# 6.2 Thermal Conductivity of Beryllium under Low Temperature High Dose Neutron Irradiation

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Thermal conductivity of compact beryllium of several Russian grades such as TE-400, TE-56, TE-30, TIP and DIP differing in the production technology, grain size and impurity content has been investigated. The thermal diffusivity of beryllium was measured on the disks in the initial and irradiated conditions using the pulse method in the range from room temperature to 200$^0$C. The thermal conductivity was calculated using the table values for the beryllium thermal capacity. The specimens and beryllium neutron source fragments were irradiated in the SM reactor at 70$^0$C and 200$^0$C to a neutron fluence of $(0.5\text{-}11.4)\cdot 10^{22}$ cm$^{-2}$ (E>0.1MeV) and in the BOR-60 reactor at 400$^0$C to $16\cdot 10^{22}$ cm$^{-2}$ (E>0.1MeV), respectively. The low-temperature irradiation leads to the drop decrease of the beryllium thermal conductivity and the effect depends on the irradiation parameters. The paper analyses the effect of irradiation parameters (temperature, neutron fluence), measurement temperature and structural factors on beryllium conductivity. The experiments have revealed that the short time post-irradiation annealing at high temperatures results in partial reduction of the thermal conductivity of irradiated beryllium.

## 1. INTRODUCTION

Thermal conductivity is a major characteristic of the structural material used under neutron irradiation conditions as it significantly effects the operating temperature of the relevant fission or fusion reactor unit. At present beryllium is used as a reflector and moderator material in nuclear research reactors. It is known [1] that the operating temperature of beryllium blocks in these reactors ranges from 70$^0$C to 200$^0$C therefore the results provided in the paper are primarily important for validation of radiation resistance of beryllium under these conditions. It is supposed that beryllium will be used as a neutron breeding material in the DEMO fusion reactor blanket in the temperature range from 100$^0$C to 800$^0$C. Therefore investigation of the regularities of the thermal-physical properties behavior for beryllium depending on the neutron irradiation parameters in the low-temperature range in the given temperature range is an urgent fusion problem too.

## 2. MATERIALS, SPECIMENS, EXPERIMENT

Beryllium of several Russian grades the chemical composition of which is presented in Table 1 has been experimentally studied. This is a hot-pressed beryllium of the outdated grades TE-400, TE-56 and TE-30 produced by hot extrusion (HE) and of grades TIP and DIP produced by hot isostatic pressing (HIP). Specimens in the form of discs of 5.9-6.0 mm in diameter and 2-3 mm thick made remotely by mechanical treatment of the cylindrical tensile specimen heads were employed to study thermal conductivity.

Table 1
Chemical composition and grain size of beryllium grades

| Grade | Technology | Middle grain size, μm | Chemical composition, % mass | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | Be | BeO | O | Fe | Al | Si | C | Mg |
| TE-56 | HE | 25 | 98.6 | 1.48 | 0.98 | 0.17 | 0.026 | 0.016 | 0.08 | н/д |
| TE-30 | HE | 15 | 98.1 | 2.5 | 1.66 | 0.11 | 0.015 | 0.013 | 0.088 | 0.002 |
| TIP | HIP | 12 | 98.8 | 1.3 | 0.89 | 0.13 | 0.013 | 0.013 | 0.07 | 0.0066 |
| DIP | GIP | 13 | 98.6 | 2.0 | 1.3 | 0.03 | 0.005 | 0.013 | 0.067 | 0.0016 |
| TE-400 | HP | ~100 | not determined | | | | | | | |

The main body of information on thermal conductivity of irradiated beryllium was obtained under investigation of the tensile specimens irradiated in the SM reactor at 70°C and 200°C into special irradiation capsules. Specimens to measure thermal conductivity of the TE-400 beryllium grade were cut out remotely from the neutron source fragments irradiated at 400°C in the BOR-60 reactor. Neutron doses of specimens are given in the Figures included in the paper. In the initial state the discs of the above dimensions were made from the rods of the corresponding beryllium grades by mechanical treatment.

The specific thermal conductivity factor was obtained by the method described in paper [2]. Measurement of the beryllium thermal diffusivity in the initial and irradiated condition was performed by the pulse method in the range from room temperature to 200°C in vacuum no less than $10^{-3}$ Torr. The density of initial and irradiated specimens was measured hydrostatically. The thermal conductivity was calculated using the table values for the beryllium thermal capacity.

## 3. EXPERIMENTAL RESULTS

In the initial state the thermal conductivity values for the beryllium grades under testing significantly differ between each other (Fig.1). The TE-56 beryllium grade possesses the best properties and TE-400 – the worst ones; the rest of the grades take the intermediate position. As the testing temperature raises from room temperature to 200° C, thermal conductivity of all grades decreases. The maximum temperature dependence is characteristic for the TE-56 beryllium grade where the difference of values at room temperature and at 200°C is about 50 W/m·K. The Figure illustrates the investigation of specimens cut out along the extrusion axis (for the anisotropic grades, i.e. produced by the HE method). For the relevant specimens cut out across axis the thermal conductivity values are, as a rule, lower by 10-15% [3].

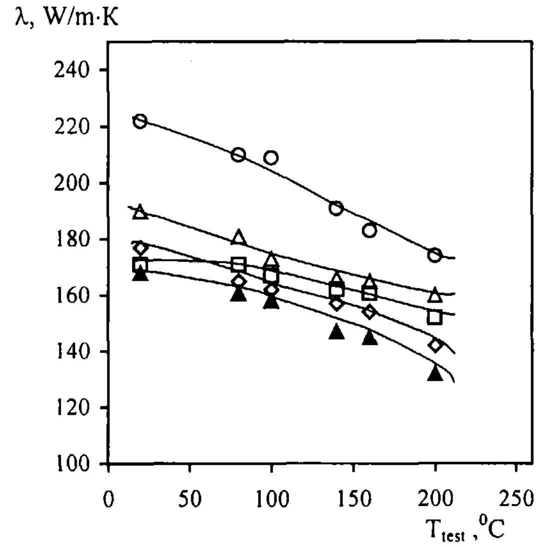

Figure 1. Temperature dependence of thermal conductivity in the initial state of various beryllium grades, along the axis:
o - TE-56
◊ - TE-30
▲ - TE-400
□ - TIP
Δ – DIP

First let's consider the effect of the irradiation parameters on thermal conductivity of the TE-56 beryllium grade, which is now the most widely used as a reflector and moderator material in nuclear research reactors. The neutron irradiation at 70°C to the neutron fluence of $2{\cdot}10^{22}$ cm$^{-2}$ (E>0.1 MeV) causes the sharp decrease of thermal conductivity of this beryllium grade (Fig.2). In our case it dropped from 210 to 46 W/m·K, i.e. by about a factor of five. The dependence of beryllium thermal conductivity on the testing temperature that was observed in the initial state disappeared, i.e. actually the thermal conductivity value in the range of room temperature to 200°C is kept at about the same level during the measurements.

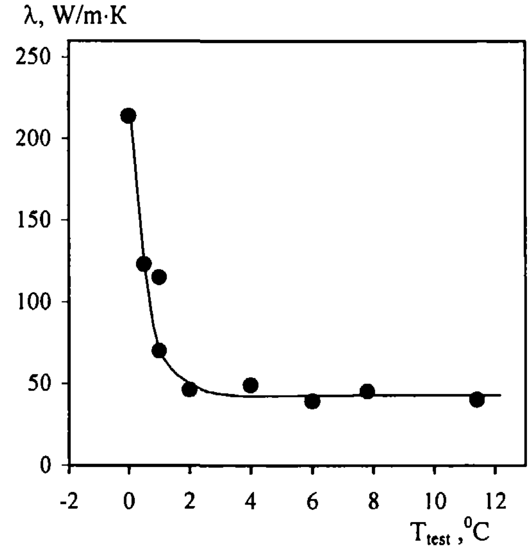

Figure 3. Dose dependence of thermal conductivity of the TE-56 beryllium grade, along the axis at $T_{irr}=T_{test}=70$°C

Dependence of thermal conductivity of the TE-56 beryllium grade on the irradiation dose at 70°C is shown in Fig.3. The sharp drop of the specific thermal conductivity factor occurs only in the interval of doses from zero to $2{\cdot}10^{22}$ cm$^{-2}$ (E>0.1 MeV). Further increase of the neutron dose up to $11.4{\cdot}10^{22}$ cm$^{-2}$ (E>0.1MeV) does not lead to thermal conductivity decrease and it remains in the range of 40-50 W/m·K.

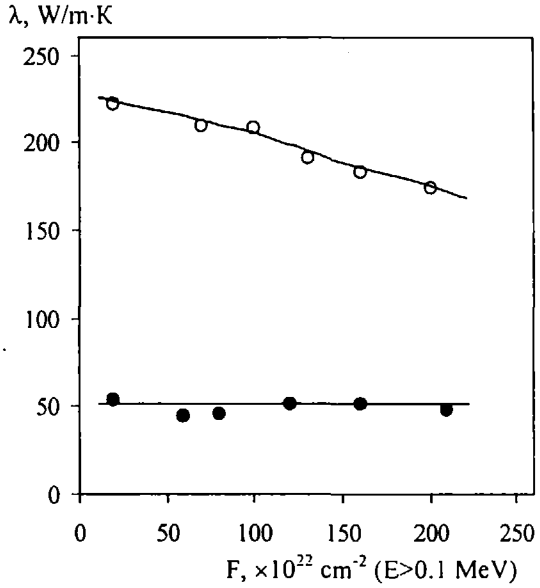

Figure 2. Temperature dependence of thermal conductivity of the TE-56 beryllium grade along the axis in the initial state (○) and after irradiation at 70°C up to fluence of $2\times10^{22}$ cm$^{-2}$ (E>0.1 MeV) (•)

The comparison of the thermal conductivity values for the TE-56 beryllium grade obtained for different irradiation temperatures (70°C and 200°C) at about comparable irradiation doses is presented in Fig.4. As evident from the Figure, thermal conductivity of specimens irradiated at a higher temperature slightly decreases in spite of a higher value of the accumulated neutron fluence. The comparison of the measurement results of the irradiated specimens cut out along and across the extrusion axis indicates to insignificant difference between them. Although it is necessary to pay attention to some instability of results at a dose of $1{\cdot}10^{22}$ cm$^{-2}$ (E>0.1 MeV) as thermal conductivity of the TE-56 beryllium grade changes most significantly somewhere at this level.

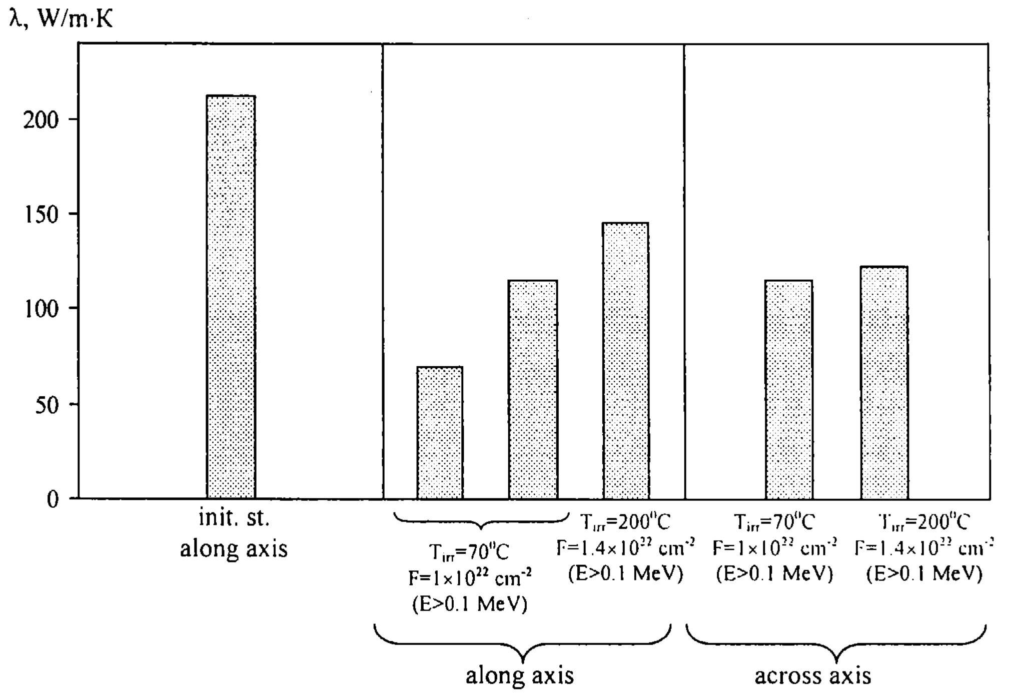

Figure 4. Influence of irradiation temperature on thermal conductivity of the TE-56 beryllium grade at compared neutron doses ($T_{test}$=70$^0$C)

The effect of the irradiation temperature on thermal conductivity of beryllium of other grades is shown in Fig.5-7. It should be noted that some of these results need to be checked as they were obtained from rather limited number of specimens. For instance, in the initial state thermal conductivity of specimens of the TE-30 beryllium grade cut out across the axis is a little higher than that of the longitudinal specimens (Fig.5). For transverse specimens of the same beryllium grade the specific thermal conductivity factor at 70$^0$C and 200$^0$C is the same. But at 200$^0$C the specimens were irradiated to rather lower fluence, therefore thermal conductivity at a fluence of 2·$10^{22}$ $cm^{-2}$ (E>0.1 MeV) is likely to be higher.

The results of the thermal conductivity measurement for beryllium of the TIP and DIP grades, produced by the HIP method, are presented in Fig. 6. Thermal conductivity of the TIP beryllium grade irradiated at 200$^0$C is slightly higher than after irradiation at 70$^0$C. The comparison of the results for the TIP and DIP specimens after irradiation at 200$^0$C suggests that the values of the specific thermal conductivity factor are very close.

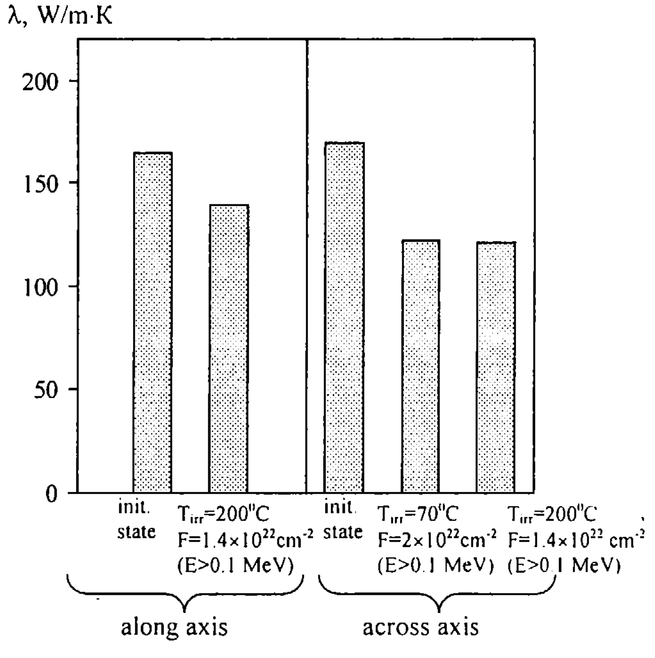

Figure 5. Influence of irradiation temperature on thermal conductivity of the TE-30 beryllium grade at compared neutron doses ($T_{test}$=70$^0$C)

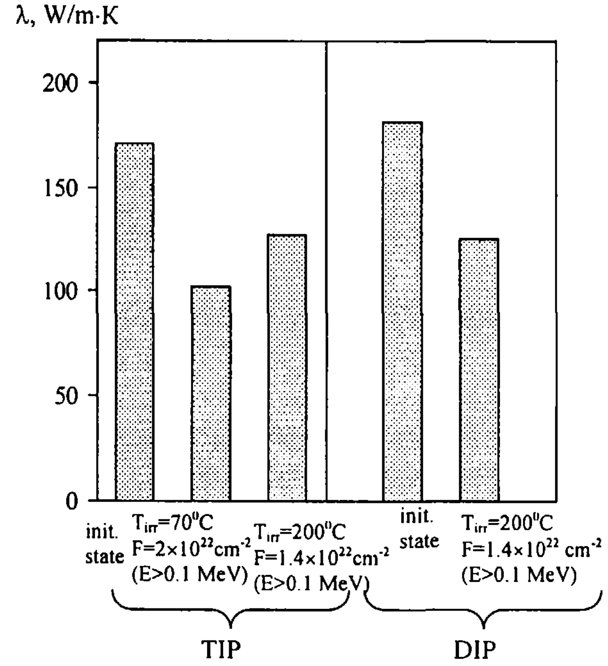

Figure 6. Influence of irradiation temperature on thermal conductivity of the TIP and DIP beryllium grades at compared neutron doses ($T_{test}$=70$^0$C)

Assessment of the effect of the irradiation temperature on thermal conductivity of beryllium irradiated at 70$^0$C and 400$^0$C at maximum neutron doses is provided in Fig.7. The measurement in both cases was made at 70$^0$C. As mentioned above, in the initial state the out-of-date TE-400 beryllium grade has the worst thermal conductivity compared to other grades. However the high dose irradiation at 400$^0$C leads to its decrease by about 15% while the similar (even a little lower) radiation dose for the TE-56 grade at 70$^0$C leads to decrease of thermal conductivity by a factor of higher than four. The TE-400 and TE-56 beryllium grades were irradiated in different reactors (BOR-60 and SM, respectively) but this is not of fundamental importance because in both cases the neutron dose was calculated for fast neutrons with the energy of more than 0.1 MeV which have the dominant role in the radiation damage of beryllium. Therefore the sharp inverse dependence of the effect of the thermal conductivity decrease on the irradiation temperature in the range of 70-400$^0$C is beyond question.

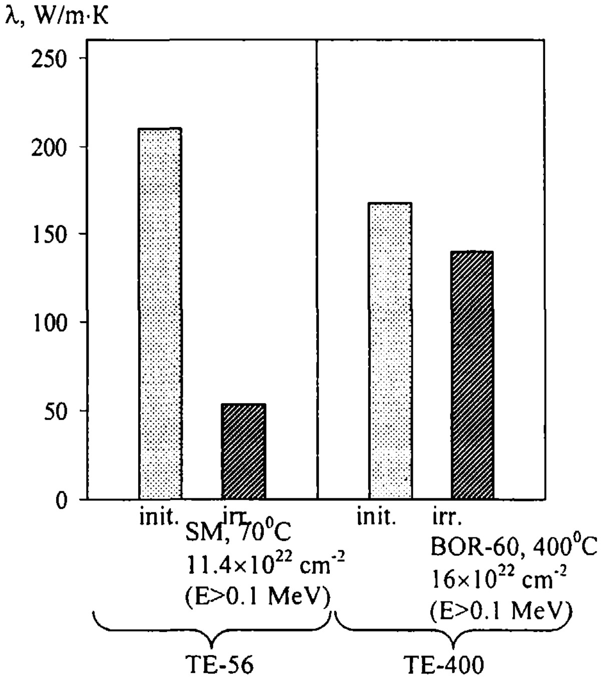

Figure 7. Influence of irradiation temperature at maximal neutron dose on thermal conductivity of the TE-56 and TE-400 beryllium grades, along axis

Fig. 8 provides the thermal conductivity factors for different beryllium grades after irradiation at two temperatures and comparable neutron doses. The comparison shows that at 70$^0$C the minimum factor of the TE-56, TE-30 and TIP grades do not much differ between each other. At 200$^0$C it's just opposite, the maximum factor is observed for the TE-56, the thermal conductivity factor of the TE-30 is close to it and the TIP and DIP factors are slightly lower.

Paper [4] describes an investigation of the effect of short-term high-temperature annealing on swelling of irradiated beryllium. It was revealed that the significant gas swelling growth starts from 600°C. Therefore in order to study the effect of annealing on thermal conductivity of the TE-56 beryllium grade, it was subjected to annealing at 500°C when the dimensional changes of specimens are insignificant. It's obvious from the results provided in Fig.9 that annealing leads to partial recovery of the specific thermal conductivity factor of irradiated beryllium and quantitatively this effect is no less than 150%.

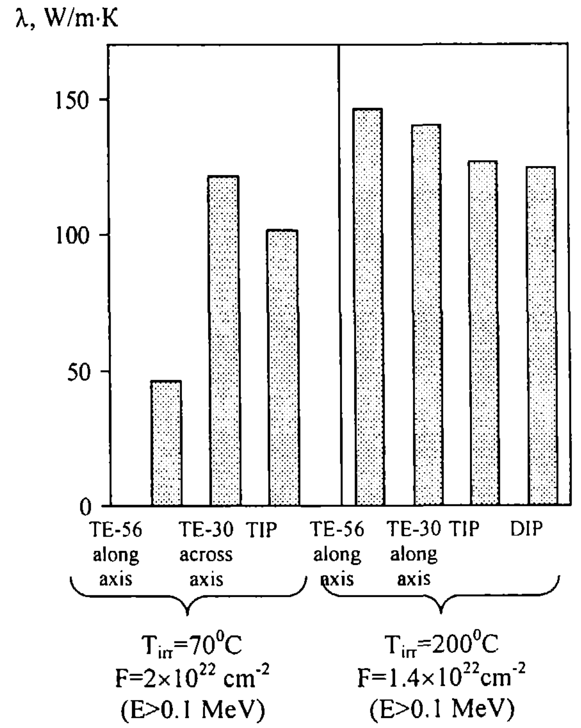

Figure 8. Comparison of the specific thermal conductivity factor of investigated beryllium grades after irradiation ($T_{test}$=70$^0$C)

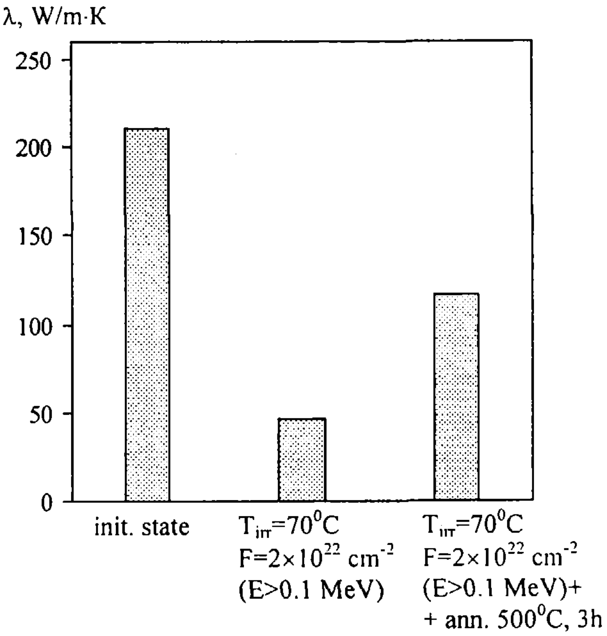

Figure 9. Influence of annealing at 500$^0$C, 3 h on thermal conductivity of the irradiated TE-56 beryllium grade, along axis ($T_{test}$=70$^0$C)

## 4. DISCUSSION

Neutron irradiation of beryllium causes damage of microstructure. These are basically radiation defects of the dislocation loops type and accumulation of significant amount of gas atoms, mainly helium ($^4$He isotope) from nuclear reactions [5]. The radiation-induced changes of thermal conductivity depending on the irradiation temperature and dose given above are caused by the change of beryllium microstructure under irradiation as the appearance of the additional electron density scattering centers in metal decreases its thermal conductive properties [6].

The dependence of transmutated helium quantity in beryllium on the neutron dose obtained from calculations and validated experimentally for the TE-56 grade is a linear dependence (Fig.10) [7].

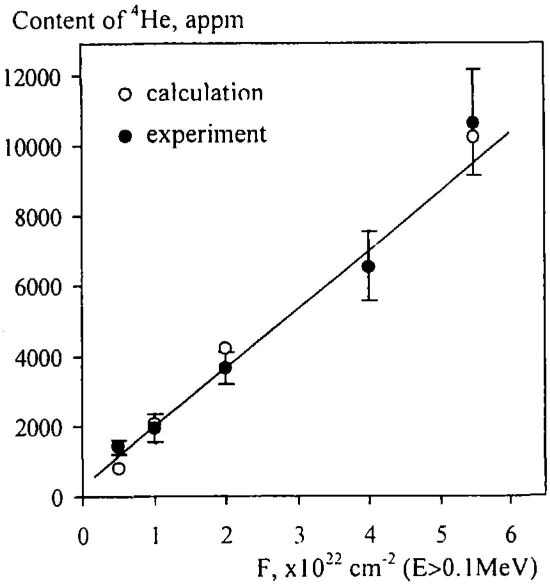

Figure 10. Dependence of transmuted helium content in the TE-56 beryllium grade on neutron dose

Superposition of this dependence on the dose dependence of the specific thermal conductivity factor of beryllium (Fig.3) shows no correlation between these two dose dependences. Sharp drop in thermal conductivity occurs in the neutron doses up to $2\cdot10^{22}$ cm$^{-2}$ (E>0.1 MeV) when the maximum content of helium in beryllium does not exceed 3000-3500 appm. An increase of transmutated helium quantity to 10000 appm (at $6{\cdot}10^{22}$ $cm^{-2}$ (E>0.1MeV)) does not cause any further decrease of thermal conductivity.

The TEM examination of beryllium microstructure shows that in the postirradiation state the main radiation defects are the dislocation loops of high volumetric density (Fig.11 (a)). Due to the low diffusion mobility of helium atoms at the low-temperature irradiation, the probability of their displacement and formation of helium atom clusters is not high in such conditions. Therefore most of the formed gas atoms remain in the region of the beryllium atom transmutated after entering the reaction. At low-temperature irradiation, helium atoms cause the so-called "solid" beryllium swelling that does not reach a significant value [8]. Dislocation loops of the interstitial type are formed in the radiation damage cascades at the elastic atom-atomic collisions. A tendency to increase the volumetric loop density is, as a rule, observed in metals with increasing the radiation dose [9]. Unfortunately, because of the strong brittleness of the irradiated discs we failed to perform the TEM examination of beryllium after high-dose neutron irradiation and to build the dose dependence of the formed dislocation loops density but there is no reason to subject to doubt the provisions of the paper [9]. Thus at the low-temperature neutron irradiation of beryllium, the helium and dislocation loops formation processes do not involve, in an explicit form, the explanations of the reasons for saturation of the specific thermal conductivity value after reaching the neutron fluence of $2{\cdot}10^{22}$ $cm^{-2}$ (E>0.1 MeV). Conceivably at high doses some superposition of these processes might occur with the radiation relaxation effect that decreases the efficiency of the newly formed scattering centers of conductivity electrons.

The short-term annealing of irradiated beryllium at $500^0$ C leads: first, to evolution of dislocation loops into the dislocation net (Fig.11 (b)); second, to the appearance of the first helium bubbles observed by TEM (Fig.11(c)) which, with increasing the annealing temperature to $700^0$C significantly increase in size (but consequently their volumetric density decreases) (Fig.11 (d)). As this takes place, the microstructural changes cause rather significant recovery of thermal conductivity.

High-dose irradiation at $400^0$C leads directly to formation of the gas pores of the plane form, which place only in certain crystallographic planes (Fig.10 (e, f)). Due to this particular structure the effect of thermal conductivity decrease of beryllium irradiated under these conditions is rather low. Probably rather large structural formations, such as helium bubbles or pores as well as dislocation net do not possess high efficiency as the scattering centers of conductivity electrons.

## 5. CONCLUSIONS

An investigation of the effect of neutron irradiation at $70\text{-}400^0$C in a neutron fluence range of $(0.5\text{-}16){\cdot}10^{22}$ $cm^{-2}$ (E>0.1 MeV) on thermal conductivity of beryllium of several Russian grades such as TE-400, TE-56, TE-30, TIP and DIP produced by hot extrusion and hot isostatic pressing methods has been undertaken. Thermal diffusivity of beryllium was measured in vacuum by pulse method from room temperature to $200^0$C. The results are as follows:

1.Neutron irradiation at $70^0$C to a neutron fluence of $2{\cdot}10^{22}$ $cm^{-2}$ (E>0.1 MeV) leads to sharp decrease of thermal conductivity of the TE-56 beryllium grade, in particular, at $70^0$C thermal conductivity decreased from 210 to 46 W/m·K, i.e. by about a factor of five. It should be noted that the thermal conductivity factor sharply decreased only in the dose range from zero to $2{\cdot}10^{22}$ $cm^{-2}$ (E>0.1 MeV). With increasing the neutron dose to $11.4{\cdot}10^{22}$ $cm^{-2}$ (E>0.1 MeV), thermal conductivity did not decrease any more.

2.In the temperature range of $70\text{-}400^0$C there is a reverse dependence of the decrease of beryllium thermal conductivity on temperature. The maximum effect of the decrease occurs at the minimum temperature and the minimum effect – at the maximum temperature in the given temperature range.

3.The comparison of the irradiation effect at $70^0$C and $200^0$C at the level of the neutron doses of $(1\text{-}2){\cdot}10^{22}$ $cm^{-2}$ (E>0.1 MeV) on thermal conductivity of the beryllium grades under study shows that there is no principle difference between them except the TE-56 grade, which has the minimum thermal conductivity factor after irradiation at $70^0$C.

4.Short-term high-temperature annealing ($500^0$C, 3 hours) leads to partial recovery of the thermal conductivity factor of the TE-56 beryllium grade irradiated at $70^0$C to $2{\cdot}10^{22}$ $cm^{-2}$ (E>0.1 MeV).

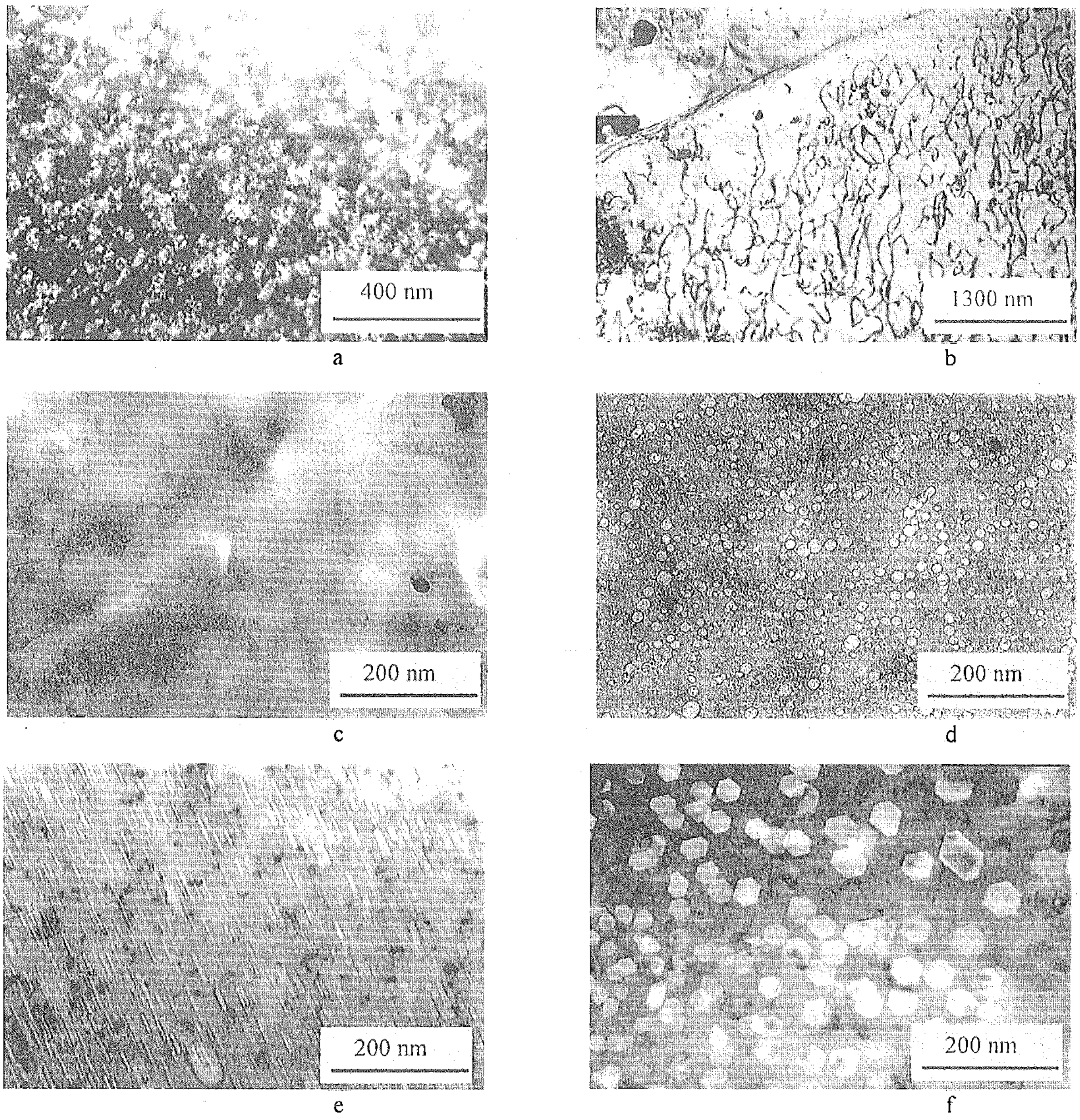

Figure 11. Influence of irradiation and postirradiation annealing on beryllium microstructure:
a) $T_{irr}=70^0$C, F=$2\cdot10^{22}$ cm$^{-2}$ (E>0.1MeV);
b) $T_{irr}=70^0$C, F=$2\cdot10^{22}$ cm$^{-2}$ (E>0.1MeV) + annealing at $500^0$C, 1 h; dislocation net;
c) $T_{irr}=70^0$C, F=$2\cdot10^{22}$ cm$^{-2}$ (E>0.1MeV) + annealing at $500^0$C, 1 h; helium bubbles;
d) $T_{irr}=70^0$C, F=$2\cdot10^{22}$ cm$^{-2}$ (E>0.1MeV) + annealing at $700^0$C, 1 h;
e,f)$T_{irr}=400^0$C, F=$16\cdot10^{22}$ cm$^{-2}$ (E>0.1MeV), plate helium pores, different crystallographic orientations

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