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# The effect of neutron irradiation on the electrical resistivity of high-strength copper alloys

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#### Abstract

The effects of neutron irradiation on the electrical resistivity of precipitation hardened (PH) and dispersion strengthened (DS) copper alloys are discussed. The analysis is based on the experimental study of radiation damage of PH and DS copper alloys, irradiated in the fast neutron reactor BOR-60 with doses of  $8-16 \times 10^{25}$  n/m<sup>2</sup> and in the mixed spectrum neutron reactor SM-2 with doses of  $3.7-5.5 \times 10^{25}$  n/m<sup>2</sup>. The experimental data on the change  $\Delta \rho$  in electrical resistivity of DS-type copper alloys irradiated in the BOR-60 reactor show that irradiation to 7-10 dpa at T = 340-450°C causes a drop in electrical conductivity by not more than 20%. The obtained results show that in mixed-spectrum reactors the rate of  $\Delta \rho$  normalized to the dpa is about 20 times as high as in fast neutron reactors. The conclusion is made that the calculations performed for ITER must take into account the presence of appreciable fluxes of thermal neutrons in certain components of the reactor. The latter will play a decisive role in the drop in thermal conductivity of copper alloys in these components. © 1997 Elsevier Science B.V.

#### 1. Introduction

Copper-based alloys are the most promising materials for the heat sink system of the ITER energy-loaded components. The most attractive feature of copper alloys is their high thermal conductivity providing an effective removal of the heat flux, which is as much as 10-20MW/m<sup>2</sup> for the ITER divertor [1].

Phase-hardened (PH) alloys of Cu–Cr–Zr, Cu–Ni–Be type and dispersion-strengthened (DS) alloys of Cu–Al<sub>2</sub>O<sub>3</sub>, Cu–Mo type have high-strength characteristics and thermal conductivity at a level of 80-90% of pure copper.

Under neutron irradiation the thermal conductivity of copper alloys drops. As massive samples are difficult to irradiate, small samples are usually used in radiation experiments and a change in their electrical resistivity is measured after irradiation, so as to measure the irradiation effects on the thermal conductivity. Then the results of these measurements are converted into a change in electrical and thermal conductivity by the well-known Wiedemann–Franz relation. The available data base on the effect of irradiation up to substantial doses (> 0.5 dpa) on copper electrical resistivity are scarce. The majority of these data was obtained during irradiation in the fast neutron reactors EBR-II and FFTF [2–5]. These investigations performed at  $T_{irr} = 380-450^{\circ}$ C in the dose range of 3–63 dpa revealed that at doses of 16 dpa the electrical conductivity of DS copper alloys is decreased by about 20% and at a larger dose (about 60 dpa) by 40%.

The main reasons for a drop in the thermal and electrical conductivity of copper alloys are the accumulation of Ni and Zn caused by transmutation and the formation of radiation defect complexes in the material (dislocation loops and, at larger doses, pores). The qualitative assessment of the contribution of each of these effects to the total change in the electrical conductivity allowed the authors [4] to make the conclusion that the transmutation is responsible for 50% of the observed effect and the complexes of radiation defects for the rest 50%. Because the transmutation products accumulation rate expected for ITER is higher than that for fast neutron reactors [1], the

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data obtained in studies [2-5] do not provide conservative assessments for a change in thermal conductivity.

It is in study [6] that the first data on the effect of irradiation in a mixed-spectrum reactor on a change in the electrical conductivity of pure copper and DS copper alloys are presented, and the rate of growth in the electrical resistivity with the dose, when irradiated in SM-2, is shown to be higher than in the fast neutron reactors. The drop in electrical and thermal conductivity causes the temperature on a copper tile to increase with a possible consequent softening (especially dangerous for PH alloys [7]) and helium embrittlement (at  $T > 0.5T_{mell}$ ).

This study presents the results of experimental investigations of changes in the electrical conductivity of pure copper and PH and DS copper alloys irradiated in the fast neutron reactor BOR-60 with doses of  $8-16 \times 10^{25} \text{ n/m}^2$ at  $T_{\text{irr}} = 340-460^{\circ}$ C and in the SM-2 reactor in the dose range of  $3.7-5.5 \times 10^{25} \text{ n/m}^2$  at  $T_{\text{irr}} = 90-430^{\circ}$ C.

#### 2. Experimental procedure

In this work pure copper (Cu 99.97%), the dispersionstrengthened (DS) copper alloys MAGT 0.2, MAGT 0.05, Cu-5%Mo and precipitation hardened (PH) copper alloys Cu-Cr-Zr, Cu-Cr-Zr-Mg have been investigated. The composition and heat treatment of alloys are presented elsewhere [7,8].

Sheet tensile samples were irradiated in the BOR-60 reactor to doses of 5.6 to 11 dpa  $(8-16 \times 10^{25} \text{ n/m}^2, E > 0.1 \text{ MeV})$  at irradiation temperatures of  $340-460^{\circ}\text{C}$  and in the SM-2 reactor to doses of 0.6 to 3.9 dpa  $(0.9-5.5 \times 10^{25} \text{ n/m}^2, E > 0.1 \text{ MeV})$  at  $T_{\text{irr}} \approx 90^{\circ}\text{C}$ ,  $180^{\circ}\text{C}$ ,  $185^{\circ}\text{C}$ ,  $240^{\circ}\text{C}$ ,  $285^{\circ}\text{C}$ ,  $310^{\circ}\text{C}$ ,  $375^{\circ}\text{C}$ ,  $400^{\circ}\text{C}$ . The

irradiation was performed in special tubular capsules. The tubes were filled with helium. The irradiation temperature regime of the specimens was measured by thermocouples. The specimen geometry consisted of a gage length of 30 mm and a thickness of 1 mm. The electrical resistivity of all samples was measured at  $(20 \pm 2)^{\circ}$ C before and after irradiation using standard four-point probe techniques. The experimental uncertainty in the resistivity measurements was  $\pm 0.3 \ n\Omega$  m. The irradiation and testing techniques are described in more detail in Refs. [7,8].

#### 3. Results

### 3.1. Effects of fast neutron reactor irradiation on electrical resistivity of copper alloys

Fig. 1 presents the dose dependence of the electrical resistivity of specimens from PH copper alloys, DS copper alloys and pure copper irradiated in the BOR-60 reactor to a dose of  $0.8-1.6 \times 10^{26} \text{ n/m}^2$  at  $T_{\text{irr}} = 340-460^{\circ}\text{C}$ . As the materials in the initial state have a different level of electrical resistivity, the comparison of their behavior is rather complicated. However, the general tendency for all DS alloys is a 5-10% increase in electrical resistivity after irradiation. The same is true for pure copper except that the rate of  $\Delta \rho$  normalized to dpa is considerably higher (nearly twice). Note, that an increase in electrical resistivity for DS alloys is of monotonous and nearly linear character, the arrangement of alloys in electrical resistivity being preserved. The MAGT 0.05 alloy, in both irradiated and unirradiated states, has the lowest and Cu-5Mo the highest electrical resistivity. The MAGT 0.2 alloy is intermediate in value.



Fig. 1. Measured electrical resistivity ( $\rho_{trr}$ ) vs. fast neutron fluence (E > 0.1 MeV) of pure copper, DS copper alloys MAGT-0.2, MAGT-0.05, Cu–5Mo and PH copper alloys Cu–Cr–Zr, Cu–Cr–Zr–Mg, irradiated in BOR-60 reactor at 340–460°C.

For Cu–Cr–Zr, Cu–Cr–Zr–Mg alloys irradiated to  $0.8-1.6 \times 10^{26} \text{ n/cm}^2$  at  $T_{\text{irr}} \approx 340-460^{\circ}\text{C}$  the electrical resistivity is not practically increased with a rise in the irradiation dose, on the contrary  $\rho$  for these materials even drops by 2–3% at doses of  $8 \times 10^{25} \text{ n/cm}^2$ . The Cu–2%Be alloy with an extremely high electrical resistivity in the initial state ( $\rho_{\text{unirr}} = 62.7 \text{ n}\Omega$  m) demonstrates nearly a two-fold drop in electrical resistivity after irradiation to 11 dpa ( $\rho_{\text{irr}} = 33.7 \text{ n}\Omega$  m).

Thus, the experimental results demonstrate the clear-cut tendency: irradiation to  $\approx 10$  dpa causes an increase in the electrical resistivity of DS copper alloys and pure copper, changes only slightly  $\rho$  of the PH alloys Cu-Cr-Zr, Cu-Cr-Zr-Mg and reduces nearly twice the electrical resistivity of the Cu-2%Be alloy.

#### 3.2. Effects of mixed spectrum reactor irradiation on electrical resistivity of copper alloys

Fig. 2 presents the dose dependence of electrical resistivity  $\rho$  for the PH copper alloys Cu-Cr-Zr and Cu-C-Zr-Mg and DS copper alloys Cu-5Mo, MAGT 0.2, MAGT 0.05 irradiated in the SM-2 reactor in the dose range of 0.6-3.8 dpa at  $T_{\rm irr} = 90-400^{\circ}$ C. A monotonous gain in  $\rho$ with the dose increasing is observed, no matter what the irradiation temperature. It should be noted here that the gain in  $\rho$  is  $\approx 20 \text{ n}\Omega$  m at a maximum dose, i.e., the electrical resistivity of the DS alloys is doubled.

The PH copper alloys of Cu-Cr-Zr and Cu-C-Zr-Mg type demonstrate the similar change in  $\rho$  under irradiation (Fig. 2). The only difference is a considerable spread in data at high irradiation doses, this not being observed for the DS alloys. Besides, the  $\rho$  gain at maximum irradiation

doses ( $\approx 3.8$  dpa) can be as much as 23-29 n $\Omega$  m, this being higher than for the DS alloys.

#### 4. Discussion

4.1. The analysis of reasons for a change in electrical resistivity of copper alloys under irradiation in fast neutron reactor

Nowadays an understanding has been reached [1], that the following two factors are responsible for a change in electrical resistivity of copper alloys: the accumulation of the transmutation products Ni and Zn and the accumulation of radiation defects (loops, pores). Note here, that relatively simple systems are convenient for the analysis. From this standpoint the PH alloys are not the type suitable for this purpose. Investigations of the mechanical properties and the structure of Cu-Cr-Zr, MZC, Cu-Be alloys performed in [6-10] demonstrate that in the PH alloys considerable changes in the dislocation structure, polygonization and recrystallization occur at  $T_{irr} = 340$ -460°C. Besides, the density of strengthening fine particles drops. These effects cause PH alloys to soften. The yield strength can be reduced by a factor of 4-5 for Cu-Cr-Zr, Cu-Cr-Zr-Mg alloys at  $T_{irr} \approx T_{test} \approx 420^{\circ}$ C [11]. The problem of assessing the contribution of both components (dislocation structure and phase structure of the PH alloys) to a change in their electrical resistivity seems to be rather complicated. For this work to be accomplished each  $\rho$ measurement is to be synchronously accompanied with the detailed TEM investigation of a specimen and with the analysis of structural changes occurring under irradiation, which is obviously beyond the scope of this work.



Fig. 2. Measured electrical resistivity ( $\rho_{irr}$ ) vs. neutron damage dose (dpa) of DS copper alloys MAGT-0.2, MAGT-0.05, Cu-5Mo and PH copper alloys Cu-Cr-Zr, Cu-Cr-Zr-Mg irradiated in SM-2 reactor at 90-400°C.

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Pure copper seems to be a much simpler system than PH alloys, but it swells in the dose range of 10-15 dpa. In our study the swelling amounted to 3.8% at 7 dpa and  $T_{\rm irr} \approx 340^{\circ}$ C [6]. In Ref. [5] the swelling of pure copper irradiated to 13.5 dpa was as much as  $\approx 7\%$ . The assessment of the contribution of pure copper swelling to a change in electrical resistivity needs a separate consideration. In this work we deliberately restrict ourselves to the analysis of one of the simplest experimentally investigated systems, i.e., DS copper alloys. As follows from our results [6] and measurements of other authors [2-5], the DS copper alloys irradiated to 16 dpa at  $T_{irr} = 340-460^{\circ}C$ do not show evidence of swelling. Besides, the TEM investigations do not also reveal pores in these alloys [10,12,13]. Fine dislocation loops are the main radiation defects in these alloys [10,13,14]. Dispersion particles (Al<sub>2</sub>O<sub>2</sub> for GlidCop and MAGT alloys and Mo particles for Cu-5Mo alloy) are stable in these alloys and are not changed in this dose-temperature irradiation range.

Thus, it may be concluded that the DS alloys are an appropriate system for the analysis of a change in  $\rho$  under irradiation. The observed effects are to be determined mainly only by two processes: the transmutation and accumulation of radiation defects. The first of these effects is analyzed, i.e., the impact of transmutation on a change in electrical resistivity of copper alloys. As mentioned above, transmutation products are accumulated in copper under irradiation. Ni and Zn, when accumulated even in extremely small amounts of  $\approx 0.05\%$ , may decrease noticeably the electrical resistivity of copper. To take into account the effect of accumulated Ni and Zn atoms on the electrical resistivity use can be made of the formula [1,15]

$$\Delta \rho_{\text{trans}} = K_{\text{Ni}} C_{\text{Ni}} + K_{\text{Zn}} C_{\text{Zn}}, \qquad (1)$$
  
where

$$K_{\rm Ni} = 11.2 {\rm n}\Omega {\rm m}, \ K_{\rm Zn} = 3.0 {\rm n}\Omega {\rm m}.$$
 (2)

As follows from Eq. (1), with Ni amounting to 0.1 at.% and Zn to 0.1 at.%, the electrical resistivity of copper is increased by  $\Delta \rho_{\text{trans}} = 1.42 \text{ n}\Omega \text{ m}$  ( $\rho_0 = 17.0 \text{ n}\Omega \text{ m}$ ), 79% of the increase being attributable to Ni and 21% to Zn. The main reason for Ni accumulation in copper in thermal neutron reactors is the reaction

$$Cu^{63}(\mathbf{n},\gamma) \rightarrow Cu^{64} \xrightarrow{Cu^{64}(\beta^{+}) \rightarrow Ni^{64}}_{Cu^{64}(\beta^{-}) \rightarrow Zn^{64}} (3)$$

For thermal neutrons the cross-section of this reaction is extremely high and ranges up to 4 barn [16]. As for fast neutrons, this cross-section is extremely small, i.e.,  $3.53 \times 10^{-27}$  cm<sup>-2</sup> (0.0035 barn). Thus, at a dose of  $10^{26}$  n/m<sup>2</sup> fast neutrons accumulated in copper are extremely small in number, i.e.,  $\approx 0.22 \times 10^{-3}$  at.% Ni and cannot affect materially the electrical resistivity.

materially the electrical resistivity. In fast reactors the reaction  ${}^{63}$ Cu(n, p) with a rather high cross-section of  $\approx 6 \times 10^{-26}$  cm<sup>-2</sup> (0.06 barn) for fast neutrons can result in an accumulation of up to 0.06% Ni, when irradiated to a dose of  $10^{26}$  n/m<sup>2</sup>. The reaction on  ${}^{65}Cu(n, \gamma) - {}^{66}Cu - {}^{66}Zn$  with a cross-section of  $\approx 0.0005$ barn may also make a certain contribution. The calculation of transmutation rate in copper for the reactor FFTF [17] gives, in particular, the assessment of  $C_{\rm Ni} = 0.41$  at.% and  $C_{Zn} = 0.4$  at.% for 300 effective days, this corresponding to a dose of  $10^{27}$  n/m<sup>2</sup>. Note, that the calculations give only the lower boundary of Ni and Zn accumulation. This is associated with that in a real situation the fast reactors always have some non-zero fluxes of thermal neutrons, the so-called thermal tails, which may increase by 20-30% the accumulation of transmutants. Even with the ratio  $Ft_{fast}/Ft_{thermal} \approx 1000$  at a fast neutron dose of  $10^{26} \text{ n/m}^2$ the thermal neutron flux amounts to  $10^{23}$  n/m<sup>2</sup>, this being enough to affect strongly the accumulation of transmutation products, as the cross-section of Ni and Zn accumulation for the reaction  ${}^{63}Cu(n, \gamma)$  is very high. Therefore, to assess the contribution of transmutation products to a change in  $\rho$  we preferred to use not the calculation results but the reported data on the measurements of the amounts of accumulated transmutants in irradiated copper alloys.

The investigations of Cu–Cr–Zr–Mg alloy specimens performed in [6] with the EDS analysis revealed that about 0.123 at.% Ni was accumulated in a specimen at a dose of  $1.5 \times 10^{26} \text{ n/m}^2$ . In study [2] the chemical analysis of pure copper specimens irradiated in EBR-II to a dose of  $1.8 \times 10^{26} \text{ n/m}^2$  was performed and the specimens were shown to contain 0.053% Ni and 0.087% Zn. The EDS method used to measure the Ni content in pure copper irradiated to  $1.69 \times 10^{27} \text{ n/m}^2$  in the FFTF reactor yielded  $(0.9 \pm 0.2) \text{ at.}\%$  Ni [18].

Based on these results an assessment is made of the increase in the electrical resistivity of copper alloy specimens irradiated in the reactors BOR-60, EBR-II, FFTF (in view of the transmutation rate in these reactors). When doing so we set rather arbitrarily  $C_{\text{Ni}} \approx C_{\text{Zn}}$  for BOR-60 and FFTF according to the calculations performed in [17]. This is of course an approximation, but it is to bear in mind that a two-fold error, when determining Zn, changes  $\Delta \rho_{\text{trans}}$  by not more than 20%.

As follows from Fig. 3, an increase in  $\Delta \rho_{\text{trans}}$  with the dose is the highest for BOR-60 and the least for EBR-II. The difference in the  $\Delta \rho_{\text{trans}}$  increase in these reactors can be explained by different spectrum forms. But undoubtedly, the main reason is the Ni and Zn accumulation on thermal neutrons caused by thermal tails whose intensity in these reactors differs essentially. One cannot but take into account the following important consideration. The accuracy of the Ni content assessment increases with the dose growth. Thus, the most reliable data are the results for FFTF, where a high concentration of  $\approx 1\%$  Ni was analyzed. The difference between the results for BOR-60, and in particular for EBR-II, and FFTF can be explained by an inaccuracy in the analysis of small amounts of (0.1-0.05) at.% Ni in irradiated specimens. Despite a relatively mod-



Fig. 3. Calculated effects of transmutation products (Ni and Zn) accumulation on the electrical resistivity change ( $\Delta \rho_{trans} = K_{Ni}C_{Ni} + K_{Zn}C_{Zn}$ ) for DS copper alloys irradiated in the fast neutron reactors – BOR-60, FFTF, EBR-II (calculations based on the results of experimental measurements of Ni concentration in irradiated specimens [2,6,19].

erate accuracy of these assessments it is possible to calculate the rate of  $\Delta \rho$  increase caused by transmutation and to assess an individual contribution of transmutation and radiation defects to the  $\Delta \rho$  increase.

Fig. 4 presents the assessments made for the same specimens, where the Ni content was measured. It is obvious that at doses of  $\simeq 10^{26} \text{ n/m}^2$  (when irradiated in

fast neutron reactors) the increase in  $\Delta \rho_{rad,def}$  is determined nearly equally both by radiation defects and transmutation. It was demonstrated in Ref. [15] that the increase in  $\Delta \rho$ , while under irradiation in the mixed spectrum reactor SM-2, rapidly reaches saturation and remains practically constant in the dose range of 1–3 dpa (at  $T_{irr} =$ 100°C).



Fig. 4. Effects of transmutation and radiation defects on the electrical resistivity change ( $\Delta \rho_{irr} = \Delta \rho_{trans} + \Delta \rho_{rad,def}$ .) of DS copper alloys MAGT-0.2, MAGT-0.05, Cu-5Mo irradiated in the BOR-60 reactor at 340-420°C.

In our case the data on irradiation in BOR-60 are not sufficient to build up the dose dependence of  $\Delta \rho_{\rm rad def}$ . But, when using the obtained calculated dependence  $\Delta \rho_{\text{trans}} = f(\text{Ft})$  for EBR-II and FFTF and BOR-60 and the experimental data on changes in  $\Delta \rho_{summ} = \Delta \rho_{rad,def} +$  $\Delta \rho_{\rm trans}$  obtained in the experiments on irradiation of DS copper alloys Al20 and Al60 in EBR-II [2] and GlidCop Al25 in FFTF [3] and MAGT 0.2 in BOR-60 [this work], it is possible to get a satisfactory dose dependence of  $\Delta \rho_{rad,def} \simeq f(Ft)$  (Fig. 5). As a whole, Fig. 5 demonstrates that in the dose range of  $0.4-25 \times 10^{26}$  n/m<sup>2</sup> ( $T_{irr} = 340-$ 450°C) the DS copper alloys have practically the same increase in  $\Delta \rho_{\rm rad, def}$  at a level of 0.8 n $\Omega$  m. This fact correlates well with the stability of the mechanical properties and the lack of swelling in DS alloys in this dose-temperature range. The TEM investigations [10,12-14] confirm that the alloy structure in this case remains rather stable, the phases and complexes of radiation defects are also changed but slightly with the irradiation dose increased in the dose range of  $(0.4-2.5) \times 10^{26}$  n/m<sup>2</sup>. Thus, the analysis performed allows also the conclusion to be made that judging by the stability of  $\Delta \rho_{rad,def}$  (for which the radiation defects are responsible) the DS copper alloys are highly stable under irradiation up to doses of  $2.5 \times 10^{26}$  $n/m^2$  and retain effectively their high resistance to radiation damage.

## 4.2. The analysis of reasons for a change in electrical resistivity of copper alloys under irradiation in mixed spectrum reactor

The observed change in electrical resistivity of copper alloys under mixed spectrum reactor irradiation is determined by both parts of neutron spectra.

#### 4.2.1. Effect of fast neutrons on Ni and Zn accumulation

As for fast neutrons, the cross-section of Eq. (3) on <sup>63</sup>Cu is extremely small (0.0035 barn). The reaction <sup>63</sup>Cu(n, p) with a rather low cross-section of (0.06 barn) for fast neutrons can result in an accumulation of up to 0.03% Ni, when irradiated to a dose of  $5 \times 10^{25} \text{ n/m}^2$ . The reaction on <sup>65</sup>Cu(n,  $\gamma$ )  $\Rightarrow$  <sup>66</sup>Cu  $\Rightarrow$  <sup>66</sup>Zn (for fast neutrons) with a cross-section of  $\approx 0.0005$  barn cannot also make a certain contribution. Thus, at a dose of  $5 \times 10^{25} \text{ n/m}^2$  fast neutrons accumulated in copper are extremely small in number, i.e.,  $\approx 0.035$  at.% Ni and cannot affect  $\rho$ .

#### 4.2.2. Effect of thermal neutrons on Ni and Zn accumulation

In a mixed-spectrum reactor Eq. (3) (on  $^{63}$ Cu(n,  $\gamma$ )) for thermal neutrons makes the main contribution in the accumulation of transmutation products Ni and Zn. The crosssection of Eq. (3) is extremely high for thermal neutrons, i.e.,  $4.5 \times 10^{-24}$  cm<sup>-2</sup> (4.5 barn), for SM-2 reactor neutron spectrum [16]. Clearly for the mixed-spectrum reactor SM-2 (Channels 2–5), where the thermal neutron flux is appreciably high (Ft<sub>therm</sub>/Ft<sub>fast</sub> = 1), this reaction will result in a substantial accumulation of Ni and Zn ( $\approx 0.27$ at.% Ni at 1 dpa). Eq. (4) (for thermal neutrons) will also contribute to Zn accumulation:

$${}^{65}\mathrm{Cu}(\mathrm{n},\gamma) \to {}^{66}\mathrm{Cu}(\beta^{-}) \to {}^{66}\mathrm{Zn}.$$
(4)

When taking into account both Eqs. (3) and (4) and calculating the gain in electrical resistivity ( $\Delta \rho_{\text{trans}} = K_{\text{Ni}}C_{\text{Ni}} + K_{\text{Zn}}C_{\text{Zn}}$ ) caused by the transmutations we obtain, in accordance with the procedure described in Ref. [15], that for a thermal neutron dose of  $10^{25} \text{ n/m}^2$  Eq. (1) yields the value  $\Delta \rho_{\text{transm}} = 2.73 \text{ n}\Omega$  m. But the experiment [9] on the spectral tailoring in the SM-2 reactor channels, where the thermal neutron flux on samples was varied by a



Fig. 5. Dose dependence of the radiation defect part ( $\Delta \rho_{rad,det} = \Delta \rho_{irr} - \Delta \rho_{trans}$ ) of the electrical resistivity change of DS copper alloys irradiated in the fast neutron reactors BOR-60, FFTF, EBR-II at 340-450°C.

Cd filter in wide ranges, allowed for the assessment of the phenomenological dependence of  $\Delta \rho_{\text{trans}}$  on (Ft)<sub>thermal</sub>.

As in our study we used for irradiation purposes Channel 3 and Channel 5, that are close in their characteristics to Channel 4 in [15], precisely this dependence will be employed to calculate the contribution of the transmutants component:

$$\Delta \rho_{\rm trans} = 3.5 \, {\rm n} \, \Omega \, \, {\rm m} \, ({\rm Ft})_{\rm thermal norm} \,, \tag{5}$$

where  $(Ft)_{thermalnorm}$  is the thermal neutrons fluence  $(10^{25} n/m^2)$ .

Measurements of Ni amounts in irradiated samples made it possible to verify the correctness of such an approach. Fig. 6 presents the results of experimental measurements of the Ni content in irradiated samples depending on the thermal neutron fluence thereon and the linear dependence of Ni accumulation calculated by Eq. (5).

It is obvious that, as predicted by the calculations, the Ni concentration in irradiated samples is increased with the growth of the thermal flux on a sample. Evidently, the results of the measurements of Ni contents in irradiated samples systematically produce higher values than those predicted by the calculations. The main reason, in our opinion, is that the Ni concentration was not accurately enough estimated by the EDS analysis with small Ni contents (0.2–0.4 at.%). Besides, the discrepancies can be caused also by inaccurate assessments of the thermal flux on a sample and the values of the reaction cross-section on  $^{63}$ Cu for the SM-2 reactor spectrum. Still, on the whole, the correlation between the calculations and experimental results seems to be satisfactory.

As demonstrated in our previous studies [6-8], the PH alloys are characterized by a dramatic change in the struc-

ture in the irradiation temperature range of 300–400°C caused by the processes of polygonization and recrystallization and coarsening of Cr phases, which develop at these irradiation temperatures [8]. This change in the structure results in a change the component  $\Delta \rho_{rad,def} = \Delta \rho_{total}$  $-\Delta \rho_{trans}$  in a rather complicated way. Based on this study on change in  $\rho$  of the PH and DS copper alloys irradiated in BOR-60 reactor, the conclusion was substantiated in Section 4.1 that dispersion-strengthened (DS) copper alloys are the most convenient ones for the analysis of the mechanism of changes in electrical resistance when under irradiation.

Actually, as follows from Fig. 7, the DS Cu-Mo alloys have practically the linear gain  $\Delta \rho - f(Ft)$  at  $T_{irr} = 370 - f(Ft)$ 430°C. The calculation of  $\Delta \rho$  gain by Eqs. (1) and (2) yields a good correlation with the experiment. This agrees well with the fact that the structure of Mo disperse particles in Cu-Mo alloys remains stable throughout the dosetemperature irradiation range. The analysis of mechanical properties of Cu-Mo alloy performed in studies [6,8] revealed that the yield strength of Cu-Mo samples does not practically change at  $T_{\rm irr} \simeq 370-430^{\circ}$ C. Complexes of radiation defects are lacking in the structure as well. As a consequence, the component  $\Delta \rho_{rad,def} \approx 0$  and  $\Delta \rho_{total} =$  $\Delta \rho_{\text{trans}}$ . The matter is quite different for Cu-Cr-Zr, Cu-Cr-Zr-Mg alloys. As seen from Fig. 8, they are characterized by a dramatic spread in data at a high-dose irradiation testifying and indicating that along with the transmutation other processes also contribute to a change in  $\Delta \rho_{\text{total}}$ .

In order to understand the character of effects observed for the PH alloys we built up the dependence (Fig. 9) of a change in the density of irradiated samples of copper alloys on their residual gain in electrical resistance



Fig. 6. Measured and calculated nickel concentration vs. thermal neutron fluence of the PH copper alloy Cu-Cr-Zr-Mg irradiated in the SM-2 reactor at 400-430°C up to 0.6-0.8 dpa.



Fig. 7. Effect of transmutants accumulation on electrical resistivity change ( $\Delta \rho_{\text{trans}} = K_{\text{Ni}}C_{\text{Ni}} + K_{Zn}C_{Zn}$ ) of the DS copper alloy Cu–5Mo irradiated in the SM-2 reactor at 375–430°C up to 0.8–3.5 dpa.

 $\Delta \rho_{\text{residual}} = \Delta \rho_{\text{total}} - \Delta \rho_{\text{transm}}$ . We consider that  $\Delta \rho_{\text{residual}}$  is not related to the accumulation of radiation defects, because at high  $T_{\text{irr}} \approx (300-400^{\circ}\text{C})$  the complexes of radiation defects, if any, are large-sized and have a low density. That is why they cannot essentially contribute to the gain in  $\rho$ .

In this study it is shown that for large doses of  $\approx 10$  dpa (irradiation in BOR-60 reactor) and at the same temperatures the gain in  $\Delta \rho$  not associated with the transmutation is not large and does not exceed  $\approx 1 \text{ n}\Omega \text{ m}$ . After

irradiation in SM-2 up to 3 dpa the value of residual gain in the electrical resistance  $\Delta \rho_{\text{resist}}$  can be as high as  $\approx 10$ n $\Omega$  m. So this gain cannot be associated with radiation defects.

The data presented in Fig. 9 allow, at a first glance, the conclusion that the gain in  $\Delta \rho$  at high irradiation doses is related to swelling. It is in the samples with a maximum density drop of 3.2% that the maximum gain in electrical resistance is observed. But, the assessments made in studies [4,5] testify that at this small swelling (0.1–3.2%), at



Fig. 8. Effect of transmutants accumulation on electrical resistivity change ( $\Delta \rho_{trr} = \Delta \rho_{trans} + \Delta \rho_{rad,def}$ ) of PH copper alloys Cu–Cr–Zr. Cu–Cr–Zr–Mg irradiated in the SM-2 reactor at 185–400°C up to 2.8–3.5 dpa.



Fig. 9. Correlation between density change and residual electrical resistivity change ( $\Delta \rho_{\text{residual}} = \Delta \rho_{\text{irr}} - \Delta \rho_{\text{trans}}$ ) of PH copper alloys irradiated in the SM-2 reactor at 310-400°C up to 2.6-3.8 dpa.

which pores are small and their density is not sufficiently large, a change in electrical resistivity caused by pores cannot exceed 5-10%. In our case it amounts to 50%. Hence, this assumption is also false.

Still, we believe that all available experimental data allow us to conclude that in PH alloys of Cu-Cr-Zr, Cu-Cr-Zr-Mg type there occurs a spinodal decay at increased irradiation temperatures. The matter is that at these doses about 1% Ni and 1% Zn are accumulated in the alloys. As a consequence, we have a Cu-1% Ni-1% Zn-0.6% Cr-0.02% Zr alloy rather than Cu-0.6% Cr-0.2% Zr. Naturally, this alloy has a quite different phase diagram. The data on the spinodal decay of Cu-Ni-Sn alloys, when under irradiation, can be found in the literature [19]. In the material under irradiation we have a massive transformation with the formation of partially ordered structures is supported by the fact that, in alloys with the maximum growth in  $\Delta \rho_{\text{residual}}$ , a slight strengthening is observed at  $T_{\text{test}} \simeq 20-100^{\circ}$ C. At high  $T_{\text{test}} = 300-400^{\circ}$ C, strengthening in irradiated samples is practically lacking.

The precise answer to this question requires that the phase stability of the Cu-Ni-Zn-Cr-Zr alloy in its initial state has to be studied. Besides, the detailed TEM investi-



Fig. 10. Comparison of the transmutation part of the electrical resistivity change  $(\Delta \rho_{trans} = K_{Ni}C_{Ni} + K_{Zn}C_{Zn})$  of the DS copper alloy Cu-5Mo irradiated in the fast neutron reactor BOR-60 and in the mixed spectrum reactor SM-2 at 375-430°C.

gations of the structure of the samples irradiated to large doses in mixed-spectrum reactors are required.

The investigations performed make it possible as well to compare the gain rate of electrical resistance in copper alloys in different reactors. When neglecting the contribution of all other effects (this being rather true for DS alloys) and comparing the dependence  $\Delta \rho - f(dpa)$  for the BOR-60 and SM-2 reactors it becomes evident that both the experiment and the calculation demonstrate that in SM-2 the gain rate of  $\Delta \rho$  is twenty times higher (Fig. 10).

In our opinion, the obtained result is of practical importance. If in the primary ITER spectrum thermal neutrons are practically lacking, thermal neutron fluxes will be quite appreciable in some components of the first wall and divertor owing to retarding in the blanket. It is not essential to the divertor as a replaceable unit, but for the first wall, for which the life time dose would exceed  $\approx 4$  dpa, the presence of thermal neutrons in some areas can result in a two-fold drop of the thermal conductivity. This will lead to a rise in the temperature gradient in the heat sink system and an undesirable increase in temperature growth in the upper part of a copper tile. Its overheating can give rise to cracks owing to helium embrittlement ( $\approx 10$  appm helium at 1 dpa) and thermal fatigue.

From this standpoint the DS copper alloys seem to be more preferable candidate materials for the heat sink system of the first wall. Determining factors are the stability of their structure, a less drop in electrical and thermal conductivity (as for the DS alloys it is determined only by transmutation). Besides, the DS alloys resist well to hightemperature helium embrittlement due to high stability of the structure and fine strengthening particles.

#### 5. Conclusion

The presented experimental data on a change in the electrical resistivity of DS-type copper alloys irradiated in the BOR-60 reactor show that irradiation to 7–10 dpa at  $T = 340-450^{\circ}$ C causes a drop in electrical resistivity by not more than 20%. Based on these data and on the experimental results in EBR-II and FFTF the analysis of the effect of transmutation and radiation defect complexes on the increase in  $\rho$  in irradiated DS alloys made possible the conclusion that at doses of  $10^{26}$  n/m<sup>2</sup> an increase in  $\rho$  is 50% determined by the transmutation effects and 50% by the complexes of radiation defects.

The analysis of the dose dependence  $\Delta \rho_{\text{rad,def}} \sim f(\text{Ft})$ reveals that in the dose range of 3–16 dpa the DS copper alloys have practically the stable level of  $\Delta \rho_{\text{rad,def}}$ , this being associated with the stabilization of the produced structure of radiation complexes, this, in its turn, is determined by defect recombination on the surface of fine particles (Al<sub>2</sub>O<sub>3</sub>, Mo). The recombination of radiation defects on the particle surface reduces the defect flux on the complexes and makes the DS structure of DS copper alloys slightly sensitive to the radiation damage. The investigation undertaken revealed that a change in electrical conductivity of a DS copper alloy of Cu–Mo type irradiated in a mixed-spectrum reactor depends on the thermal flux on a sample. The calculations show that the experimentally measured gain in  $\Delta \rho_{\text{total}}$  agrees well with the calculated value of  $\Delta \rho_{\text{transm}}$ . Hence, a change in electrical conductivity is determined mainly by the accumulation of transmutation products Ni and Zn.

For PH copper alloys at increased  $T_{\rm irr} \approx 300-400^{\circ}$ C,  $\Delta \rho_{\rm total}$  is affected along with the transmutation by the structural and phase transformations. The correlation observed in a change in  $\Delta \rho$  and density of samples allows for the conclusion that the processes of spinodal decay proceed in the alloys of the Cu-Cr-Zr, Cu-Cr-Zr-Mg system irradiated to doses of  $\approx 3$  dpa at  $T_{\rm irr} \approx 310-400^{\circ}$ C. The obtained results show that the rate of  $\Delta \rho$  gain with a dose in mixed-spectrum reactors is about 20 times as high as in fast neutron reactors.

The conclusion is made that the calculations made for ITER must take into account the presence of appreciable fluxes of thermal neutrons in certain components of the reactor. The latter will play a decisive role in the drop of the thermal conductivity of copper alloys in these components.

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