EMBRITTLEMENT SATURATION OF REACTOR PRESSURE VESSEL STEELS

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NRC Seventh Water Reactor Safety
Research Information Meeting
November 5-9, 1979
National Bureau of Standards (NBS)
Gaithersburg, Maryland

Nuclear Research Centre Jülich, F.R.G.

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INTRODUCTION

Recently irradiation results of a surveillance program were discussed about the saturation of irradiation embrittlement at neutron fluences as low as 10^{19} n/cm^2. To explain this saturation phenomenon it was surmized that at long-term irradiations other annealing processes are occuring than at short term irradiation with a high neutron flux. This would mean that the irradiation embrittlement was dependent on the neutron flux. If that is true, this would have significant consequences for irradiation programs. One could no longer, without reservations, perform irradiations in power and research reactors, unless one is familiar with the transferability of these results to the results obtained in the wall of a reactor pressure vessel.

A saturation of irradiation embrittlement, however, has also been noted for short-term irradiations and this in turn leads to the assumption that it is not the duration of irradiation that is significant for the occurance of saturation. This assumption is supported by the results of annealing tests with short and long-term irradiated specimens which were subjected to equal neutron fluences. Short and long-term irradiated specimens showed the same annealing responses.

IRRADIATION RESULTS

Subsequently, the irradiation and annealing results of three different Reactor pressure vessel steels will be reported. The steels were irradiated at different irradiation temperatures, with different neutron fluxes and up to different neutron fluences (E > 1 MeV) in the research reactors FRJ-1 and FRJ-2 in Jülich. In table 1 the chemical analyses of the steels are shown. First steel A, a basic material according to ASTM A 533 B, produced by Lukens as HSST 03, which has been thoroughly investigated in the IAEA-coordinated irradiation program. Secondly steel B, a modified A 533 B having 0.15% vanadium and steel C with a somewhat higher nickel content. The latter two were testing materials in the German VDEh-steel irradiation program [1,2]

In addition, the results of two forgings produced by the Klöckner Company according to A 302 B have been presented. Here too one of the two forgings was slightly modified with Vanadium. In order to obtain a general understanding of the irradiation embrittlement, the following figures show, as well as the trend curve for the increase in transition temperature, the trend curve for the relative increase of Vickers hardness, the relative decrease of the upper shelf energy and the relative increase of yield strength.

Figure 1 shows the increase of transition temperature as a function of neutron fluence (E > 1 MeV) up to fluences of $6 \times 10^{20} n/cm^2$. Each irradiation temperature is represented by a different colour, blue is for 150 °C, green for 280-310 °C and red for 400 °C. The green trend curves show expected embrittlement for the running temperature of power reactors. These will therefore be examined first. The increase of transition temperature is considerably high at a neutron fluence of $10^{20} n/cm^2$; 100 °C for the less sensitive steel A and 200 °C for the more sensitive steel B. For higher neutron fluences the increase of transition temperature tends towards saturation. This is for steel B, curve 1 and for steel A, curve 4. The points for steel A whose fluences are greater than $1.5 \times 10^{20} n/cm^2$, are from a publication by Russ. Hawthorne, NRL [3]. The measurements were for steel of ASTM specification A 533 B, but not from HSST-plate 03.

The trend curve for steel C in the $5 \times 10^{19} n/cm^2$ range is especially interesting. At a neutron fluence of $10^{19} n/cm^2$ an apparent saturation effect occurs. The saturation remains to more than $5 \times 10^{19} n/cm^2$, but at $10^{20} n/cm$ a considerably higher increase of transition temperature takes place. This behaviour was seen for other steels irradiated at 300 °C. Similar apparent saturation can be seen from the results of steel A for the 150 °C irradiations (blue) and the 400 °C irradiations (red). The neutron fluence range between 10^{19} and $5 \times 10^{19} n/cm^2$ is important because it corresponds to the end of life neutron fluence of power reactors.

For this reason the fluence range is shown in greater detail in Fig. 2. The 300 °C irradiation results have already been discussed, the differences between the different steels irradiated at 300 °C are clear. However when one considers the 150 °C and 400 °C results, it can be seen that the two steels A and B are equally sensitive to irradiation. They already show a saturation behaviour which lasts up to 5 x 10^{19}n/cm^2 at 150 °C-irradiation and up to 10^{20}n/cm^2 at 400 °C irradiation.

The numerous fluence values at an irradiation of 150 °C were chosen in order to investigate the embrittlement growth by annealing after irradiation. The temperature of 150 °C guarantees that no noticeable annealing of the irradiation damage occurs during the irradiation.

Normally irradiation was carried out at a neutron flux of 3 x 10^{13}n/(cm^2·s). In order to investigate the effect of flux, steels A and B were also irradiated at a flux of 6 x 10^{11}n/cm^2/s at 150 °C for fluences up to 1.5 x 10^{19}n/cm^2. The same embrittlement values were obtained as with a neutron flux of 3 x 10^{13}n/cm^2 per second.

Also Fig. 2 contains additionally the results for low flux irradiations of A 302 B material at 300 °C. The A 302 B specimens were irradiated in the VAK power reactor to 1.2 x 10^{20}n/cm^2. With these specimens the duration of irradiation was ten years with a neutron flux of about 6 x 10^{11}n/cm^2 per second. It was a lucky chance that we could test this steel with such a long irradiation period, if one can call it a lucky chance when a capsule gets stuck. For the A 302 B steel, the lower point indicates saturation. This point corresponds to the saturation value of steel C. The A 302 B material with 0.12 Vanadium had nearly the same embrittlement as steel A irradiated with 3 x 10^{13}n/cm^2/s at 300 °C.

The trend curves for the relative increase of Vickers Hardness of the three steels at the three irradiation temperatures are shown in Figure 3. The results of the 300 °C irradiations show the same behaviour as the increase of transition temperature, except steel B which was somewhat more sensitive. The 150 °C and 400 °C irradiations show that here steel B is much more irradiation sensitive than steel A. This is in contrast to the ΔTT-behaviour where at the 150 °C and 400 °C irradiations steel A and steel B were equally sensitive. At 400 °C steels A and C indicate a saturation in the range between 5×10^{19} and $10^{20} n/cm^2$.

The behaviour of another mechanical property, the relative decrease of upper shelf energy is in Figure 4. In this case the behaviour of steels A and B at 300 °C is different from the other mechanical properties. A measurable change first occurs at $5 \times 10^{19} n/cm^2$. In contrast to the previous cases in this graph steel B is as sensitive as steel A. Steel C shows no great change in behaviour. Up to neutron fluences of $5 \times 10^{19} n/cm^2$ the relative decrease of upper shelf energy is small but for $10^{20} n/cm^2$ increases by a factor of 5. The 150 °C irradiations show two saturations. One between 10^{19} and $5 \times 10^{19} n/cm^2$ and one between 5×10^{19} and $10^{20} n/cm^2$, for the 400 °C irradiations the upper shelf energy was not changed.

The last mechanical property is the relative increase of yield strength, which is shown in Figure 5. The behaviour of the 300 °C irradiation is the same as for the increase of transition temperature and relative increase of Vickers Hardness. The 150 °C irradiations produce a strong increase in yield strength of 70 % for $1.5 \times 10^{19} n/cm^2$. No change occurs for steel B at higher fluences, but steel A increases 20 % at $7 \times 10^{19} n/cm^2$. The 400 °C irradiations indicated no change and were therefore in agreement with the behaviour of the upper shelf energy.

After having discussed the trend curves for various mechanical properties it can be summarized that

1. apparent saturation effects often occur in limited neutron ranges
2. the trend curves of the various mechanical properties are different. This leads to the assumption that different damage mechanisms are responsible. It is noteworthy that the Vickers Hardness showed the least similarity to the others.

3. the irradiation temperature influences the embrittlement greatly. For example, at 5 x $10^{19} n/cm^2$ the relative increase of yield strength is 70 % at 150 °C, 20 % at 300 °C and 0 % at 400 °C.

ANNEALING RESULTS

The strong decrease of embrittlement with temperature indicates that the higher the irradiation temperature the fewer the defect mechanisms which remain, or in other words, at higher temperatures some defects anneal out. Heat treatment after irradiation has the same effect as high temperature irradiation. The embrittlement starts to anneal at about 220 °C and at 500 °C after 4 hours is nearly the same as normal unirradiated pressure vessel steel [4, 5].

To explain the apparent saturation, the annealing of irradiation damage and the processes noted must be briefly refered to. During the annealing tests the Vickers Hardness was measured in terms of the hardness increase produced by irradiation relative to the initial hardness.

The next illustration Figure 6 shows such a annealing process for steel A. It shows the relative hardness increase produced by irradiation dependence on the annealing temperature. A further parameter is the annealing time. The curve indicates the behaviour for an annealing duration of four hours. Shorter times shift the curve to the right and longer times shift it to the left. During the annealing, various processes occur in different temperatures ranges. These are characterized by various activation energies. The individual processes were determined by the different time dependence at various temperatures [6].

A given state of annealing is always the sum of the processes which occur at a given temperature. By marking the processes with different colours, it can be seen which process is actually annealing. One can also estimate the influence of the irradiation temperature. At an irradiation temperature of 300 °C process number one has no influence owing to the high mobility of the corresponding defects. At the irradiation temperature of

400 °C only process 4 is important and this causes further lowering of the embrittlement.

The manner in which the individual process build up can be seen in Figure 7. Irradiations at 150 °C for all neutron fluence values were investigated. Again the temperature dependence of relative hardness increase after annealing is shown. The curve sections 1, 2, 3, 4 are related once more to the current annealing processes. This indicates quite clearly what is happening in the fluence range from 1.5×10^{19} to $7 \times 10^{19} n/cm^2$. The yellow part, i.e., process number 3 is decreasing while the red part i.e., process number 4 is increasing correspondingly. Thus there occurs, if you wish, an increment of process number 3 towards a higher activation energy, i.e. the defects are probably larger. The effect of higher neutron fluence is the conversion of type 3 defects into type 4 defects, with the overall defect density being almost unaltered. A quasi saturation has been reached. Furthermore, process number 2 accounts for 10 % of hardness increase for neutron fluences up to $3 \times 10^{19} n/cm$. However, for $7 \times 10^{19} n/cm^2$ the contribution increases to 20 %.

The conversion of type 3 defects into type 4 defects at the irradiation temperature of 300 °C must be the reason for annealing behaviour of the A 302-steel irradiated at low flux in the VAK-Reactor (Fig. 7). The development of process number 3 for the vanadium containing charge proves this because the specimens were inside the same capsule; only process number 4 occurs for the lower embrittlement A 302 B steel.

As well as with hardness testing, annealing characteristics were investigated by tensile testing and Charpy-V-testing. The processes seen were confirmed by these methods. It was established for steel A that the relative decrease of upper shelf depends mainly on process number 2, the relative increase of yield strength depends mainly on processes 1, 2, 3 and the increase in transition temperature depends on processes number 2, 3, 4 of the hardness annealing. For steel B an additional process, number 5, was found, which only affected the Vickers Hardness.

These results explain the observed behaviour of the increase of transition temperature curve, increaseof Vickers Hardness curve, decrease of upper shelf energy curve and increase of yield strength curve.

The occurance of a process depends on the irradiation temperature. For example at an irradiation temperature of 350 °C, the contribution of process number 2 to the embrittlement is negligible and furthermore, no decrease in upper shelf energy is expected.

SUMMARY

Summarizing the following conclusions can be made:
A saturation of embrittlement was often found within certain neutron fluence ranges. It occurs for irradiations at a high neutron flux of $3 \times 10^{13} n/(cm^2 s)$ as well as with a low neutron flux of $6 \times 10^{11} n/(cm^2.s)$, and also at 150 °C irradiations as well as for higher temperature irradiations. With the available data it cannot be said under which prerequisites a saturation occurs more readily.

Two causes for the apparent saturation were discovered from the behaviour of the annealing curves. Firstly, the production of defects of higher activation energy without altering the effective defect density and secondly the tendency of one process to remain constant in spite of increasing neutron fluence. It is clear that the demonstrated processes are the effects of microscopic mechanisms and that the irradiation behaviour can be described by the growth of these processes but not understood. This requires microscopic investigations. Such investigations have already been started by using the neutron small angle scattering technique and simultaneously annealing and measuring the Vickers Hardness /7/. It is intended to use electron microscopy to better advantage in the future.

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/3/ Hawthorne,J.R., "Further Observations on A 533-B Steel Plate , Tailored for Improved Radiation Embrittlement Resistance", Journal of Pressure Vessel Technology, Mai 1976, Vol. 98

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STEEL A	Manganese - Nickel - Molybdenum, according to ASTM A 533-B, HSST - plate 03			
	0.22 C, 1.4 Mn, 0.012 P, 0.008 S, 0.3 Si, 0.6 Ni, 0.12 Cr, 0.5 Mo, 0.12 Cu, 0.01 V			
	Transition Temp. (51 J/cm^2)	Vickers Hardness	Upper Shelf Energy	Yield Strength
	25 ^0C	2000 N/mm^2	160 J/cm^2	510 N/mm^2
STEEL B	Manganese - Nickel - Molybdenum - Vanadium			
	0.11 C, 1.3 Mn, 0.012 P, 0.008 S, 0.3 Si, 1.5 Ni, 0.11 Cr, 0.4 Mo, 0.11 Cu, 0.15 V			
	Transition Temp. (51 J/cm^2)	Vickers Hardness	Upper Shelf Energy	Yield Strength
	- 14 ^0C	1860 N/mm^2	210 J/cm^2	500 N/mm^2
STEEL C	Nickel - Chromium - Molybdenum			
	0.15 C, 0.4 Mn, 0.010 P, 0.020 S, 0.2 Si, 3.2 Ni, 0.34 Cr, 0.4 Mo, 0.15 Cu, 0.06 V			
	Transition Temp. (51 J/cm^2)	Vickers Hardness	Upper Shelf Energy	Yield Strength
	- 65 ^0C	2400 N/mm^2	190 J/cm^2	620 N/mm^2

Table 1 : Chemical Composition in wt % and Material Data

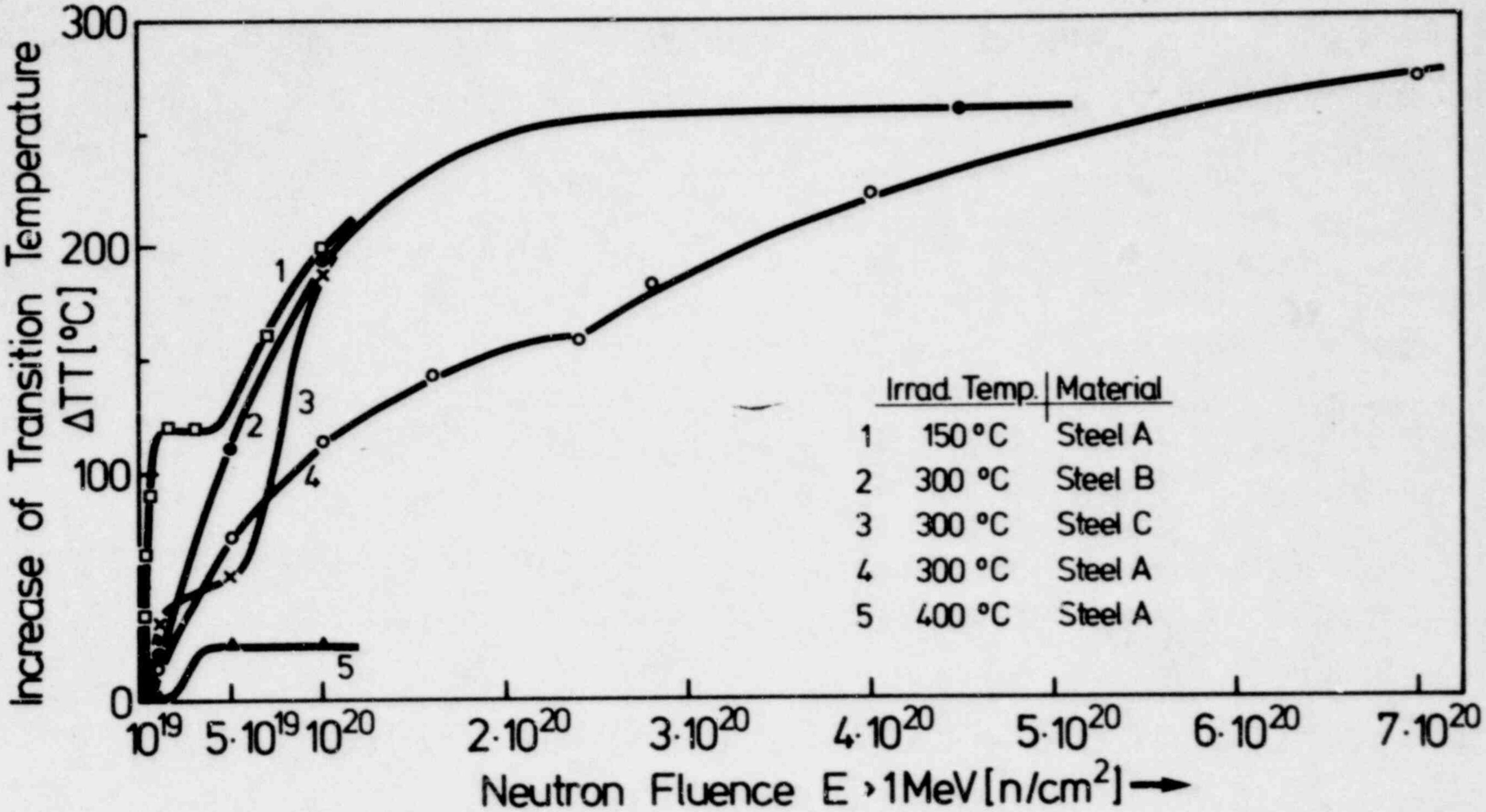

Fig.1 : Effect of High Neutron-Fluence (E > 1 MeV) on Increase of Transition Temperature

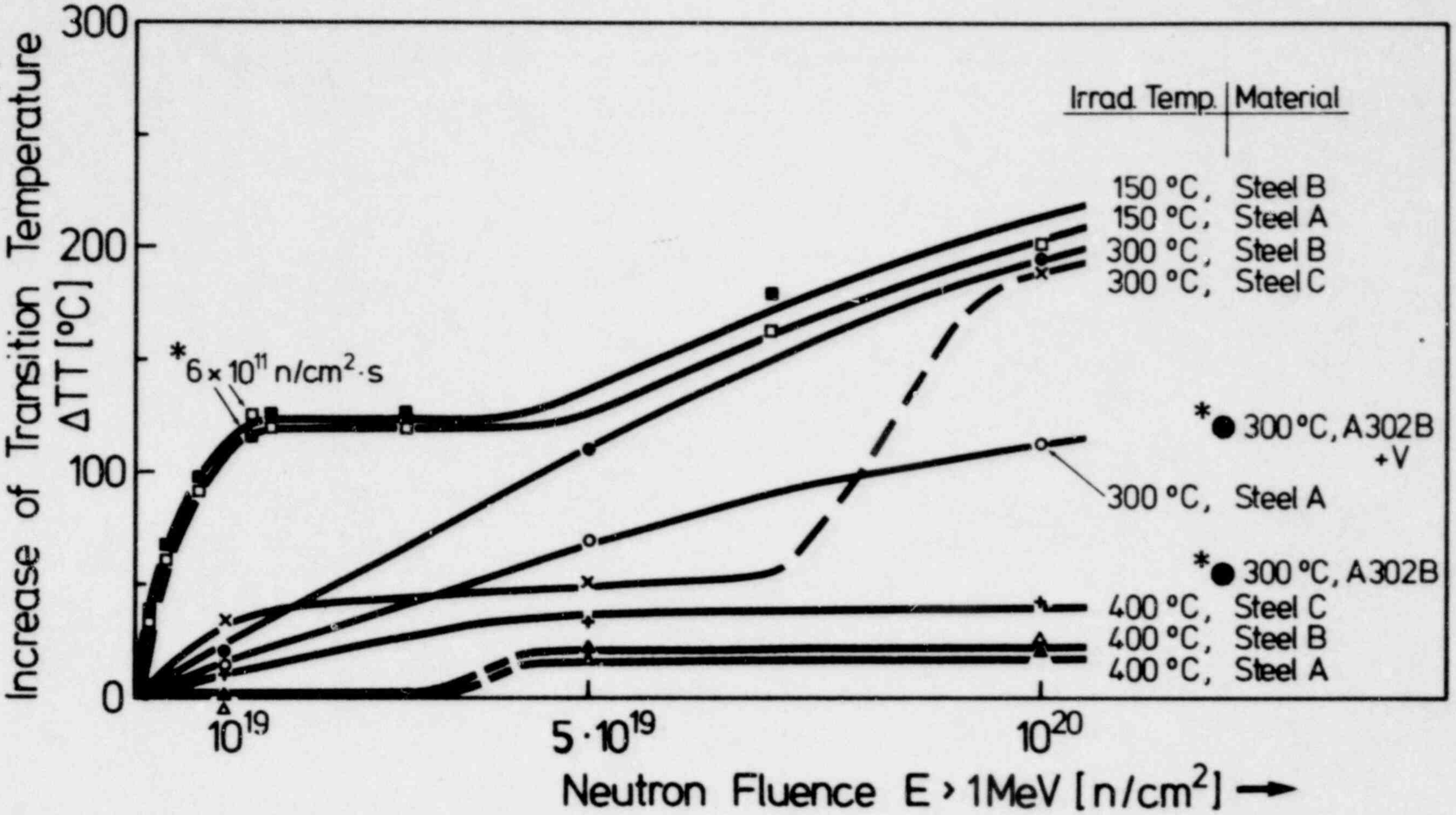

Fig.2 : Effect of Neutron-Fluence (E >1 MeV) on Increase of Transition Temperature

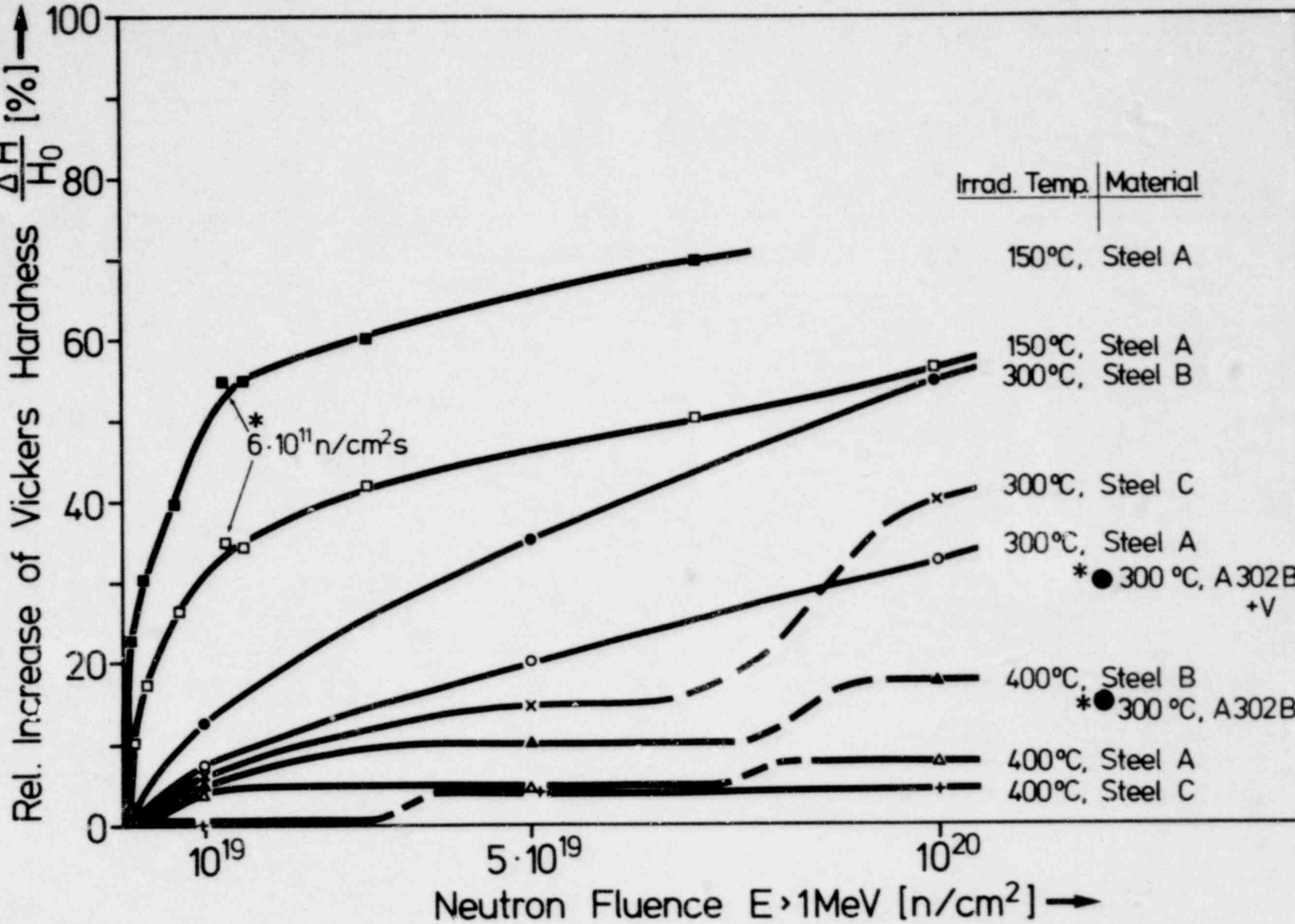

Fig.3 : Effect of Neutron-Fluence (E > 1 MeV) on Relative Increase of Vickers Hardness

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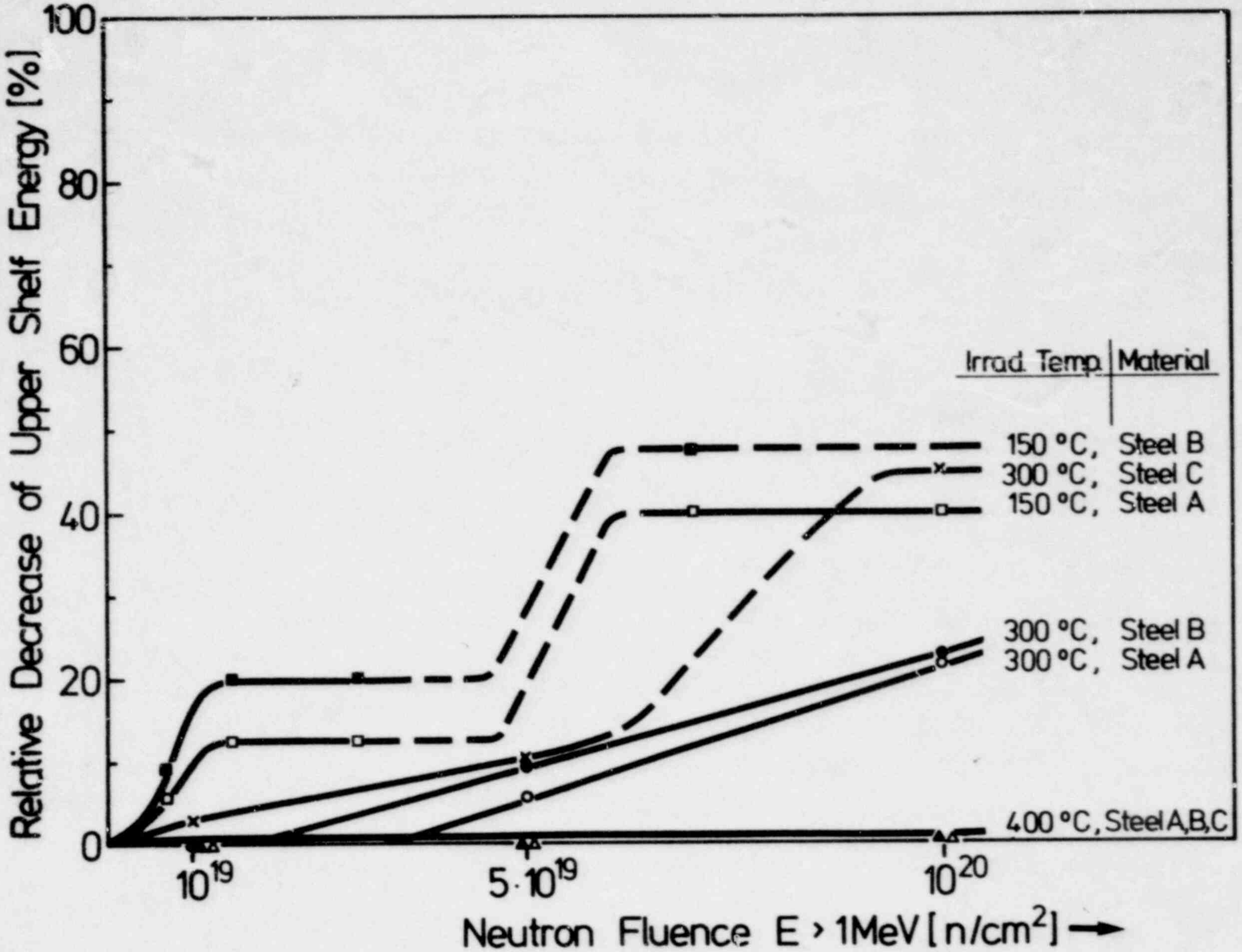

Fig.4 : Effect of Neutron-Fluence (E > 1 MeV) on Decrease of Upper Shelf Energy

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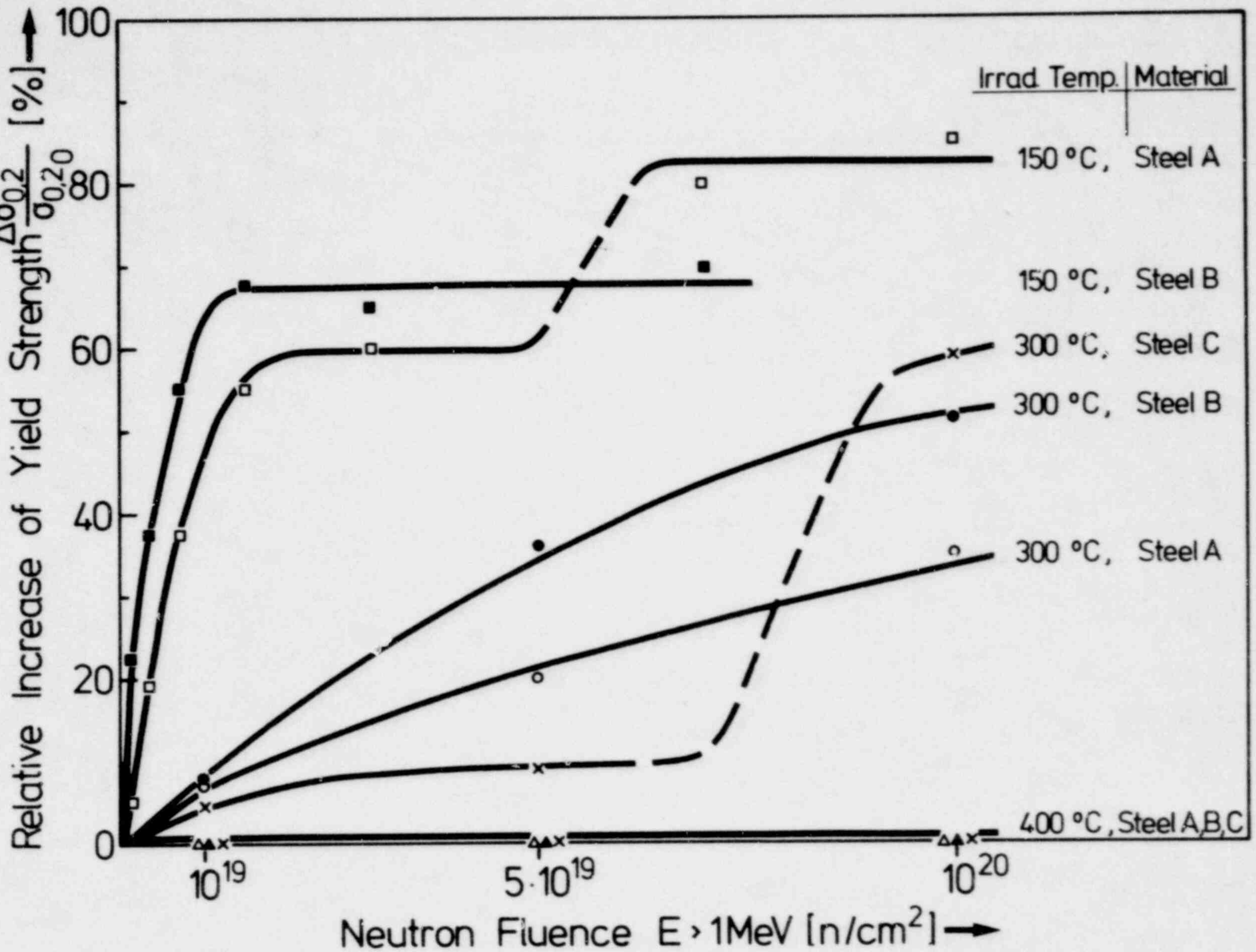

Fig.5 : Effect of Neutron-Fluence (E >1 MeV) on Relative Increase of Yield Strength

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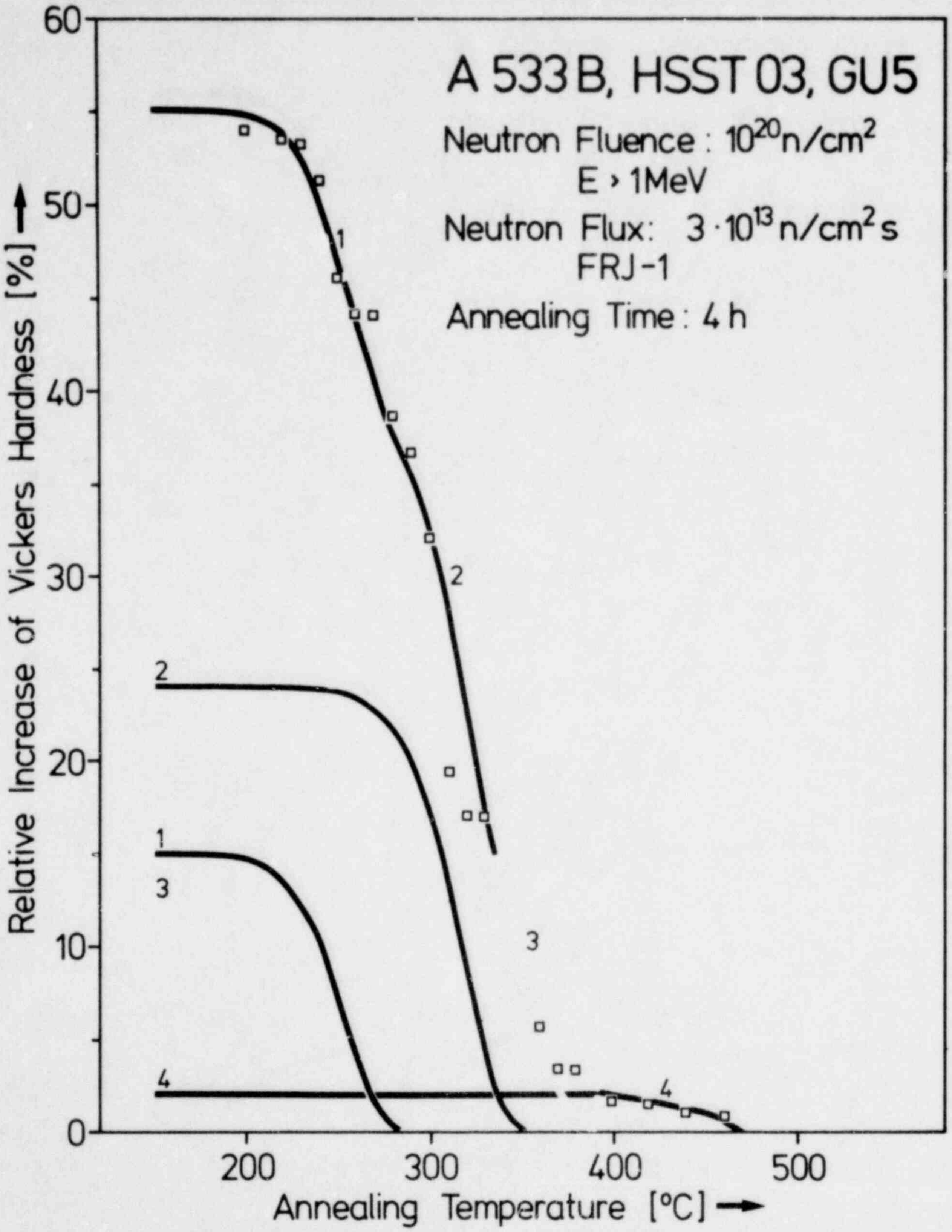

Fig. 6 : Recovery of the Relative Increase of Vickers Hardness for Steel A irrad. at 150 °C as a Function of Annealing Temperature, curve calculated according to equation

$$\Delta H = \Sigma_n H_n \cdot e^{-\lambda_o \cdot t \cdot e^{-U_n/kT}}$$

The contribution of the individual process is shown.

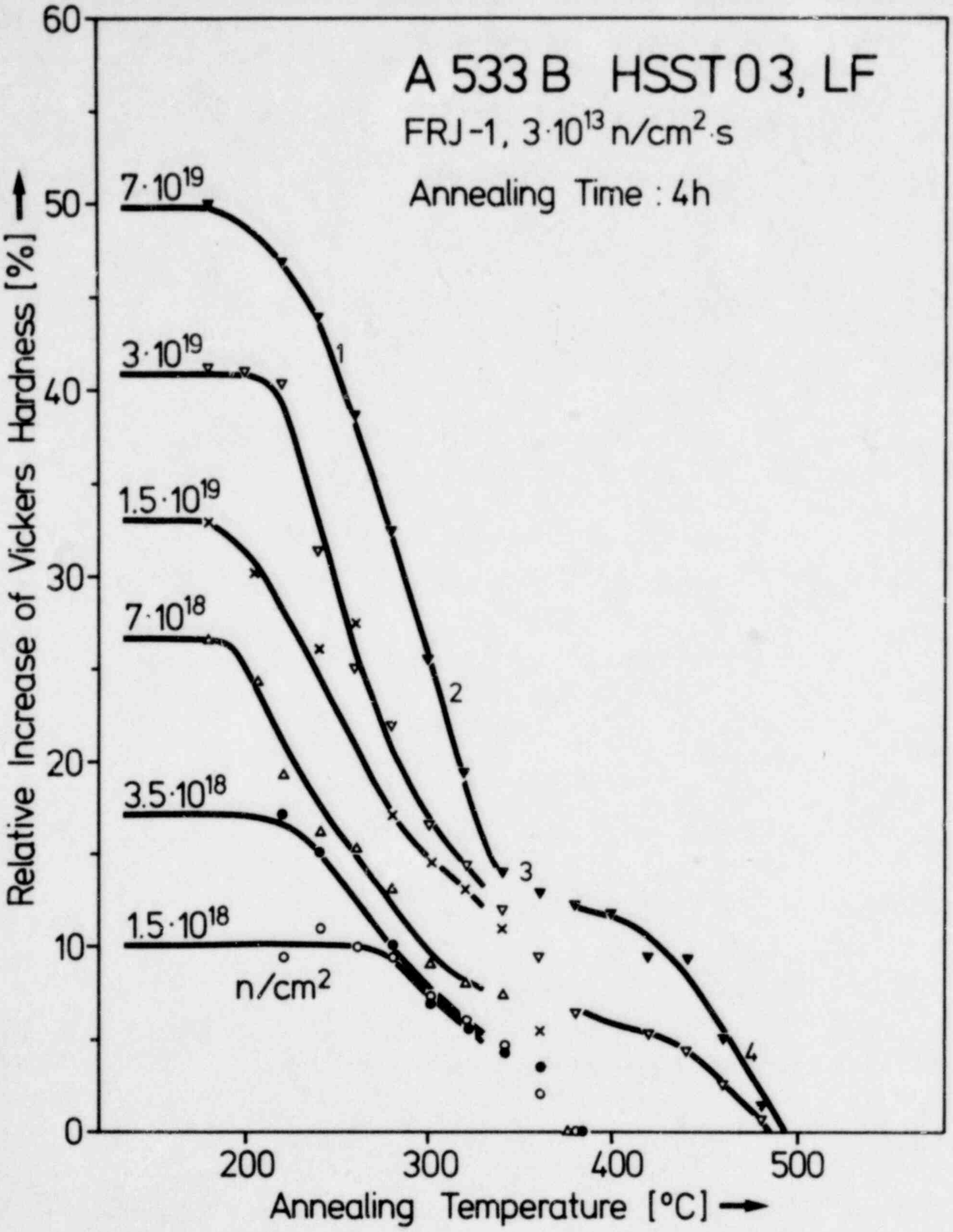

Fig. 7 : Recovery of the Relative Increase of Vickers Hardness as a Function of Annealing Temperature (isochronal 4 h) for Steel A Irradiated at 150 °C up to Various Neutron Fluences

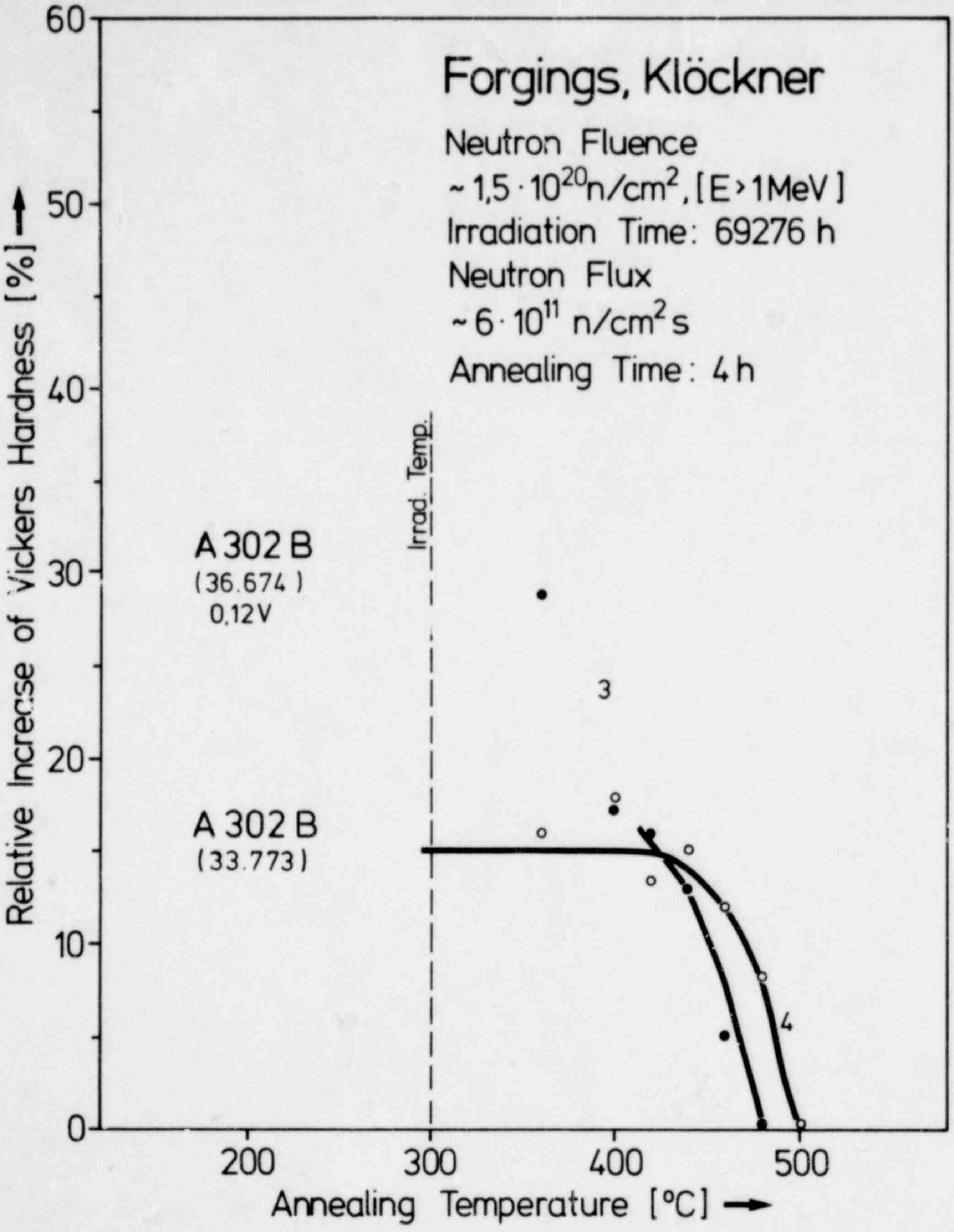

Fig. 8 : Recovery of the Relative Increase of Vickers Hardness as a Function of Annealing Temperatur (isochronal 4 h) for Steel A 302, Klöckner Forging, irradiated at ∿300 °C to 1.2 x 10^{20} n/cm^2

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