of 5×10^{19} n/cm^2 (thermal). Afterwards the mechanical properties were determined in a tensile test at different temperature with a strain-rate of 0.01/min. Experimental results indicate, that the strength of all tested materials at temperatures around the α-γ-transformation was not affected by irradiation.

In the following the influence of the irradiation on the ferritic and austenitic structure will be demonstrated by the values of total elongation and by some metallographic pictures of the specimens. The influence of irradiation on the elongation of the 11%-Cr-steel in three heat-treated conditions between 650 and 950 °C is shown in fig. 1.

No high-temperature embrittlement is to be seen when the specimens are in the ferritic condition. The embrittlement starts with the

TABLE 1

	Temperature of α-γ-transformation (°C)	C	Cr	Ni (wt %)	Mo	V	Nb	B (ppm)
X18CrMoVNb12I	825	0.11	11.3	0.7	0.5	0.3	0.25	70
X7Cr14	850	0.08	14,6	–	–	–	–	1
X8Cr17	900	0.05	17.7	–	–	–	–	1
15Mo3	730	0.13	–	–	0.24	–	–	2

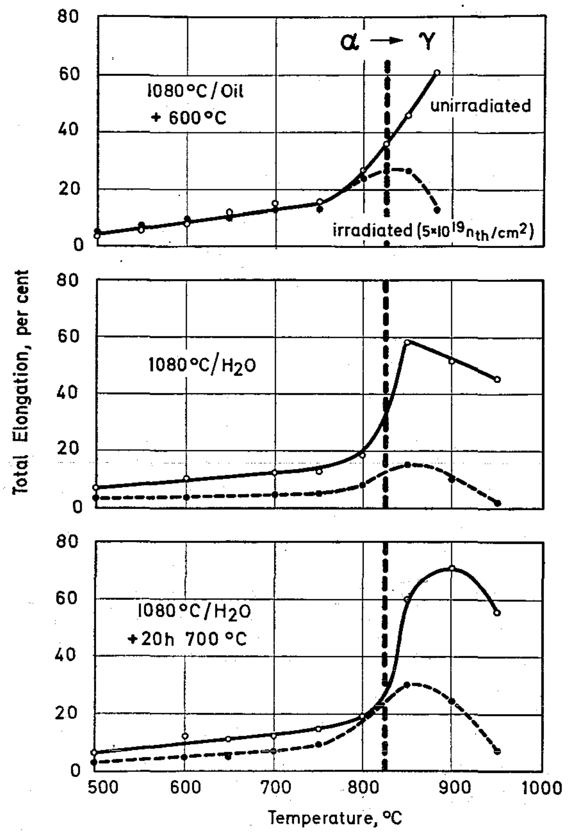


Fig. 1. Pre- and post-irradiation tensile ductility of X18CrMoVNb12 1-steel in different heat-treated conditions.

beginning of the α-γ-transformation. The specimens, which are in condition (a) do not show high-temperature embrittlement below the α-γ-transformation.

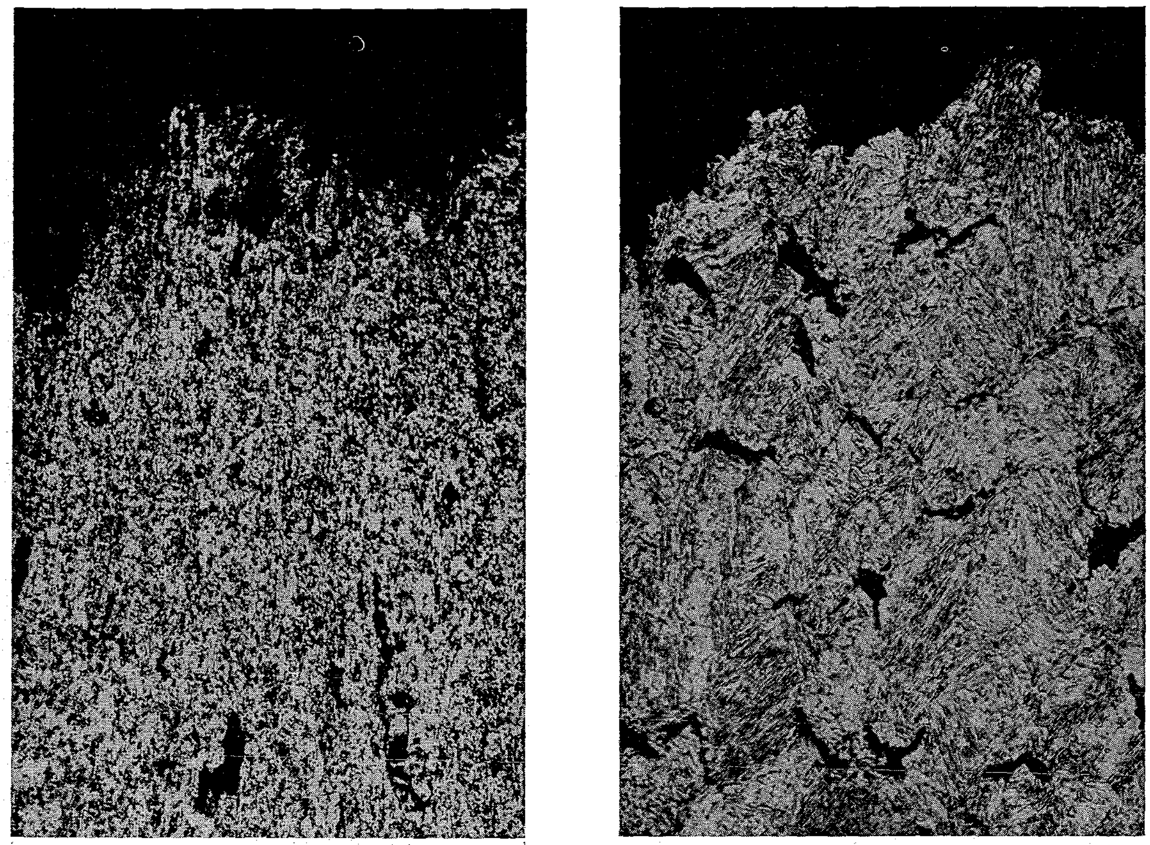
The two other diagrams (condition b and c) demonstrate a similar behaviour. The remarkable difference is a slight influence of neutron irradiation in the α-range already. Here again serious embrittlement starts just above the temperature of α-γ-transformation. To check the obvious assumption, that the favourable irradiation behaviour below the transformation temperatures may be the result of carbide particles existing in the ferritic structure, the following testing-methods were chosen: The irradiated and unirradiated specimens were annealed in the furnace of the tensile-testing-machine at a temperature beyond the α-γ-transformation. Then, after cooling slightly below the transformation-temperature, they were tested. The result was an increase of the total elongation, but no high-temperature embrittlement could be observed. Therefore one can conclude that the favourable irradiation behaviour in the ferritic condition is not an effect of carbide particles.

A good demonstration of the different irradiation behaviour of ferrite and austenite is given by the metallographic picture of the fractured specimens (fig. 2). These irradiated specimens were tested at 800 and 850 °C, that means in the α- respectively in the γ-range. The specimens tested at 800 °C distinctly show a transgranular fracture. The structure does not reveal any intergranular cracks. A completely different fracture behaviour can be observed on the sample tested at 850 °C. Here the fracture is fully intergranular and shows a lot of intergranular cracks, which obviously lie at the original austenitic grain boundaries.

As already mentioned the 11%-Cr-steel shows a complete α-γ-transformation, whereas the 14%-Cr-steel has only a partial transformation. The elongation of this steel in the pre- and post-irradiated condition can be seen in fig. 3. The transformation starts at 850 °C. The results clearly indicate that here again no embrittlement can be seen in the ferritic condition, and the results already found with the 11%-Cr-steel are confirmed. The obviously small amount of embrittlement, which still allows an elongation of nearly 30%, may be seen in connection with the smaller amount of transformation in comparison with the 11%-Cr-steel.

The metallographic investigation shows the influence of the transformation on the fracture behaviour of irradiated samples very clearly. Fig. 4 shows the fractured sections of specimens tested at 850 and 900 °C. Besides the considerably smaller reduction in area at 900 °C, the great amount of intergranular cracks indicates the change in fracture behaviour beyond the transformation temperature.

The third examined steel was a 17%-Cr-steel. Its chemical composition leads to a rather stable



(a) (b)

Fig. 2. Fracture sections of irradiated tensile specimens (X18CrMoVNb12 1-steel). Test temperature 800 °C (a); 850 °C (b). ×400

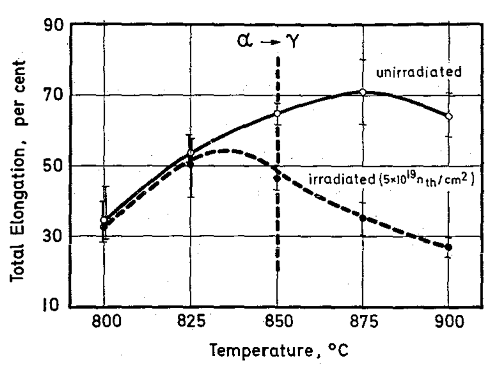
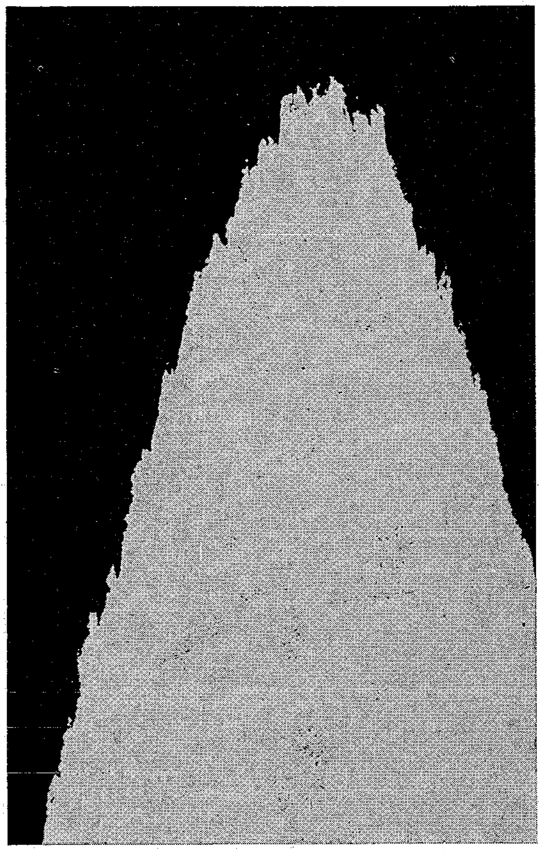


Fig. 3. Pre- and post-irradiation tensile ductility of X7Cr14-steel.

ferritic structure even at higher temperatures. The dilatometric curve indicates only a very slight amount of transformation at 900 °C.

Fig. 5 shows the influence of the irradiation on the ductility of this steel. As can be expected from the results mentioned before, there is no embrittlement up to a temperature of 900 °C. The embrittlement starts beyond the transformation temperature. The strong scattering in the range of 750 °C may be due to different amounts of carbide precipitations, which are caused by heating during tensile testing. The ferritic steels investigated up to now were chosen under the point of view, that they have a strong tendency to transgranular fracture in the ferritic condition.

We think it would be an unallowed interpretation of the results if one would conclude that high-temperature embrittlement cannot occur at all in ferritic steels. But one can conclude that the tendency to intergranular fracture is not so strong in the ferritic condition.



(a) (b)

Fig. 4. Fracture sections of irradiated tensile specimens (X7Cr14-steel). Test temperature 850 °C (a); 900 °C (b). ×30

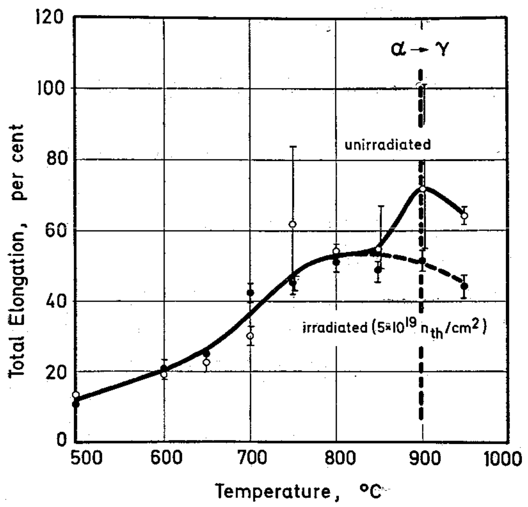


Fig. 5. Pre- and post-irradiation tensile ductility of X8Cr17-steel.

The reason for this phenomenon can certainly be found in the fracture mechanism of the unirradiated materials. As a prove for this consideration a ferritic steel was irradiated, which shows a tendency to intergranular fracture in the unirradiated condition. The pre- and post-irradiation ductility of this steel can be seen in fig. 6. It is clearly demonstrated that this steel shows a strong embrittlement even in the ferritic condition. The α-γ-transformation has no influence on the high-temperature embrittlement.

3. Discussion

The results clearly indicate that the α-γ-transformation in several ferritic steels is

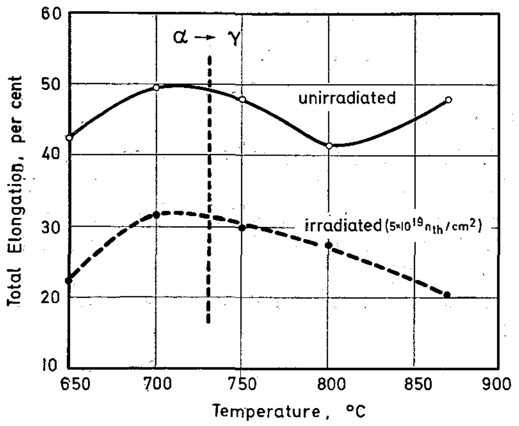


Fig. 6. Pre- and post-irradiation tensile ductility of 15Mo3-steel.

connected with a change in irradiation behaviour. No influence of the transformation is found when the steel shows a tendency to intergranular fracture in the ferritic condition. So it may not be concluded from these results, that the bcc-alloys do not show high-temperature embrittlement at all. The irradiation- and fracture-behaviour indicate only a minor tendency of most ferritic steels to high-temperature embrittlement in the α-condition. At first it must be taken into account whether the He-distribution, due to more homogenous B-distribution or a fixing of He-bubbles by incoherent carbide-particles within the grain, can be different in the α- and γ-condition and can lead to a different irradiation behaviour. The results of the tests on the 11%-Cr-steels in various heat-treated conditions show that this cannot be the reason for the different behaviour. So the interpretation of these results must start with considering the factors which are governing the appearance of intergranular fracture. Based upon the theories of intergranular fracture there are at least the following factors which seem to be very important:

Besides the effective surface energy, there are the mobility of the grain boundaries and the recovery which both may influence the stress-concentration at grain boundaries.

Often it is assumed that the appearance of intergranular cracks depends on the reduction of surface energy due to a segregation of impurety atoms at the grain boundary. The effective surface energy for intergranular fracture is approximately $\gamma = (2\gamma_S - \gamma_B) + a\sigma^2$, where γ_S is the surface energy, γ_B the grain boundary energy and $a\sigma^2$ a term representing the plastic work done at the crack tip. The segregation of solute atoms at grain boundaries normally lowers the grain boundary energy γ_B as well as the much larger term $2\gamma_S$ for the surface energy. This is the reason for the preferred intergranular fracture due to grain boundary segregations.

If the change in fracture behaviour, when crossing the α-γ-transformation temperature is due to differences in surface energy, this energy must be lower for the fcc-structure. Assuming that the structure of the grain boundaries is not different in bcc and fcc-metals a difference in surface energy could arise from a different amount of segregation of interstitial impurities. Because of the much higher distortion energy for the solution of carbon, nitrogen, and oxygen in α-Fe than in γ-Fe, the driving force for grain boundary segregation of these elements is smaller in γ- than in α-Fe. Therefore it is not probable that the change in fracture characteristics is due to a higher decrease in surface energy in the γ-phase due to grain-boundary segregation.

So it is more probable that those factors are responsible for the influence of structure, which influence the stress concentration at grain boundaries. It is well known that every process which will lead to a stress relaxation at the grain boundaries will delay or prevent intergranular fracture. So the larger grain boundary mobility and/or the higher recovery rate due to the higher rate of self-diffusion in α-Fe may be one of the reasons for the lower tendency to intergranular fracture. On the other hand the higher diffusion rate in the bcc structure of iron excludes that the difference in fracture behaviour may result from a faster growth of the voids in the fcc structure.

Last but not least one has to consider whether the dislocation reactions in bcc metals may perhaps favour the nucleation of transgranular cracks.

In all considerations it must be taken into account that certain ferritic steels do show intergranular fracture and some austenitic steels have no tendency towards intergranular fracture. So the main result is that in several ferritic steels the change in irradiation behaviour occurs when the α-γ-transformation starts. This change in irradiation behaviour must be connected with an influence of structure on the fracture characteristics.

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