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HELIUM EFFECT ON PHASE STRUCTURAL CHANGES IN CARBON STEEL UNDER POST-IRRADIATION PHASE RECRYSTALLIZATION

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Introduction

It is known that under neutron irradiation of materials not only the formation of radiation defects takes place but accumulation of transmutation elements as a result of various nuclear reactions as well. Though radiation defects are resistant in materials at temperatures of annealing not more than $0.6T_m$, transmutation elements can be conserved in irradiated materials up to melting point.

Among the most important in practical terms of transmutation elements, e.g. in thermonuclear materials science, are helium atoms which under high temperature irradiation or post-irradiation annealing are clustered in the form of helium bubbles remaining in materials till their melting point. Such heat resistance of helium bubbles is the reason of irreversible loss of constructional material plasticity. Moreover, in a number of works other peculiarities of helium effect on properties and structure of irradiated materials are determined. So, in the work [1] shown that helium promotes to remain interstitial dislocation loops during post-irradiation annealing up to high temperatures. Besides, the the work [2] it has been discovered that helium has a pronounced effect on redistribution of impurity elements and also on radiation-stimulated processes of phase structural destabilization in stainless steels [3].

In the work [4] one has found a new phenomenon in carbon steels irradiated by high-energy alpha-particles. It involves the formation of fine-grained ferrite-pearlite structure with simultaneous essential increase of ferrite phase fraction in local volumes of steel sample exposed to alpha-particles bombardment and post-irradiation annealing at temperatures of austenitic region of Fe-C diagram.

In this connection it is of interest to define the role of radiation defects and helium atoms in phase changes of carbon steel under post-irradiation $\alpha \Leftrightarrow \gamma$-transformation.

Experimental

As an object of investigations the U7 carbon steel (0,7% carbon) has been taken. After annealing in vacuum at 850°C for two hours ferrite-pearlite structure with a ferrite component distributed along the pearlite grain boundaries has been produced in samples.

The samples have been irradiated on the cyclotron by alpha-particles with energies of 29 and 50 MeV up to dose $1.5 \times 10^{17}\,cm^{-2}$ and by protons with energy of 10 MeV up to dose $2.5 \times 10^{17}\,cm^{-2}$ at temperatures not more than 100°C.

Two methods of irradiation with alpha-particles and protons have been applied. They are differ from one another by the condition of formation of zone doped with helium or hydrogen, respectively, in volume of samples (see Fig.1). As seen from Fig.1a under irradiation by the first method (method 1) the zone doped with helium (or hydrogen) is parallel to the bombarded surfaces at a depth corresponding to projective range of charged particles. The helium (or hydrogen) doped zone formed by irradiation on the second method (method 2, see Fig.1b) has a shape of a "chute"

separating the local irradiated zone from unirradiated matrix. Similar shapes of doping zones located on the bombarded sample surface at regular intervals have been produced under special irradiation conditions. One should note that the results of studying peculiarities of similar doping shaping zone in bulk pure metals indicate that due to lateral straggling of the projectiles the actual sizes of doping zone under irradiation by the method 2 are 5-7 times larger than under irradiation by the method 1 (Fig.2).

After irradiation the samples have been annealed in vacuum at temperature between 400 and 1200°C. Some irradiated samples after total decarburization at 1200°C have been exposed to one-side carburization (on the bombarded surface side) in activated carbon powder at 950°C. Carburizing treatment has been performed by two consequent stages to form a carburized case depth of 0,3 mm and up to total carburization of the whole sample volume.

The phase and structural changes in irradiated and unirradiated regions of samples have been detected by means of optical microscopy, X-ray diffraction and microhardness measuring.

Results

After irradiation both by alpha-particles and protons no structural changes have been observed in the investigated samples.

Microhardness measuring both on the bombarded surface and along the direct of particle motion showed the presence of radiation strengthening. As this takes place (see Fig.3), the changes of the value of microhardness growth along the range of projectiles are nonmonotonic nature. In the first zone the microhardness growth value is independent on particles energy. In the second one corresponding to the end of particle range (field of ion doping) a drastic increase of microhardness value is observed. In the sample region beyond the zone of ion range of the particles the misrohardness value corresponds to the unirradiated state of carbon steel. The observed difference in the microhardness growh value of samples irradiated by alpha-particles and protons corresponds in the first region to the level of damage by the given particles and in the second region - not only to the level of damage at the end of the range but to the effect of helium atoms on the portion increase of the retained defects which are responsible for radiation strengthening as well.

Thus, microhardness measuring evidences significant radiation damage of the irradiated part of the carbon steel samples.

Post-irradiation annealings at temperatures 400-900°G of the U7 steel samples irradiated by alpha-particles and protons (Fig.4) showed that in the sample regions irradiated by protons for "shooting through" and one doped with hydrogen the total recovery of microhardness growth has been observed even at 400°C. In samples irradiated by alpha-particles for "shooting through" the total recovery of microhardness growth has occurred at 600°C. At the same time in the helium doped sample region the total recovery of

microhardness growth has not been observed. Moreover, in the given region after annealing at 900°C the effect of "softening" has been noted, i.e. the value of microhardness decreases by 15% in comparison with unirradiated state.

Metallographic investigations of all as-irradiated and annealed at temperatures 400-600°C samples have not revealed any changes in pearlite structure. Temperature increase of post-irradiation annealing up to 900°C has left structural state of the U7 steel irradiated by protons invariant, while the annealing temperature exceeds 160°C of the $\alpha \to \gamma$-transition temperature for the given steel. Only in samples irradiated by alpha-particles after annealing at 900°C the essential structural changes have been observed (Fig.5). As seen from figure in the irradiated sample regions firstly, fine-grained ferrite-pearlite structure is produced, and secondly, the portion of ferrite phase practically is doubled. At the same time it has been noted that if the size of the "shooting through" region increases (see Fig.5b), similar structural changes take place only in helium doped sample regions (Fig.5c). Taking into account the absence of an analogous structural change in the U7 steel irradiated by protons (both in the region of "shooting through" by protons or alpha-particles and in the region of hydrogen doping) one can conclude that the presence of helium atoms in steel is a necessary condition for revealing the phenomen of structural changes in the carbon steel after irradiation and post-irradiation phase recrystallization.

X-rays diffraction study of the given sample of the U7 steel in the helium doped regions and unirradiated one indicate the same qualitative structure: they involve α-Fe solid solution and cementite. Comparising of cementite reflection intensity we suggest that with an accuracy of 20% the quality of cementite phase in all investigated regions is the same. A small intensity and width of diffraction lines make more detailed analysis of cementite phase difficult. At the same time the α-Fe solid solution reflections (110), (200) and (211) in the heliun doped sample regions and unexposed to irradiation one, differ both in intensity and shape: the lines obtained in the unirradiated regions of the sample have the greatest width (Fig.6). The reason of decreasing the α-Fe line width in the helium doped region sample is vague. Increasing of the intensity of one reflection is accompaned by decreasing the intensity of another one, but summar intensity is practically invariant. This, probably, is due to orientation redistributions of α-Fe grains in the helium doped regions in comparison with unirradiated regions during post-irradiation phase recrystallization.

Thus, X-ray diffraction data suggest that qualitative phase structure of the U7 carbon steel in the helium doped regions and unexposed to irradiation one is the same after annealing at 900°C.

The increase of the time of post-irradiation annealing at 900°C up to 6 h and further increasing of annealing temperature up to 1100°C showed that the above-mentioned phenomen of anomalous structural change in the helium doped region of the U7 steel sample is conserved. The temperature increase of post-irradiation annealing of the given sample

up to 1200°C (for 1 h) has resulted in its total decarburization and large helium bubbles formation in the helium doped regions (Fig.7).

In this connection, it was interesting to establish the fact of thermal irreversibility of the observable anomalous structural change in the helium doped U7 steel under post-irradiation phase recrystallization by means of carburization of the irradiated sample which decarbornizing treated by post-irradiation annealing at 1200°C. For this purpose the given sample has been subjected to one-side (on the bombarded side) carburization in activated carbon powder at 950°C for 5 and 15 minutes. Typical microstructures of the samples after one-side carburization at a depth of 0,3 mm (5 min) and till total carburization of the whole volume (15 min) are shown in Fig.8. It is well seen that the effect of anomalous structural change in helium doped sample zones manifesting in fine-grained ferrite-pearlite structure formation with an essential superiority of ferrite phase portion has been repeated. This proves conclusively the thermal irreversibility of the helium effect on phase and structural changes in the carbon steel under post-irradiation phase recrystallization.

Therefore the above results allow one to emphasize two important facts. First, the irradiation of the U7 carbon steel by high-energy protons and alpha-particles which results in formation of radiation defects and hydrogen doping leaves any structural changes invariant in the process of post-irradiation annealing at temperatures 400-1100°C. Second, due to post-irradiation annealing in the range of temperatures above the $\alpha\to\gamma$-transition temperature in the helium doped sample regions of the U7 steel observes thermal irreversible structural changes (perhaps phase changes too): the formation of fine-grained ferrite-pearlite structure with an essential superiority of ferrite phase portion.

From the above it might be assumed that the main reason of the observed effect is thermally stable complexes of helium atoms and vacancies in the form of helium bubbles rather than radiation defects. First, helium bubbles can essentially effect on the processes of nucleation and growth of austenite grains during phase recrystallization as a result of which fine-grained austenite is formed. The direct experiments performed with fine-grained austenitic steel doped with helium under alpha-particles irradiation by the method 2 and annealed to post-irradiation recrystallization have confirmed the given assumption.

As to the observed phenomen involving practically double increase of ferrite phase portion one can assume here the following. Firstly, on the base of X-ray diffraction data testifying the absence of phase and structural transition in the helium doped sample region one can suppose that helium bubbles promote the formation of globular pearlite rather than lamellar ones. The first has a less value of microhardness in comparison with a lamellar pearlite. Secondly, it is beyond reason to exclude from consideration a possibility of cementite decomposition on ferrite and graphite or graphite formation on the helium bubbles surface. And, finallly, helium bubbles growing in local

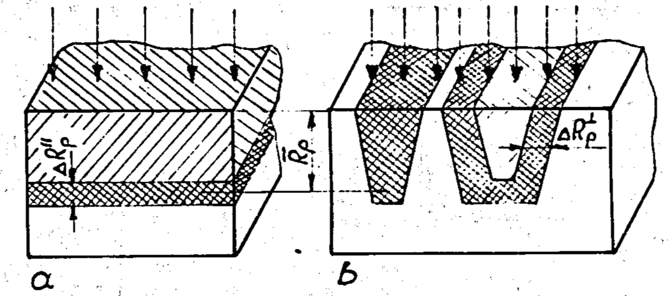

Fig.1 The scheme zones production in samples of the U7 steel irradiated by high-energy particles by the method 1 (**a**) and method 2 (**b**): – the region of alpha-particles (proton) "shooting through", – the helium (hydrogen) doped region, – unirradiated region, R_p – projective range of charged particles, $\Delta R_p^{\parallel}$ – longitudinal straggling, $\Delta R_p^{\perp}$ – lateral straggling.

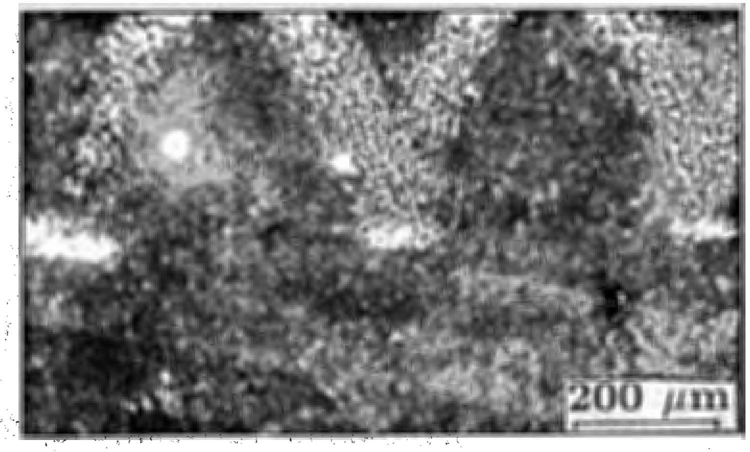

Fig.2 Helium bubbles in zones of copper doping irradiated by alpha-particles with E = 50 MeV by the method 2.

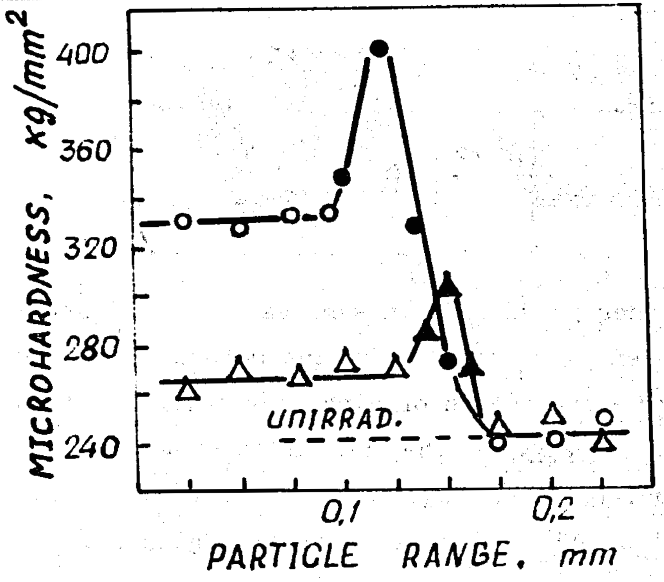

Fig.3 The change of the U7 steel microhardness value along the range of alpha-particles (E = 29 MeV) - (o,●) and protons (10 MeV) - (Δ,▲).

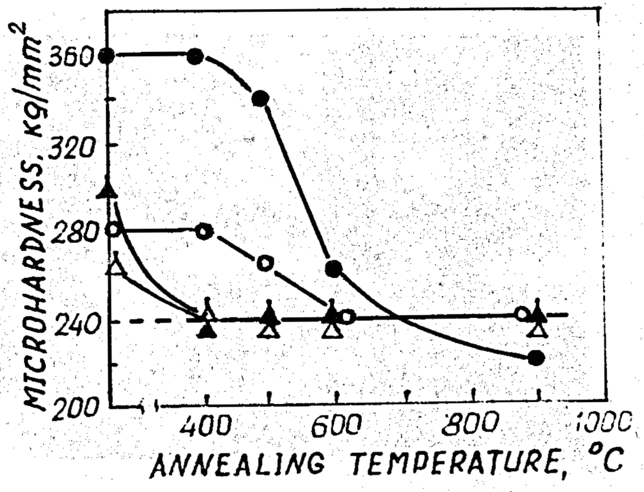

Fig.4 The change of microhardness value of the U7 steel irradiated by protons (Δ - region of irradiation for "shooting through", ▲ - hydrogen doped region) and alpha-particles (o - region of irradiation for "shooting through", ● - helium doped region) in dependence of post-irradiation annealing temperature.

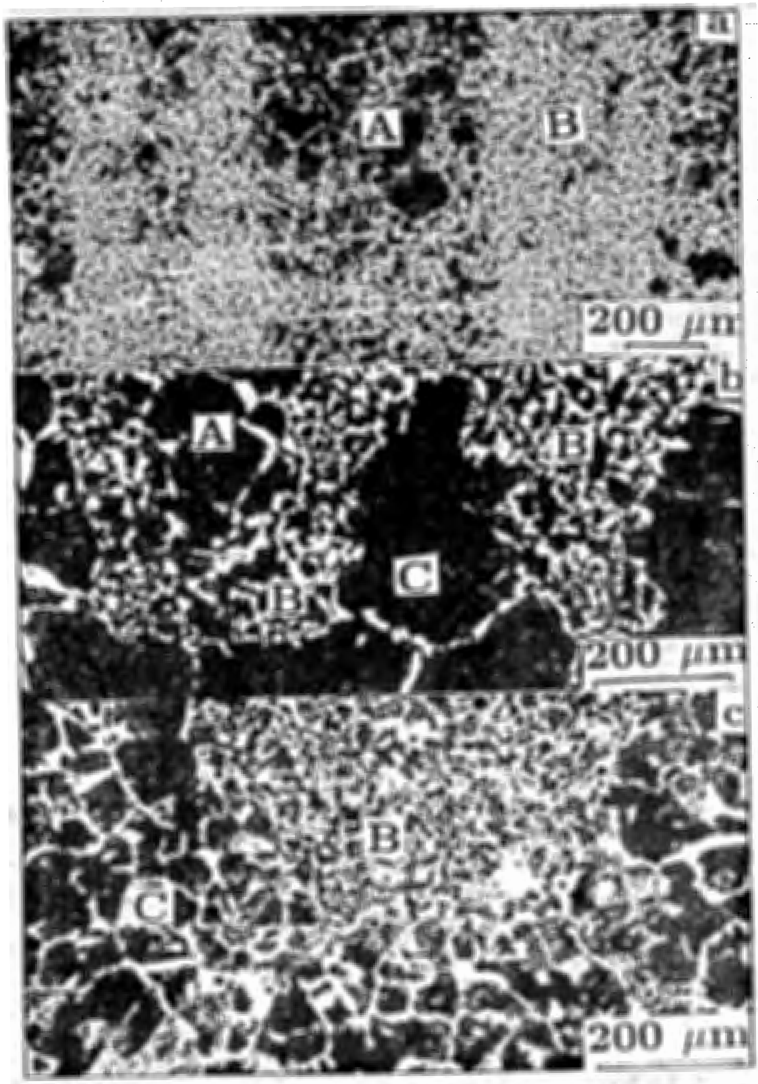

Fig.5 Structural changes in the regions of the U7 steel sample irradiated by alpha-particles by the method 2 and annealed at 900°C: a - structure of bombarded surface, b, c - structure along the direction of alpha-particle motion. A - region of irradiation for "shooting through", B - helium doped region, C - unirradiated region.

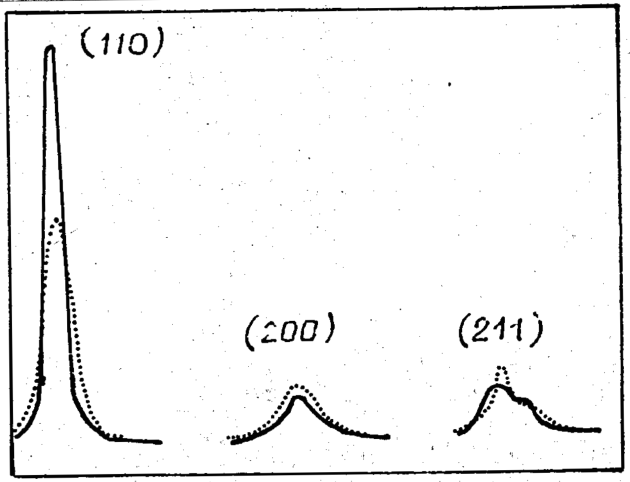

Fig.6 X-ray diffraction pattern of α-Fe in the U7 steel sample irradiated by alpha-particles and annealed at 950°C. (···) - unirradiated region of a sample, (——) - helium doped sample region.

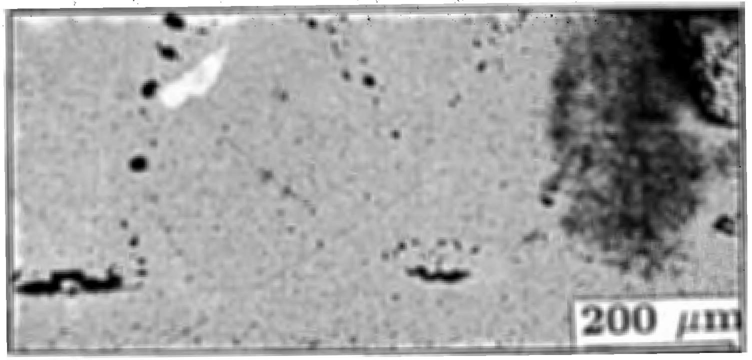

Fig.7 Decarburization and growth of helium porosity in the U7 steel irradiated by alpha-particles under post-irradiation annealing in vacuum at 1200°C for 1 hour.

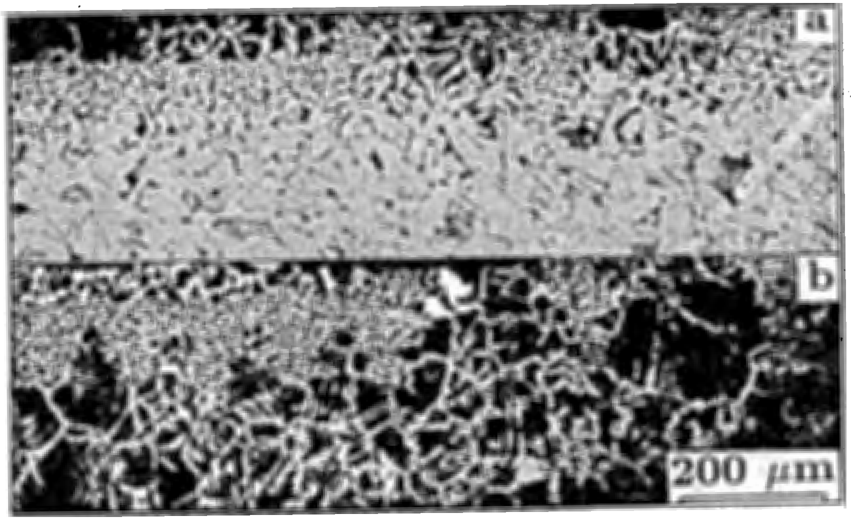

Fig.8 Structural changes in the U7 steel after alpha-particle irradiation, decarburization at 1200°C and followed by one-side carburization (on bombarded surface side) at 950°C for 5 (**a**) and 15 (**b**) min.

helium doped sample regions can cause a high level of compressive stresses (due to gas swelling) in them. A similar stress condition of the helium doped sample region of the U7 steel can facilitate both the increase of carbon desorption rate from it under post-irradiation annealing and, vice versa, the decrease of carbon desorption under high temperature post-irradiation carburization.

It is clear that a new helium role manifestation observed in phase and structural destabilization of carbon steel and, perhaps, in other polymorphous metals and alloys require for further studies of this problem by means of transmission electron microscopy, that appears to be important both for extending our knowledges of inert gas behaviour in metals and for radiation solid state physics as a whole, in particular, in the field of studying the effect of thermal stable radiation defects on allotropic transformations.

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Реутов В.Ф., Семина В.К., Турубарова Л.Г. E14-95-55
Влияние гелия на структурно-фазовые изменения в углеродистой стали
в условиях послерадиационной фазовой перекристаллизации

Методом металлографического анализа углеродистой стали У7 (0,7% С) после облучения протонами с $E = 10$ МэВ или α-частицами с $E = 29$ и 50 МэВ, отжига и цементации полностью обезуглероженных образцов установили два факта: 1) облучение высокоэнергетичными протонами и α-частицами, результатом которого является создание радиационных дефектов и легирование водородом, не вызывает каких-либо структурных изменений в процессе послерадиационного отжига в интервале 400—1200°С; 2) в участках образца, легированных гелием, в результате послерадиационного отжига при температуре выше температуры $\alpha \to \gamma$-перехода наблюдались термически необратимые структурные (возможно, и фазовые) изменения: образование мелкозернистой феррито-перлитной структуры с существенным преобладанием доли ферритной фазы. Предполагается, что основной причиной наблюдаемого эффекта являются не радиационные дефекты, а термически стабильные комплексы атомов гелия и вакансий в форме гелиевых пор.

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Helium Effect on Phase Structural Changes in Carbon Steel
Under Post-Irradiation Phase Recrystallization

The results of metallography study of as-irradiated with protons ($E = 10$ MeV) or with α-particles ($E = 29$ and 50 MeV), annealed and carburized after full decarburisation samples of the U7 carbon steel (0.7% C) allow to establish two important facts: 1) the irradiation with high-energy protons and α-particles which results in formation of radiation defects and doping with hydrogen leaves any structural changes invariant in the process of post-irradiation annealing in the range of 400—1100°C; 2) due to post-irradiation annealing above the $\alpha \to \gamma$ phase-transition temperature in the helium doped regions of α-irradiated U7 steel samples one observes thermal irreversible structural changes (perhaps phase changes too): the formation of fine-grained ferrite-pearlite structure with an essential superiority of ferrite phase portion. It might be assumed that the main reason of the observed effect is thermally stable complexes of helium atoms and vacancies in the form of helium bubbles rather than radiation defects.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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