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# CORRELATION OF MECHANICAL PROPERTY CHANGES IN NEUTRON IRRADIATED PRESSURE VESSEL STEELS ON THE BASIS OF SPECTRAL EFFECTS

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# CORRELATION OF MECHANICAL PROPERTY CHANGES IN NEUTRON IRRADIATED PRESSURE VESSEL STEELS ON THE BASIS OF SPECTRAL EFFECTS - H. L. Heinisch, (Pacific Northwest Laboratory)[a]

## OBJECTIVE

To investigate the effects of the neutron spectrum on mechanical property changes in metals.

## SUMMARY

Defect production functions derived from atomistic modeling were evaluated for use in correlating yield stress changes of A212B and A302B pressure vessel steels irradiated in a wide variety of neutron spectra at low temperatures (40-90°C) and low doses (< 0.1 dpa). The irradiations were performed in RTNS-II, OWR, ORR and the HFIR pressure vessel surveillance positions. The data from RTNS-II, OWR and ORR are correlated fairly well on the basis of dpa, but the data from HFIR show that only one tenth as many dpa are needed to produce the same radiation-induced yield stress changes as in the other neutron spectra. About 96% of the neutrons in the HFIR surveillance position are thermal neutrons, and a significant fraction of the displacements is produced by recoils from thermal neutron captures. The best correlation of all the data is achieved when the property changes are compared on the basis of the production of freely migrating self-interstitial defects, which better represents the defects participating in the radiation strengthening process.

## PROGRESS AND STATUS

### Introduction

The effects of the neutron spectrum on tensile property changes of metals at relatively low doses (< 0.1 dpa) and irradiation temperatures from 25 - 450°C have been studied in an ongoing series of experiments that includes A302B pressure vessel steel[1]. Irradiations were performed with 14 MeV neutrons at the Rotating Target Neutron Source (RTNS-II) and with pool type reactor neutrons at the Omega West Reactor (OWR). A212B pressure vessel steel was later included[2] in OWR irradiations to extend the data base for the pressure vessel steel of the High Flux Isotope Reactor (HFIR), which had been observed to undergo embrittlement much sooner than original projections had predicted[3].

A212B and A302B have very similar compositions, especially with respect to the elements suspected of affecting embrittlement. A212B and A302B steels displayed the same irradiation hardening when irradiated in OWR[2], and it is reasonable to assume they will harden the same in any neutron spectrum. This assumption effectively broadens the data base for low-dose radiation-induced yield stress changes of A302B and A212B to include four very different neutron spectra. Tensile data have been obtained for A212B irradiated at 50°C in the HFIR pressure vessel surveillance positions and in the Oak Ridge Reactor (ORR)[3], for A212B at 90°C in OWR[2] and for A302B at 90°C in OWR and RTNS-II[1].

In this report the radiation-induced changes in yield stress occurring in all four neutron spectra will be compared on the basis of several damage parameters, including defect production functions derived from atomistic modeling results. In making these comparisons it is assumed that A302B and A212B behave similarly in low-dose, low-temperature irradiations and that any differences in behavior due to the differences in irradiation temperatures (90°C and 50°C) are small.

### The Neutron Spectra

Characteristics of the four neutron spectra used in the computations are listed in Table 1. The HFIR and ORR spectra were supplied by K. Farrell of Oak Ridge National Laboratory (ORNL). The HFIR spectrum is a calculated spectrum. The ORR spectrum is a modification of that for position A9 within the core, where A212B Charpy specimens were irradiated. The tensile specimens were irradiated outside the core at P8, where the neutron spectrum is similar at high energies, but has about 70% more thermal neutrons. An additional thermal neutron contribution was added to the A9 spectrum, which was then used in the computations. The RTNS-II and OWR spectra were supplied by L. R. Greenwood of Argonne National Laboratory (ANL). The variation in neutron spectra is quite extreme: the HFIR pressure vessel surveillance flux is dominated by thermal neutrons, while the RTNS-II flux consists entirely of 14 MeV neutrons. In RTNS-II the magnitude of the neutron flux decreases with distance from the source, so doses varying by about a factor of 100 were achieved by placing specimens at increasing distances from the source during the same run. Damage rates in RTNS-II varied from $3 \times 10^{-11}$ to $3 \times 10^{-9}$ dpa/s, placing the damage rate of RTNS-II between that of HFIR and the other reactors.

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TABLE 1
Characteristics of Neutron Spectra

| | HFIR[a] | ORR[b] | OWR[c] | RTNS-II[d] |
|---|---|---|---|---|
| Total Flux ($n/cm^2/s$) | 1.2e+10[f] | 1.8e+14 | 1.9e+14 | 1.0e+12 |
| Thermal Flux Fraction | 0.96 | 0.56 | 0.42 | 0.0 |
| Flux Fraction ($E > 0.1$ MeV) | 0.022 | 0.15 | 0.32 | 1.0 |
| Flux Fraction ($E > 1.0$ MeV) | 0.015 | 0.072 | 0.17 | 1.0 |
| Damage Rate in Iron (dpa/s) | 3.9e-13 | 1.9e-8 | 5.7e-8 | 3.0e-9 to 3.0e-11 |

a surveillance position, key 7, location 7, at pressure vessel
b location P8, outside core (based on in-core spectrum at A9)
c in core
d peak flux; fluxes in RTNS-II vary with distance from the source
f "e+10" should be read "x $10^{10}$"

### Damage Correlation Parameters

Meaningful comparison of mechanical property changes of materials irradiated in different neutron environments requires a damage correlation parameter that accounts for the effects of the spectrum of neutron energies in the materials. Comparing property changes on the basis of the measured fast neutron fluence (e.g. $E > 1$ MeV) is one attempt at incorporating spectral sensitivity, recognizing that more defects per neutron are produced by higher energy neutrons. How well this works depends on the neutron spectra involved. Clearly, damage in a largely thermal neutron spectrum, which can produce recoil atoms through thermal neutron-gamma capture reactions, is poorly represented by the fast neutron flux.

Dpa is in wide use as an exposure index and as a correlation parameter. It is a measure of the average number of times an atom of the material can be displaced to a stable defect position during an irradiation, and it takes into account the energy lost to inelastic processes that cannot produce displacement damage. The displacement cross section or displacement energy cross section is calculated for the given neutron spectrum and material[4]. Calculation of dpa requires a neutron spectrum, a set of neutron reaction cross sections, a model of the kinematics of the reactions that produce primary atomic recoils, a model for the dissipation of the primary recoil energy as electronic excitation and damage energy, and a model for the conversion of damage energy into dpa.

Dpa is a measure of the potential to create point defects. It is not necessarily equal to, or even proportional to, the actual number of residual point defects. The actual number of point defects present in a material at any time depends on the temperature and the material's history, including the neutron fluence. It also depends on the number of defects produced during primary damage production in the recoil events, which is dependent on the recoil energy[5].

Within a collision cascade created by a high-energy recoil, a significant fraction of the hundreds of initially produced point defect pairs (Frenkel pairs of vacant sites and self-interstitial atoms, referred to here as "vacancies" and "interstitials") will recombine as the locally high energy density in the cascade region dissipates. Of the remaining defects, most form into clusters (the stability of which depends on the crystal temperature), while a small fraction escapes the cascade region, becoming freely migrating defects. The fractions of initially produced defects that recombine or become freely migrating defects are constant above a minimum recoil energy (on the order of 10-100 keV). At the other end of the energy scale, low-energy recoils that can create only a few isolated defect pairs do not have high enough energy density to drive significant correlated recombination or clustering; nearly all the defects produced become freely migrating defects. Thus, the efficiency of defect production relative to calculated dpa values is a function of recoil energy, becoming constant at higher energies.

Dpa can be an effective damage correlation parameter for irradiation in different neutron environments only if the property change of interest is influenced by a quantity that is proportional to dpa. Proportionality of the damage to dpa can be influenced by the rate of damage production as well as the spectrum. Since environments with different neutron spectra usually have different damage rates, failure to correlate data on the basis of dpa has often been attributed to rate effects.

For example, initial comparison[3] of HFIR pressure vessel surveillance Charpy data with reference data at higher damage rates was made on the basis of fast neutron fluence. The order-of-magnitude "accelerated embrittlement" of the HFIR pressure vessel steel was attributed to unspecified rate effects. Comparison of the HFIR data with recent low-fluence tests by Nanstad et al. in ORR[6] showed the same accelerated embrittlement in both tensile and Charpy tests when compared on the basis of fast fluence (and dpa obtained from the fast fluence). The concluded that the results could be explained by either rate effects or spectral effects, but not unambiguously, since the effects are concurrent and inseparable in this case.

Failure of data to correlate on the basis of dpa can be due to the importance of the more detailed spectral effects. Property changes are likely to be sensitive to the number of residual point defects, the numbers of freely migrating defects, or the numbers of defect clusters contained in collapsed displacement cascades. All of these quantities vary with the recoil spectrum and are proportional to dpa only at high recoil energies.

The existence of tensile data for these pressure vessel steels over a wide range of neutron spectra and damage rates, from RTNS-II to HFIR, provides an opportunity to investigate other spectrally sensitive quantities as correlation parameters. In an earlier consideration of these data with respect to damage rate[2], it was found that there was no apparent effect of damage rate for the OWR, ORR and RTNS-II irradiated material. Furthermore, if the "accelerated embrittlement" of the HFIR surveillance material is due to a rate effect, then this effect would be important only below a very low flux. We will examine here possible spectral effects on these steels, based on damage production functions derived from atomistic modeling results for freely migrating vacancies, freely migrating interstitials, and total residual point defect pairs.

## Computations

Greenwood's SPECTER computer code[7] was used to generate the primary recoil spectra for iron into which the damage correlation functions were folded. Some issues with regard to using this code should be discussed. The SPECTER code is not particularly well suited to cases where thermal neutrons play a large role in the damage process. For most cases for which the code was developed, thermal neutrons have been thought to have little effect. Thus, in calculating some quantities, including the recoil spectra, the code omits the contributions due to thermal neutron captures. They can easily be added manually, at least to the level of precision necessary for the present investigation. Another more serious difficulty in the computations is that the integration schemes for obtaining and utilizing the recoil spectra lead to errors on the order of 20-30% in calculating the quantities from which they were derived. The effect of this for our application is that the criteria for "correlation" must be somewhat looser than if these quantities could be calculated exactly. Nevertheless, the extreme cases of neutron spectra are so different that the effects are large, and the main points of this report are well-established.

The damage correlation functions reported in reference 8 were used, although they were originally derived from simulations of cascades in copper. Parameter values in the expression for residual defects were modified according to Simons' prescription[5] to reflect more appropriate values for iron. The recoil energy-dependent damage functions that describe the partitioning of the residual defects into freely migrating defects and clusters were taken to be the same for iron as for copper. While the magnitudes are expected to be somewhat different for copper and iron, the recoil energy dependence of these functions should be qualitatively the same for both metals.

## Results

Irradiation temperatures for HFIR and ORR were 49°C and 43°C, respectively, while the RTNS-II and OWR irradiations were at 90°C. All the tensile tests were done on miniature tensile specimens. Those used at ORNL for HFIR and ORR were about four times larger in cross-sectional area than those for the RTNS-II and OWR tests done at PNL.

In Fig. 1 the radiation-induced yield stress changes at room temperature in A302B and A212B pressure vessel steels are plotted as a function of fast neutron fluence (E > 1 MeV) in the various irradiation environments. Each point is the result of an individual test. The hand-drawn curve is simply to aid visual organization. It is reproduced in the same position relative to the OWR data in each figure.

(At the highest dose in ORR the yield stress changes are significantly smaller than in OWR. It is possible that this difference may be due to the irradiation environment. The ORR specimens were irradiated in contact with the reactor coolant water, and it was found[3] that, despite being anodized to reduce rusting, those specimens irradiated for more than a few days displayed considerable rusting. The OWR specimens were irradiated in helium-filled capsules. However, in several of the earlier OWR runs the capsules developed leaks, and the specimens were exposed to reactor coolant water for several days or more, becoming rusty. The irradiations were redone, and both the clean and rusty specimens were tested. The rusty specimens at three fluences uniformly yielded at 70-80 MPa less than the clean specimens. No further tests have been done to determine the cause of these differences. Assuming the differences are related to the specimen surface, one expects the smaller OWR specimens to be more affected than the ORR specimens. Nevertheless, the ORR results with rusty specimens may result in somewhat lower values than expected for clean specimens.)

While the general trend of radiation hardening is observed in the data for all spectra in Fig. 1, the data can hardly be said to be well-correlated. In particular, in HFIR the fast fluence necessary to produce a yield stress change of 50 MPa is less by an order of magnitude than that in the other spectra.

In Fig. 2 the data are replotted as a function of calculated dpa. For all but HFIR, the data grouping is much tighter, being within a factor of 2 in dpa.

Fig. 3 shows the yield stress data replotted as a function of freely migrating interstitials per atom (fmipa). The number of fmipa needed to produce a yield stress change of 50 MPa in HFIR is within a factor of two of that needed in OWR, ORR and RTNS-II. Relative to their separation in the dpa plot, Fig. 2, the HFIR data have been shifted toward the OWR data by a factor of 4.4.

In Fig. 4 the yield stress data are plotted as a function of freely migrating vacancies per atom (fmvpa). Relative to the dpa plot, the HFIR data are shifted toward the OWR data by a factor of 14.2, which places them at a higher fmvpa level than OWR for the production of the same yield stress change.

Compared on the basis of total residual defect pairs (not shown), the tensile data for HFIR shift toward the data for the other spectra by a factor of 1.5 compared to the dpa plot.

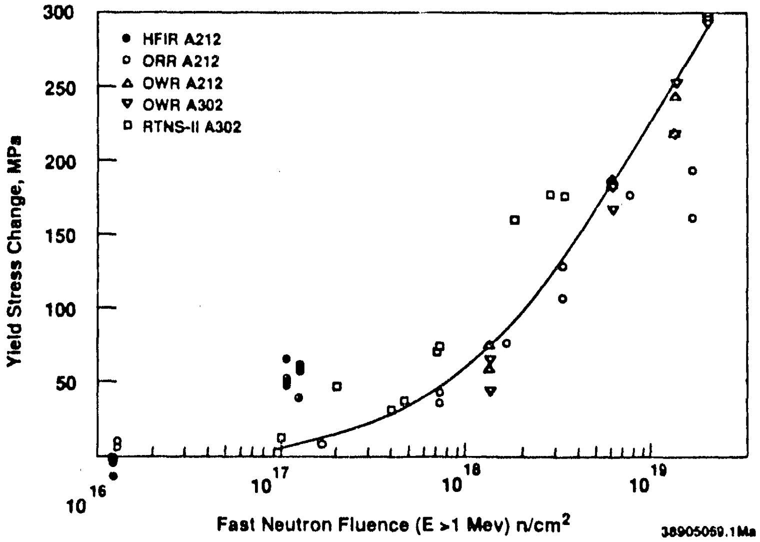

Figure 1. Change in 0.2% offset yield stress of A302B and A212B pressure vessel steels as a function of fast neutron fluence (E > 1 MeV). The hand-drawn curve is simply to aid visual organization. It is reproduced in the same position relative to the OWR data in each figure.

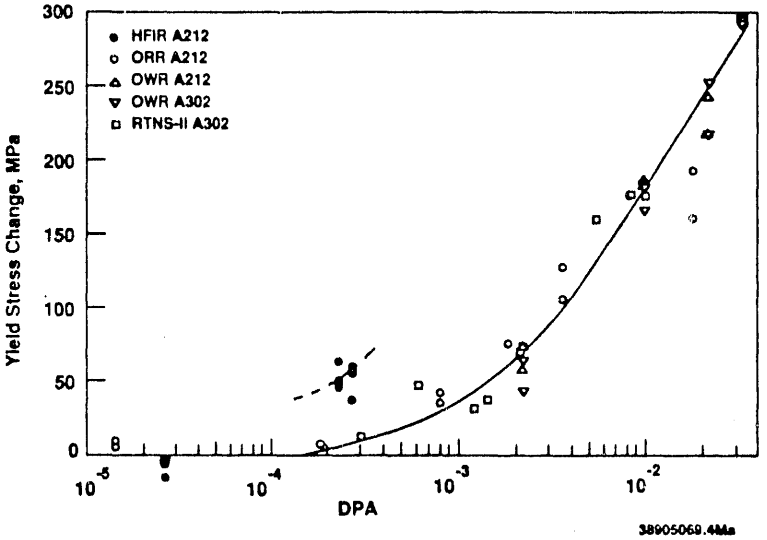

Figure 2. Change in 0.2% offset yield stress of A302B and A212B pressure vessel steels as a function of dpa.

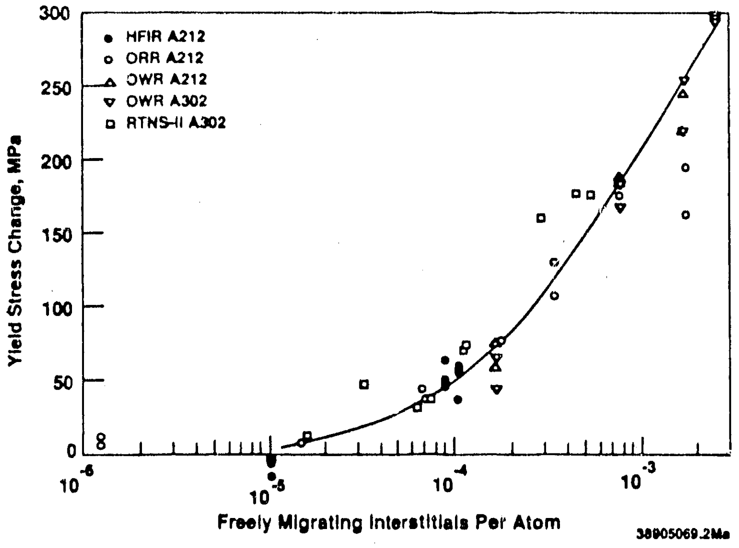

Figure 3. Change in 0.2% offset yield stress of A302B and A212B pressure vessel steels as a function of freely migrating interstitials per atom (fmipa).

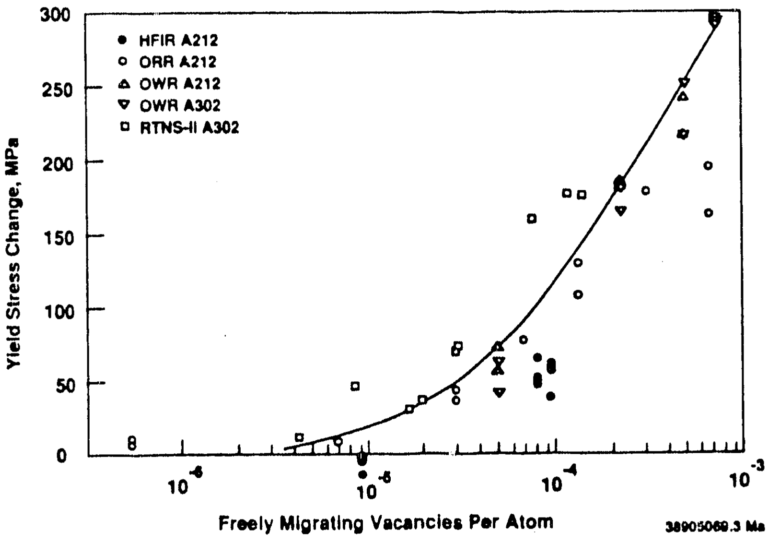

Figure 4. Change in 0.2% offset yield stress of A302B and A212B pressure vessel steels as a function of freely migrating vacancies per atom (fmvpa).

## Discussion

The best correlation of the yield stress changes of A212B and A302B pressure vessel steels irradiated in different neutron facilities is obtained when the data are compared on the basis of fmipa. Since the Charpy test data and tensile test data exhibit the same dependence on fluence[3], the same correlation will occur for embrittlement when fmipa is used as a damage parameter.

The HFIR results show a dramatic difference relative to the results in the other spectra when fmipa or fmvpa are used as damage parameters instead of dpa. This is the result of the 96% thermal neutron spectrum at the HFIR surveillance positions, combined with the increased efficiency of freely migrating defects per dpa at low energies. Thermal neutron captures in iron produce recoils with an average energy of 395 eV, enough to make about 4 displacements per recoil. Of the displaced atoms, an average of 2.4 displaced atoms per recoil become freely migrating interstitials (according to the model used here). In contrast, a 200 keV recoil produces on average about 1100 displacements and 63 freely migrating interstitials.

The defect production functions used in these calculations were derived from binary collision simulations of collision cascades and random jump simulations of short-term annealing in copper. Parameters in the semi-empirical annealing simulation were adjusted such that the fractions of freely migrating defects inferred from experiments were reproduced for non-interacting higher energy cascades (where they are independent of recoil energy). The energy dependence of freely migrating defect production was determined by modeling the annealing of thousands of cascades over a wide range of energy. Thus, the shapes of the defect production curves are a consequence of the initial spatial distributions of the defects. Cascades in iron have a slightly more diffuse character compared to cascades in copper because of the relatively more open bcc crystal structure. The efficiency of producing residual defects is higher in iron than copper (40% vs 30%). However, the two metals should exhibit the same relative energy dependence of free defect production because increased efficiency at lower energies is due more to the small number of defects produced than the details of the energy density.

Various types of microscopic analyses provide good evidence that the increases in yield stress and embrittlement of A212B irradiated in HFIR and ORR are due to irradiation hardening involving hardening centers too small to be imaged by transmission electron microscopy[3]. Post-irradiation annealing studies on A212B[3] showed no significant annealing below 300°C. Thus, at irradiation temperatures below 100°C small clusters are expected to be relatively stable.

Hardening centers can consist of defect clusters formed both immediately within cascades and by diffusion of the freely migrating defects. The freely migrating defects can recombine, go to existing sinks, form clusters or form complexes with interstitial impurities. Effectiveness of pure vacancy or interstitial clusters as hardening centers depends on the size of the clusters, although observations show that no clusters are very large. If pure interstitial or vacancy clusters are the primary source of hardening, then the data should correlate well on the basis of total residual point defects (especially if cluster size distributions are about the same), but they do not. Impurity complexes may be the most effective hardening centers. This is consistent with the successful correlation of the tensile data on the basis of freely migrating defects.

Since pure vacancy and interstitial clusters probably contribute to the hardness, the best damage correlation parameter would be a properly weighted combination of freely migrating interstitials and clusters produced directly in cascades. Additional experimental information or more extensive modeling will be necessary to determine the optimum combination for use as a damage function.

The above arguments suppose the effects of damage rate are not important. Indeed, there is no evidence of damage rate effects in tensile data in the range from $3 \times 10^{-11}$ to $2 \times 10^{-8}$ dpa/s[2]. A damage rate effect would have to be significant only at a threshold rate somewhat less than $10^{-11}$ dpa/s. Irradiations of these pressure vessel steels to low doses in a very high flux of thermal neutrons would help determine whether spectral or rate effects are responsible for the behavior observed in the HFIR surveillance specimens.

## REFERENCES

1. H. L. Heinisch, "Effects of the Neutron Spectrum on Mechanical Property Changes in Low Dose Irradiations," J. Nuc. Mater. 155-157, 121 (1988).

2. M. L. Hamilton and H. L. Heinisch, "Tensile Properties of Neutron Irradiated A212B Pressure Vessel Steel," Proceedings of the 14th ASTM International Symposium on Radiation Effects in Metals, Andover, MA, June 27-29, 1988.

3. "Evaluation of HFIR Pressure-vessel Integrity Considering Radiation Embrittlement," R. D. Cheverton, J. G. Merkle and R. K. Nanstad, eds., ORNL/TM-10444 (1987).

4. D. G. Doran and N. J. Graves, "Displacement Cross-sections and PKA Spectra: Tables and Applications," HEDL-TME 76-70 (1976).

5. R. L. Simons, "Evaluation of Defect Production Cross Sections and Calculated Fission-fusion Neutron Spectrum Sensitivity," J. Nuc. Mater. 141-143, 665 (1986).

6. R. K. Nanstad, K. Farrell, D. N. Braski and W. R. Corwin, "Accelerated Neutron Embrittlement of Ferritic Steels at Low Fluence: Flux and Spectrum Effects," J. Nuc. Mater. 158, 1 (1988).

7. L. R. Greenwood and R. K. Smither, "SPECTER: Neutron Damage Calculations for Materials Irradiations," ANL/FPP/TM-197 (1985).

8. H. L. Heinisch and F. M. Mann, "Neutron Cross Sections for Defect Production by High Energy Displacement Cascades in Copper," J. Nuc. Mater. 122-123, 1023 (1984).

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