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Strengthening, loss of strength and embrittlement of beryllium under high temperature neutron irradiation

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Abstract

The 'dose' dependence of short-term mechanical properties of beryllium DIP-600 with 2.3 wt% of oxygen was determined. Irradiation temperature was 650°C, post-irradiation test temperature was 20°C. The influence of the main structural factors on ultimate strength of warm- and hot-pressed materials, characterized by oxygen content 0.7–4.9 wt%, grain size 8–50 μm, initial porosity 0.81–2.26% was revealed. The materials were irradiated to a fast fluence of 4.67×10^{21} n/cm^2 ($E \geqslant 0.85$ MeV) at 620°C and that of 5.69×10^{21} n/cm^2 ($E \geqslant 0.85$ MeV) at 680°C. © 1999 Elsevier Science B.V. All rights reserved.

1. 'Dose' dependence of ultimate strength variation. Irradiation temperature 650°C

It was reported in [1,2] that 'dose' dependencies of the short-term mechanical properties of beryllium under cryogenic and low temperature irradiation are qualitatively similar and fully dominated by two main competing processes:

- strengthening due to dislocation pinning by the defects generated by irradiation with $(\Delta\sigma_{\mathrm{us}}^{\mathrm{el}})_{\mathrm{str}} \sim (\Delta V/V_0)^{1/3}$, where $(\Delta\sigma_{\mathrm{us}}^{\mathrm{el}})_{\mathrm{str}}$ denotes strengthening in the dependence of tensile strength $\sigma_{\mathrm{us}}^{\mathrm{el}}$ versus swelling $\Delta V/V_0$ of the material;
- softening initiated by the occurrence and growth of stresses of 'radiation-swelling' anisotropy with $(\Delta\sigma_{\mathrm{us}}^{\mathrm{el}})_{\mathrm{soft}} \sim (\Delta V/V_0)^{1/3}$, where $(\Delta\sigma_{\mathrm{us}}^{\mathrm{el}})_{\mathrm{soft}}$ denoted strengthening in the dependence of tensile strength versus swelling of the material.

The processes, being similar in their consequences, though different in their physical origin, can proceed under high temperature irradiation conditions as well.

Thus, the material will be noticeably strengthened owing to gas bubble formation and dislocation pinning by these bubbles. There are reasons to forecast that $(\Delta\sigma_{\mathrm{us}})_{\mathrm{str}}$ will be proportional to $(\Delta V/V_0)^{1/6}$ or $F_{\mathrm{f}}^{1/4}$, where F_{f} is a fast neutron fluence ($E \geqslant 0.85$ MeV).

At the same time the material will be softened substantially due to 'flow-to-sink' of the bubbles from the near boundary areas to the grain boundaries as such. Here $(\Delta\sigma_{\mathrm{us}})_{\mathrm{soft}}$ will be proportional to $(\Delta V/V_0)^{2/3}$ or F_{f}, which is easy to find.

Hot-pressed Be powder of grain size <600 μm, containing 2.3 wt% O, was irradiated at 650°C to fluences of $(0; 0.6; 3.0; 5.7) \times 10^{21}$ n/cm^2 ($E \geqslant 0.85$ MeV) to verify and refine the above statements.

Compression tests were performed at 20°C in a remote-controlled MM-150D machine. The samples tested had a diameter of 6 mm and a height of 9 mm. Experimental values were determined by averaging the data from three or four independent tests.

The results obtained are shown in Fig. 1. As it was anticipated, the figure proves that:

- strengthening, softening and embrittlement are the main manifestations of beryllium radiation damageability in the region of Be bubble swelling;
- strengthening dominates at $F_{\mathrm{f}} < (F_{\mathrm{f}})_{\max} < 6 \times 10^{20}$ n/cm^2, $E \geqslant 0.85$ MeV $[\Delta V/V_0 < (\Delta V/V_0)_{\max} < 0.22\%]$; [1]

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[1] Here and after, swelling is calculated from the expression [3]: $\Delta V/V_0 = 1.17 \times 10^{-34}\, Te^{-2.1/(4kT)} F_{\mathrm{f}}^{\ 3/2}$.

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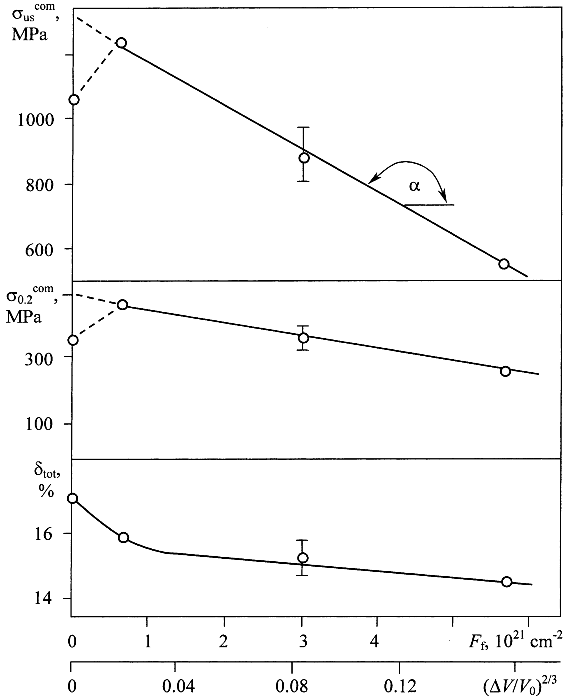

Fig. 1. Dose dependence of ultimate strength, σ_{us}, relative yield stress, $\sigma_{0.2}$, and general relative elongation, δ_{tot}, of hot-pressed (out of the powder of the size $\leqslant$ 600 μm) beryllium. $T_{irr} = 650°C$, $T_{test} = 20°C$.

$\mathrm{tg}\ \alpha = \Delta\sigma_{us}^{com}/\Delta F_f = -1.31 \times 10^{-19}$ MPa cm^2.

- the maximum attainable increment of ultimate strength $[(\sigma_{us}^{com})_{max} - (\sigma_{us}^{com})_0]$ does not exceed 17–23%. Here $\Delta\sigma_{us}^{com}$ is compression strength, the subscript '0' corresponds to an unirradiated material and the subscript 'max' denotes the maximum value for an irradiated material;
- softening prevails at $F_f > (F_f)_{max}$ and seems to continue until the material completely loses its 'load bearing capability';
- at the softening stage the dependencies $\sigma_{us}^{com} = f(F_f)$, $\sigma_{us}^{com} = f[(\Delta V/V_0)^{2/3}]$ are close to linear ones.

The obtained results combined with the above qualitative peculiarities of the expressions $(\Delta\sigma_{us})_{str}$, $(\Delta\sigma_{us})_{soft}$ allow to determine the shape of the dose dependence of Be ultimate strength $\Delta\sigma_{us}^{com}$ and reveal its consequences.

Actually, setting up different slopes of the decreasing dependencies of $(\Delta\sigma_{us}^{com})_{soft} \sim F_f$ and defining the relationships complementing them and verifying the latter for linearity in the coordinates $(\Delta\sigma_{us}^{com})_{str} - (\Delta V/V_0)^{1/6}$, one finds the dose dependence is described by the expression

$$\begin{aligned}\sigma_{us}^{com}\ [\mathrm{MPa}] &= (\sigma_{us}^{com})_0\ [\mathrm{MPa}] + 1.68 \\ &\quad \times 10^{-3}\ [\mathrm{MPa\ cm}^{1/2}] \cdot F_f^{1/4}\ [\mathrm{cm}^{-1/2}] \\ &\quad - 1.66 \times 10^{-19}\ [\mathrm{MPa\ cm}^2] \cdot F_f\ [\mathrm{cm}^{-2}] \\ &= (\sigma_{us}^{com})_0\ [\mathrm{MPa}] + 7.3 \times 10^2\ [\mathrm{MPa}] \\ &\quad \times (\Delta V/V_0)^{1/6} - 6.01 \times 10^3\ [\mathrm{MPa}] \\ &\quad \times (\Delta V/V_0)^{2/3}.\end{aligned}$$

While searching for an extremum in the above expression, we find that $(F_f)_{max} = 3.43 \times 10^{20}$ n/cm^2 ($E \geqslant 0.85$ MeV), $(\Delta V/V_0)_{max} = 0.09\%$, $(\sigma_{us}^{com})_{max} = 1253$ MPa, and setting its right terms equal to zero, we obtain $(F_f)^d = 1.02 \times 10^{22}$ n/cm^2 ($E \geqslant 0.85$ MeV), $(\Delta V/V_0)^d = 14.58\%$, where $(F_f)^d$ and $(\Delta V/V)^d$ correspond to fluence and swelling, respectively, which completely soften the material.

2. The dependence of ultimate strength on main structure factors. $T_{irr} = 620°C$ and 680°C

A correct choice of the most radiation resistant grades of Be is impossible without comparative investigations of a substantial number of commercial and laboratory modifications of Be. We have studied specially prepared warm and hot extruded (Tables 1 and 2) modifications. Radiation test parameters of experimental (rupture) samples were relevant to the values from Table 3.

Pre- and post-irradiation mechanical tests were performed at the temperatures of 20°C and 650°C. Experimental points were obtained by averaging the data obtained from individual tests of three or four identical samples. The results are shown in Figs. 2–6.

It can be seen that the main manifestations of Be radiation damageability there are again strengthening, softening and embrittlement of the material. The results lead to some other, less evident and therefore not formulated as yet, conclusions.

Thus, it follows from Figs. 2–6, that

- only the modifications with the $(\sigma_{us}^{el})_0 < 200$ MPa ($T_{test} = 650°C$) undergo radiation strengthening. The latter value is reduced with the increase of irradiation temperature and neutron fluence;
- the degree of softening increases with the initial strength. The dependencies of $\Delta\sigma_{us}^{el} = \sigma_{us}^{el} - (\sigma_{us}^{el})_0$ on $(\sigma_{us}^{el})_0$ have the form:
- $\Delta\sigma_{us}^{el}$ [MPa] $= -1.62(\sigma_{us}^{el})_0$ [MPa] + 333 for $T_{irr} = 620°C$,
- $F_f = 4.67 \times 10^{21}$ n/cm^2 ($E \geqslant 0.85$ MeV), $T_{test} = 650°C$,
- $\Delta\sigma_{us}^{el}$ [MPa] $= -1.80(\sigma_{us}^{el})_0$ [MPa] + 277 for $T_{irr} = 680°C$,
- $F_f = 5.69 \times 10^{21}$ n/cm^2 ($E \geqslant 0.85$ MeV), $T_{test} = 650°C$;

Table 1
Chemical content of Be modifications studied

Modification number	Element content, wt%							
	O	Fe	Ni	C	Al	Cr	Cu	Be
1	0.70	0.058	0.003	0.012	0.019	0.007	0.013	balance
2	1.75	0.071	0.003	0.012	0.029	0.009	0.015	balance
3	2.75	0.068	0.004	0.012	0.042	0.012	0.032	balance
4	3.10	0.024	–	0.010	0.050	0.070	0.014	balance
5	4.90	0.024	0.001	0.010	0.004	0.001	0.001	balance

Table 2
Oxygen content, density, porosity and grain size of Be modifications

Modification size number	Manufacturing technology	Oxygen content, C, wt%	Initial density, γ_0, g/cm^3	Porosity, P_0, %	Mean grain Size, d_g, μm
1	Hot-pressing + sintering	0.70	1.834	0.81	50
2	$T = 1160$–1180°C + warm	1.75	1.852	1.13	35
3	($T = 450$–500°C) extrusion with deformation 82%	2.75	1.846	1.75	20
4	Hot pressing	3.10	1.849	1.83	15
5	($T = 1170$–1180°C) ($P = 589$–638 MPa, $\tau = 2$ h) + hot ($T = 1000$°C) extrusion with deformation 82%	4.90	1.859	2.26	8

Table 3
Irradiaton parameters of samples

Modification number	Irradiation temperature (°C)	Exposure time		Neutron fluence ($E \geqslant 0.85$ MeV), 10^{21} n/cm^2
		Calendar days	Effective days	
1–3	680	245	233.8	5.69
3–4	620	245	233.8	4.67

- most softening is characteristic for the modifications with the values $P_0 \cdot d_g{}^2 = 35$–45% μm or $C \cdot d_g^2 =$ 43–57 wt% μm;
- the manner of the relation 'temporal resistance – relative ultimate strength' practically does not change.

They also allow one to reconstruct the form of the dropping legs of the dose dependencies of the temporal resistance of modification 3.

Actually, since the mechanical properties of the material are determined by its defect structure and the main (and, practically, the only) defects in the material irradiated within the high temperature range are gas bubbles, we can find the secondary missing points of dose dependencies σ_{us}^{el} with the usage of the derivative equation

$$(F_f)_X = (T_2/T_1)^{2/3}\ \exp[2.1 \cdot 2/(3 \cdot 4k) \times (1/T_1 - 1/T_2)] \cdot (F_f)_2$$

for the corresponding swelling [3] from Ref. [3] and substitution of T_1, T_2, $(F_f)_2$ by their values from Table 3. Hence, the missing points are: 7.9×10^{21} n/cm^2 ($E \geqslant 0.85$ MeV), 100 [MPa] for $T_{irr} = 620$°C, $T_{test} = 650$°C and 3.37×10^{21} n/cm^2 ($E \geqslant 0.85$ MeV), 220 [MPa] for $T_{irr} = 680$°C, $T_{test} = 650$°C and the dependencies have the form (Fig. 7)

- σ_{us}^{el} [MPa] $= -3.66 \times 10^{-20} \cdot F_f\ [\mathrm{n/cm^2}\ (E \geqslant 0.85\ \mathrm{MeV})] + 392$ for $T_{irr} = 620°C$, $T_{test} = 650°C$,
- σ_{us}^{el} [MPa] $= -5.09 \times 10^{-20} \cdot F_f\ [\mathrm{n/cm^2}\ (E \geqslant 0.85\ \mathrm{MeV})] + 392$ for $T_{irr} = 680°C$, $T_{test} = 650°C$.

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[2] The values proportional to specific grain boundary porosity P_0 (i.e. relevant to unit grain boundary area) and specific grain boundary oxygen content C (i.e. corresponding to a unit grain boundary area).

[3] $\Delta V/V_0 = M \cdot T \cdot \exp[-2.1/(4kT)] \cdot F_f^{3/2}$.

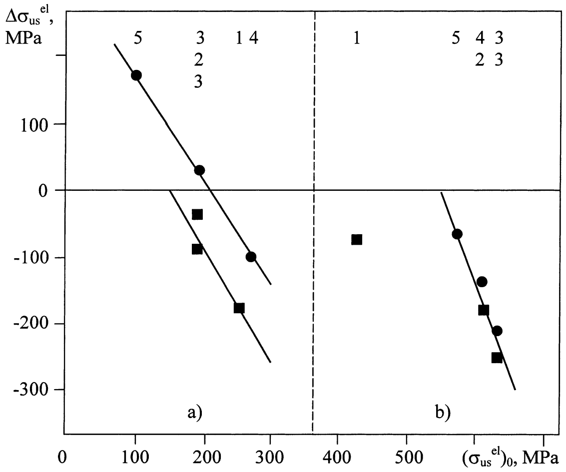

Fig. 2. Dependence of variation of material strength on its initial value. Dots in the areas (a) and (b) correspond to the temperatures of tests (650°C and 20°C). Numbers over the points are numbers of specimens types from Tables 1–3.

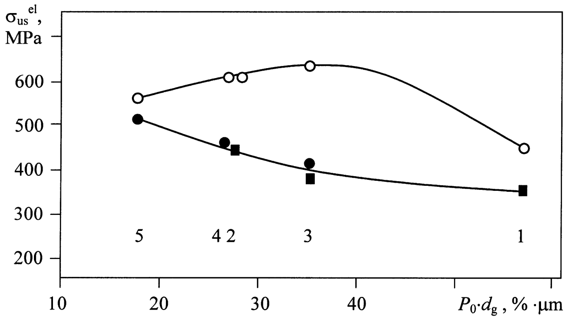

Fig. 3. The effect of irradiation and the parameter $P_0 \cdot d_g$ on the ultimate strength of the material at 20°C: ○ – before irradiation; ● – $T_{irr} = 620°C$, $F_f = 4.67 \times 10^{21}$ n/cm^2 ($E \geqslant 0.85$ MeV); ■ – $T_{irr} = 680°C$, $F_f = 5.69 \times 10^{21}$ n/cm^2 ($E \geqslant 0.85$ MeV). Numbers under the points are specimens types from Tables 1–3.

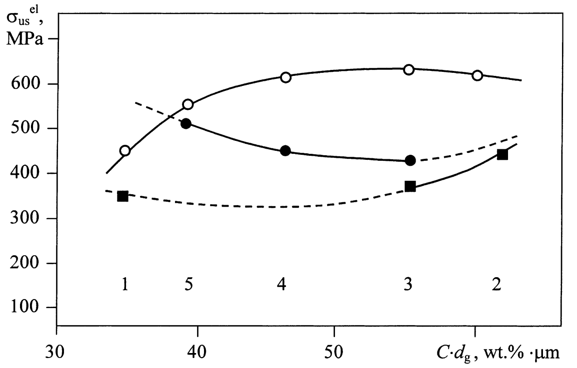

Fig. 4. The effect of irradiation and the parameter $C \cdot d_g$ on the ultimate strength of the material at 20°C: ○ – before irradiation; ● – $T_{irr} = 620°C$, $F_f = 4.67 \times 10^{21}$ n/cm^2 ($E \geqslant 0.85$ MeV); ■ – $T_{irr} = 680°C$, $F_f = 5.69 \times 10^{21}$ n/cm^2 ($E \geqslant 0.85$ MeV). Numbers under the points are specimens types from Tables 1–3.

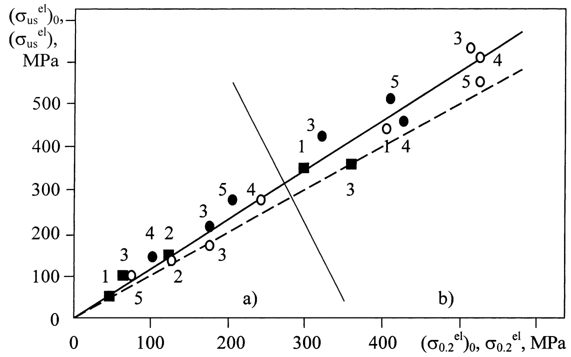

Fig. 5. Irradiation effect on the character of relationship "temporal resistance versus yield stress": ○ – before irradiation; ● – $T_{irr} = 620°C$, $F_f = 4.67 \times 10^{21}$ n/cm^2 ($E \geqslant 0.85$ MeV); ■ – $T_{irr} = 680°C$, $F_f = 5.69 \times 10^{21}$ n/cm^2 ($E \geqslant 0.85$ MeV). (a) region corresponds to $T_{test} = 650°C$; (b) region corresponds to $T_{test} = 20°C$. Numbers under the points are numbers of specimens types from Tables 1–3. Dashed line corresponds to the condition $(\sigma^{el}_{us})_0$ (and $\sigma^{el}_{us}) = (\sigma^{el}_{0.2})_0$ (and $\sigma^{el}_{0.2}$).

Therefore, the value of critical (i.e. fully softening the modification 3) fluence $(F_f)_{Tirr=650°C}$ is within the interval 7.7×10^{21}–10.7×10^{21} n/cm^2 ($E \geqslant 0.85$ MeV).

3. Conclusions

Thus, we observe that

- The neutron fluence dependence of the ultimate strength σ^{com}_{us} of beryllium DGP-600 irradiated at 650°C, includes two stages with the terms, denoting the observed effect, having different signs.
- At the first stage, when $(F_f)_1 < (F_f)_{max} \approx 3.43 \times 10^{20}$ n/cm^2 ($E \geqslant 0.85$ MeV), the changes of $(\sigma^{com}_{us})_1$ have a positive sign and reach the value $0.19 \cdot (\sigma^{com}_{us})_0$ at the maximum point.
- At the second one, when $(F_f)_2 > (F_f)_{max}$, the changes have a negative sign and attain the value $-(\sigma^{com}_{us})_0$ at a critical fluence
- Dose dependence σ^{com}_{us} has the form

$$\sigma^{com}_{us}\ [\mathrm{MPa}] = (\sigma^{com}_{us})0\ [\mathrm{MPa}] + 1.68 \times 10^{-3}\ [\mathrm{MPa\ cm}^{1/2}] \cdot F_f^{1/4}\ [\mathrm{cm}^{-1/2}] - 1.66 \times 10^{-19}\ [\mathrm{MPa\ cm}^2] \times F_f\ [\mathrm{cm}^{-2}].$$

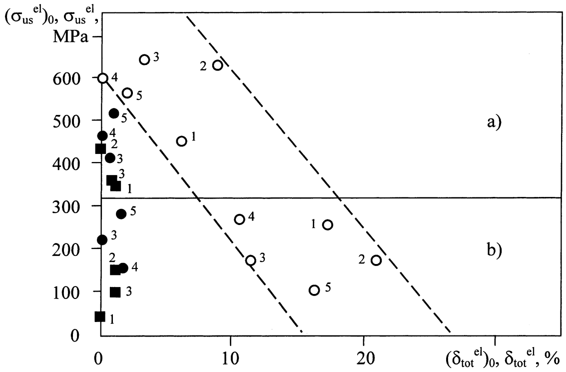

Fig. 6. Irradiation effect on general relative elongation of the material: ○ – before irradiation; ● – $T_{irr} = 620°C$, $F_f = 4.67 \times 10^{21}$ n/cm^2 ($E \geqslant 0.85$ MeV); ■ – $T_{irr} = 680°C$, $F_f = 5.69 \times 10^{21}$ n/cm^2 ($E \geqslant 0.85$ MeV); (a) region corresponds to $T_{test} = 20°C$; (b) region corresponds to $T_{test} = 650°C$. Numbers under the points are numbers of specimens types from Tables 1–3.

- The condition $\sigma_{us}^{com} < (\sigma_{us}^{com})_0$ holds at $F_f > 2 \times 10^{21}$ n/cm^2 ($E \geqslant 0.85$ MeV),$(\Delta V/V_0) > 1.3\%$.
- The value of the critical fluence, which fully softens the material, $(F_f)^d$ is 1.02×10^{22} n/cm^2 ($E \geqslant 0.85$ MeV).
- The value of the critical swelling, which fully softens the material, is $(\Delta V/V_0)^d$ and equals 14.58%.
- The important manifestation of Be radiation damageability is its embrittlement.
- The main manifestations of radiation damage of Be under irradiation to $F_f = 4.67$ and 5.69×10^{21} n/cm^2 ($E \geqslant 0.85$ MeV), at 620°C and 680°C are strengthening, softening and embrittlement.

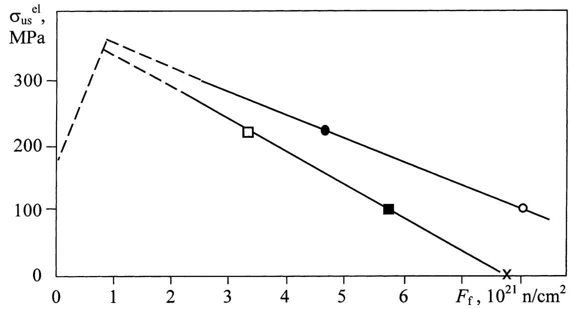

Fig. 7. An appearance of the dependencies $\sigma_{us} = f(F_f)$ of specimen type 3 when being softened $T_{test} = 650°C$: ●, ○ – experimental and calculated values of σ_{us} for $T_{irr} = 620°C$; ■, □ – experimental and calculated values of σ_{us} for $T_{irr} = 680°C$; x – critical value (at which the material is fully softened) of F_f for $T_{irr} = 680°C$.

- Strengthening-sensitive modifications are only those with $(\sigma_{us}^{el})_0$ <200 MPa ($T_{test} = 650°C$), and $(\sigma_{us}^{el})_0 <$ 550 MPa ($T_{test} = 20°C$).
- The degree of Be softening increases with its initial strength.
- The most softening is observed with the modifications of the values $P_0 \cdot d_g = 35$–45% μm or $C \cdot d_g = 43 - 57$ wt% μm.
- The critical fluence for complete softening of Be modification with $C = 2.75$ wt% O, $d_g = 20$ μm, $P_0 = 1.75\%$ is within the interval 7.7×10^{21}–10.7×10^{21} n/cm^2 ($E \geqslant 0.85$ MeV).

References

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[2] G.A. Sernyaev, VANT. Ser.: Nucl. Tech. Eng. 5 (1992) 48.
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