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Investigation of cryogenic irradiation influence on mechanical and physical properties of ITER magnetic system insulation materials

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Abstract

A set of methods of cryogenic irradiation influence test on mechanical and physical properties of insulation of ITER magnetic system are presented in this paper. Investigations are carried out without intermediate warming up of samples. A Russian insulating composite material was irradiated in the IVV-2M Reactor. The ratio of energy absorbed by insulation materials from neutron irradiation to that from gamma irradiation can be varied from ~(25:75) % to ~(50:50) % in the Reactor. The test results on the thermal expansion, thermal conductivity and gas evolution of the above material are presented. It was shown, that cryogenic irradiation up to the fluence $\sim 2 \times 10^{22}$ n/m^2 ($E \geqslant 0.1$ MeV) leads to 0.27% linear size changes along layers of fiber-glass, the thermal conductivity coefficient is decreased on 15% at 100 k in perpendicular direction to fiber-glass plane, and thermal coefficient of linear expansion (TCLE) has anomalous temperature dependence. © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

Insulation materials of ITER magnetic system are subjected by cryogenic gamma–neutron irradiation effect during their work. It leads to their gas and structure evolution. Both strength, thermal and electric properties and size are changed. We should pay attention to these factors during working out of ITER magnetic system. It is difficult to investigate such changes because of a number of properties are restored during samples warming up to room temperature. Therefore, investigations of samples irradiated under cryogenic temperature, carried out with intermediate warming up do not give the adequate picture of properties changes in real FR magnetic system conditions. It is necessary to have a scheme of experimental investigation realization, where irradiation and tests are performed at the same temperatures without samples intermediate warming up.

A methodical complex was developed in SB PDIPE. It allows to carry out samples irradiation in IVV-2M at 78 K, then to transportate them in liquid nitrogen medium (temperature is not changed), to determine a number of physical and mechanical properties of insulation materials and gluing joints and to perform gas evolution investigations in the temperature range 80–300 K.

2. A methodical complex facility to study a cryogenic irradiation effect on mechanical and physical properties of insulation materials

The investigation complex consists of both a low-temperature irradiation channel-cryostat (LTIC), located in the IVV-2M reactor core and a set of techniques realized in the hot cell laboratory.

The LTIC is 2-circuit liquid-nitrogen loop with aluminum capsule with samples located in the reactor core. The temperature of liquid-nitrogen coolant boiling under pressure is 78 K. Structure and dimensions of the

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channel are in detail described in Ref. [1]. Samples are placed into irradiating capsule, so that they blocked no more than 1/2 of capsule cross-section. It is necessary for normal circulation of liquid nitrogen. Therefore, limit size of the samples depends on their location in the capsule. Limit size and mass of the samples under simultaneous irradiation are shown in Table 1.

In order to control ratio of absorbed dose from gamma and neutron irradiation components a reactor cell (where LTIC is located) was equipped with special shielding screen. It allows to carry out irradiation under conditions with different gamma–neutron spectrum characteristics. Neutron irradiation monitoring was carried out by tracking indicator of 54Fe to control fluence. Both a set of activation detectors made of 197 Au, 59Co, 55Mn, 115In, 56Fe, 27Al and intrazone control channels based on ionization and fission chambers were used to measure flux differential density. After irradiation indicators were studded by neutron-activation analysis method. Neutron flux differential density is restored using program means of information-computing complex (ICC) and MCU programs.

Absorbed dose determination was performed by both carbon and polyethylene indicators and ionization chambers using ICC and information about chemical structure of insulation materials. Characteristics of both neutron flux and absorbed dose rate under irradiation conditions (“with shielding” and “without shielding”) of Russian candidate insulation material (fiber-glass laminated) are shown in Table 2.

The relation of absorbed doses from neutron and gamma components was (∼25%:75%) under irradiation in a mode without shielding and this relation was (∼50%:50%) in a mode with shielding.

After irradiation the capsule filled with liquid nitrogen is removed from the LTIC to a storage-cryostat for the decay of induced radioactivity for a month. Then it is transported in a shielding container-cryostat to a hot cell laboratory for investigations. There samples are sorted and distributed into cases with liquid nitrogen. All sorting operations are held in a bath filled with liquid nitrogen. The samples reloading are performed by a pouring method, so it excludes an opportunity of intermediate warming up of test materials. The samples are placed into a testing unit using the same method, with the exception of gas evolution investigation, where the sample is taken out from nitrogen for a short-term and placed into a work cell which is connected with measurement vacuum chamber.

Table 1
Sizes and mass of the samples for simultaneous cryogenic irradiation

Characteristics	Value
Maximum diameter (at vertical position), mm	28
Maximum diameter (at horizontal position), mm	20
Maximum length of the sample, mm	140
Total mass of the samples, g	50

Table 2
Characteristics of neutron fluxes and gamma dose rate

	Characteristics	Value
Without shielding	Neutron flux ($E < 0.1$ MeV), m^{-2} s^{-1}	3.7×10^{18}
	Neutron flux ($E > 0.1$ MeV), m^{-2} s^{-1}	1.8×10^{18}
	Gamma dose rate, Gy s^{-1}	3.3×10^{3}
With shielding	Neutron flux ($E < 0.1$ MeV), m^{-2} s^{-1}	2.8×10^{18}
	Neutron flux ($E > 0.1$ MeV), m^{-2} s^{-1}	1.4×10^{18}
	Gamma dose rate, Gy s^{-1}	1.6×10^{3}

The post-irradiation complex to study insulation and gluing joint materials without warming up of samples includes:

- measurement of size changes and thermal expansion characteristics,
- thermal conductivity characteristics measurement,
- gas evolution investigations,
- shear–compression tests of gluing joint material (insulator–metal) at different angles between loading axis and plane of shear.

If it is necessary, we can carry out shear, compression, elongation, 3-point bend tests of insulation materials.

Moreover, we can perform microstructure investigations at room temperature by the methods of optics and electron microscopy.

3. Comparison of IVV-2M and ITER reactors neutron spectrums

A calculation neutron energy spectrum for the operation area of the ITER magnetic system (obtained by Dr. G. Shatalov in SSC RF Kurchatov Institute) and an energy neutron spectrum in IVV-2M reactor were compared. Relative neutron contribution with different energies and integral flux density were also compared for the reactors. It was shown that relative neutron contribution with the energies (from 0.8 to 10 MeV) is higher in the IVV-2M reactor than in the ITER. At energies smaller than 0.8 MeV this contribution is higher in the ITER reactor. Besides, neutrons with energy higher than 10 MeV are occurred in the ITER, but their relative contribution is not more than 0.1% (see Table 3). Integral flux density in the IVV-2M reactor is higher than in the ITER magnetic system on 3 degree.

Close difference is observed in absorbed doze rate of gamma irradiation in the reactors. It allows to gather

Table 3
Different energy neutrons contribution into total flux

Energy interval		Relative contribution		
E min (MeV)	*E* max (MeV)	IVV-2M Without shielding	IVV-2M With shielding	ITER
10	14.2	0	0	0.001
0.8	10	0.164	0.169	0.021
0.1	0.8	0.159	0.160	0.165
0	0.1	0.677	0.671	0.813
Total flux (n m^{-2} s^{-1})		5.52×10^{18}	4.23×10^{18}	5.28×10^{15}

fluences and absorbed doses programming on lifetime of ITER for much less time in the IVV-2M reactor. Difference in irradiation intensity must not essentially influence on samples properties irradiated to identical fluence until both probability of overlaps damage is small and irradiation is carried out at lower temperature in spectrum with lower intensity.

4. Cryogenic irradiation influence on insulation material properties

Cryogenic irradiation influence investigation on a set service characteristics of insulation material was performed on Russian candidate samples (fibre-glass laminate). They were irradiated in a mode with shielding up to the fluences: 3×10^{21}, 1.1×10^{22}, 2.2×10^{22} n/m^2 ($E > 0.1$ MeV). Then size changes are studded on the samples at their warming up to room temperature. Thermal coefficient of liner expansion (TCLE) and coefficient of thermal conductivity (CTC) are estimated and gas evolution was also investigated. Sample size changes in the direction of fibre-glass layers are increased with fluence growth. Length measurement results of the samples at their warming up from 77 K to room temperature before irradiation, up to the fluence 2.2×10^{22} n/m^2 ($E > 0.1$ MeV) after irradiation and after their annealing at room temperature are shown in Fig. 1.

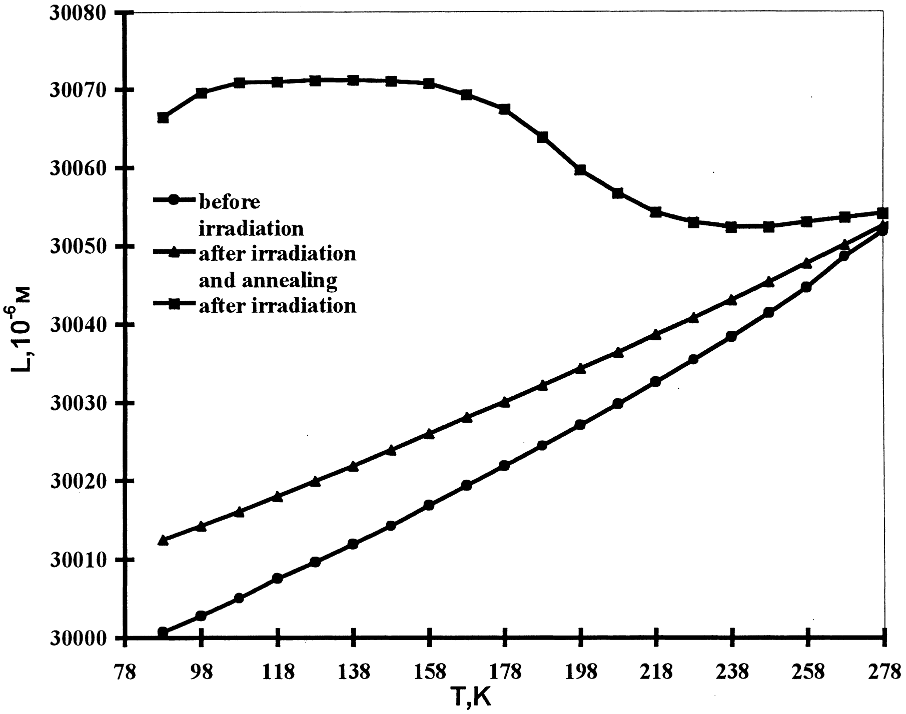

Fig. 1. Absolute length change of a sample during its warming-up ($F = 2.2 \times 10^{22}$ n/m^2).

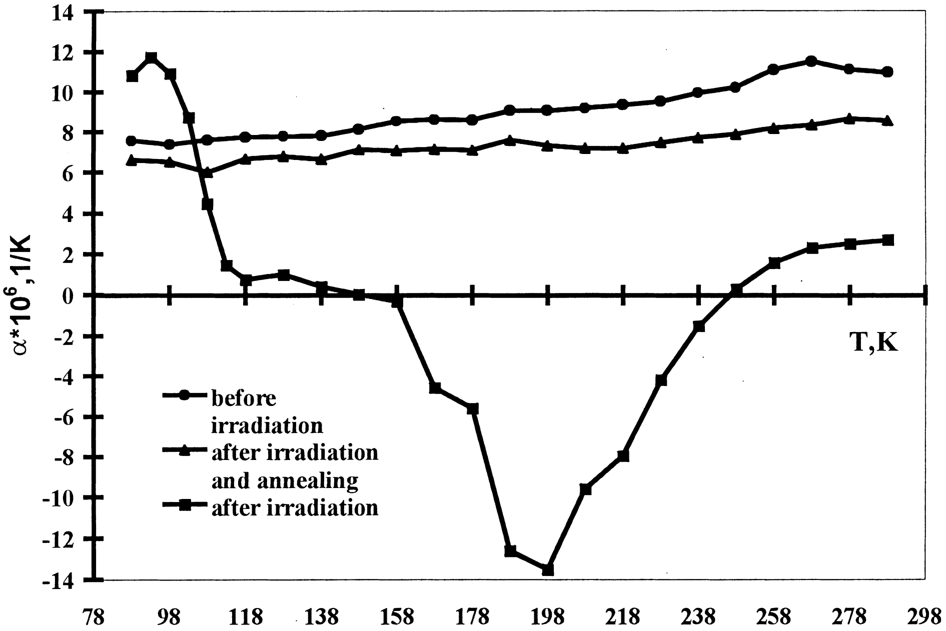

Fig. 2. TCLE of fiber-glass laminate before and after low-temperature irradiation ($F = 2.2 \times 10^{22}$ n/m^2).

The most length changes along fiber-glass layers reach 0.27% after cryogenic irradiation. Temperature dependence of TCLE of samples irradiated up to maximum fluence is shown in Fig. 2.

One can see, that TCLE decreases at temperature increasing above ~100 K. In the temperature range 150–250 K its value becomes negative, that is, the length is decreased under warming up to room temperature. It leads to incomplete restoration of linear sizes. Residual size changes are ~0.05%. Cooling up to 77 K and second measurements show that TCLE–temperature relationship of irradiated samples is the same as for nonirradiated samples at more lower values level.

Thermal conductivity measurement was carried out in the direction perpendicular to fiber-glass layers. Coefficient of thermal conductivity is computed using the measurement results. Its relative changes in the range 98–323 K, carried out on samples irradiated up to different fluences without intermediate warming up and after their warming up to room temperature are shown in Fig. 3.

One can see, that CTC is practically identically increased on ~15% for all samples after cryogenic irradiation. However, warming up leads to CTC decreasing which depends on fluence. The CTC of samples irradiated up to fluence 2.2×10^{22} n/m^2 ($E > 0.1$ MeV) is decreased after the second cooling on ~30%.

Gas evolution investigation is performed after irradiation up to fluences: 3.2×10^{21} n/m^2 – on one sample, 1.1×10^{22} n/m^2 – on two samples, 2.2×10^{22} n/m^2 – on one sample. Time of sample reloading and its pressurization in a working chamber did not exceed 10 c. Then the chamber was joined to vacuum measurement system. Amount of evolved gas was determined by pressure, structure of its components was determined by mass-spectrometer. The volume of evolved gas was determined on time base ~200 min (sec Fig. 4).

Then after longer hold times, gas structure and the volume of each component were determined (see Table 4).

The main components measured were: hydrogen, methane, carbon dioxide and nitrogen. We also measured small amounts of water steam, oxygen and argon. Nitrogen presence probably is due to its leakage into cracks formed in insulation material under cryogenic irradiation during its storing. Analysis of results carried out shows that amount of gas generated in insulation material is large and it increases with fluence's increase. Specific volume of evolved hydrogen is nearly 20 cm^3 under irradiation up to fluence 2.2×10^{22} n/m^2 ($E > 0.1$ MeV) under normal conditions on 1 cm^3 of insulation material.

5. Conclusion

Investigation results carried out show that cryogenic irradiation exerts essentially influence on thermal properties and leads to changes in thermal conductivity and size of insulation materials. Probably, the reason for such changes is radiation-induced gas evolution proceeding due to radiation defect generation, structure and

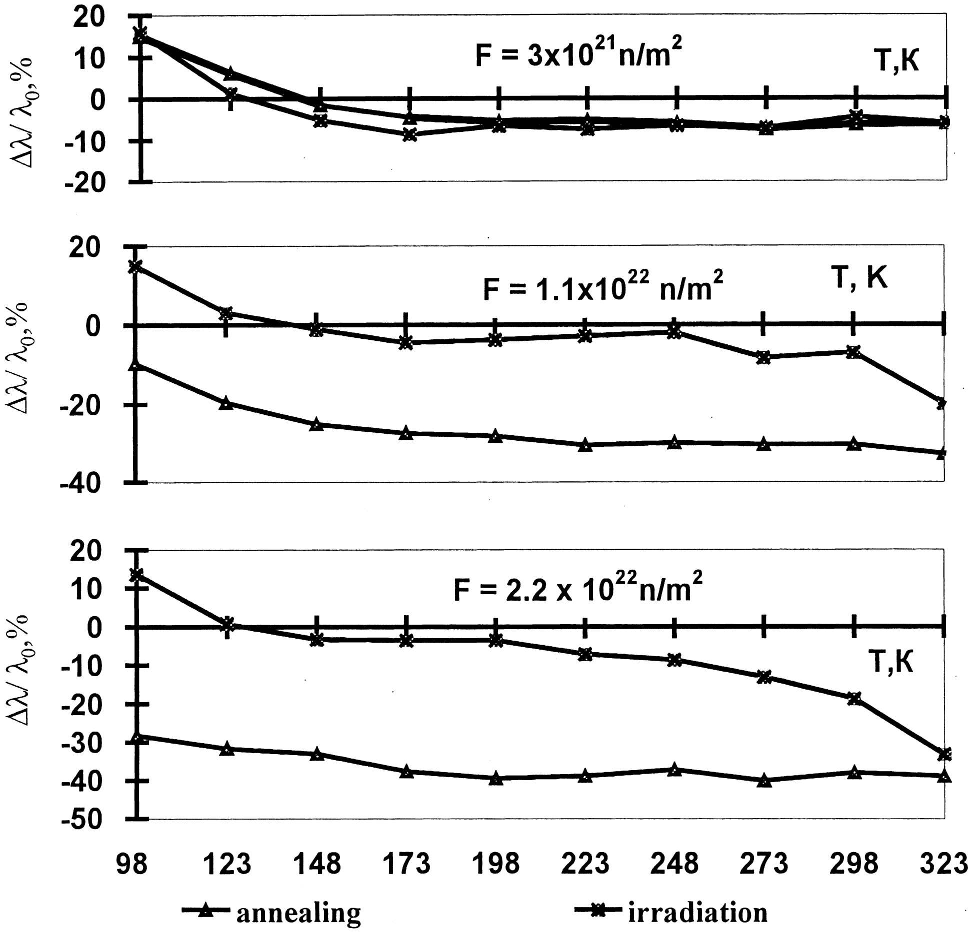

Fig. 3. Relative change of thermal conductivity coefficient of fiber-glass laminate after low-temperature irradiation.

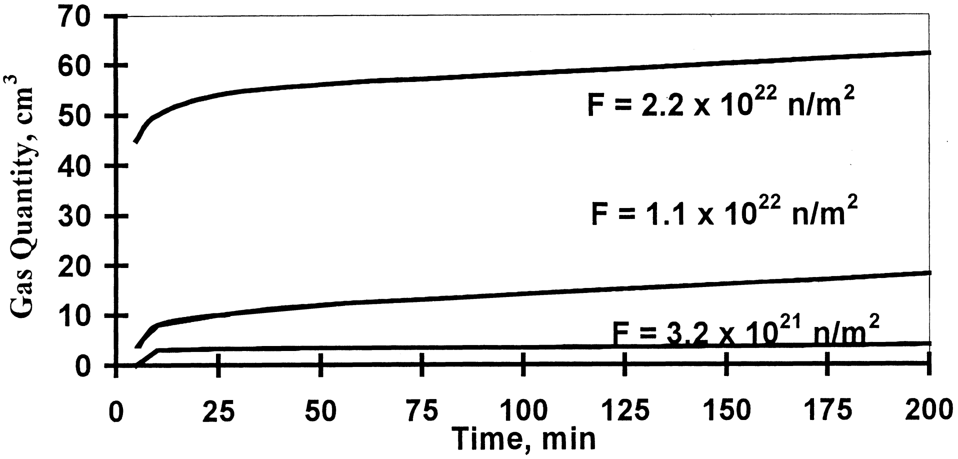

Fig. 4. Gas evolution kinetics.

Table 4
Component volume of gas evolved from irradiated insulation material

Fluence (neutron/m^2) (E > 0.1 MeV)	No. of sample	Time (h)	Quantity of evolved gas (normal cm^3)				
			H_2	CH_4	CO_2	N_2	Total [a]
$F = 3 \times 10^{21}$	1	187	0.0 ± 0.0	0.0 ± 0.0	0.21 ± 0.07	0 ± 9	2.86
		202	0.0 ± 0.0	0.0 ± 0.0	0.21 ± 0.05	1 ± 8	2.95
$F = 1.1 \times 10^{22}$	2-1	1205	13 ± 2	0.10 ± 0.01	0.65 ± 0.06	15 ± 10	26.5
		1225	13 ± 2	0.10 ± 0.01	0.78 ± 0.13	15 ± 10	26.6
		1230	13 ± 2	0.11 ± 0.01	0.66 ± 0.10	15 ± 11	26.6
	2-2	417	10 ± 1	0.00 ± 0.00	1.0 ± 0.2	15 ± 10	22.5
		1777	13 ± 2	0.03 ± 0.00	1.1 ± 0.2	16 ± 11	29.4
		4282	16 ± 2	0.14 ± 0.01	1.4 ± 0.2	17 ± 12	32.9
$F = 2.2 \times 10^{22}$	3	236	35 ± 2	2.1 ± 0.2	3.8 ± 0.2	23 ± 10	62.1
		1286	37 ± 6	2.7 ± 0.2	4.2 ± 0.3	22 ± 12	64.0
		1301	35 ± 6	2.8 ± 0.2	4.3 ± 0.5	23 ± 14	64.1
		1316	36 ± 5	2.8 ± 0.2	5.4 ± 0.7	24 ± 13	64.2
		1321	36 ± 6	2.8 ± 0.2	4.8 ± 0.7	23 ± 14	64.5

[a] There are no values for H_2O, O_2 and Ar whose contribution to gas evolution was small.

chemical composition changes of insulation material components. During the following warming up mobility of homogenous distribution gas is increased. The gas leaves to internal and external outlet. It is shown in gas evolution measurement results and size changes. A large part of gas leaves a sample, but near 10–20% of accrued gas stays in internal outlet (in pores and cracks). It must lead to decreasing of thermal conductivity and mechanical strengthening.

Most investigators measure both the sizes of insulation materials irradiated under cryogenic temperature after samples warming up and residual gas evolution. Such measurements allow one to evaluate only small part radiation affect. It is necessary to perform measurement of a set of characteristics without intermediate warming up in order to determine the value of insulation material properties and the amount of gas evolved from it.

In our opinion, it is not spared sufficient attention to the question of irradiation fluence determination in research reactor, which should influence on insulation materials, like ITER (equivalent fluence). To determine the fluence we must correctly select a criterion of cryogenic irradiation affect on insulation material. Damage dose called displacements per atom (dpa) is a criterion to select metal material. But in insulation material it is important to take into consideration several process simultaneously, such as:

- atom gas generation of different types and their diffusion mobility;
- point radiation defect formation, influenced significantly on diffusion process intensity;
- chemical structure changes of insulation material component by radiation-induced chemical reactions.

Using as a criterion the value of absorbed energy at close gamma–neutron component ratio of different reactors irradiation spectrum is insufficiently satisfied. Estimations show that difference only in energy distribution of neutron component can lead to difference in the amount of knock-out gas atoms in different reactors at the same fluence in several times. Therefore, it is necessary to carry out experimental investigations of cryogenic irradiation influence on candidate insulation material of ITER magnetic system. At the same time, it is worthwhile to develop quantitative model of processes occurred under irradiation in the materials and using it, to select criterion of radiation damage of insulation materials based on the model.

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