Development of indigenous insulation material for superconducting magnets and study of its characteristics under influence of intense neutron irradiation

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Abstract. Epoxy based glass fiber reinforced composites are the main insulation system for the superconducting magnets of fusion machines. 14MeV neutrons are generated during the DT fusion process, however the energy spectra and flux gets modified to a great extent when they reach the superconducting magnets. Mechanical properties of the GFRP insulation material is reported to degrade up to 30%. As a part of R & D activity, a joint collaboration with IGCAR, Kalpakkam has been established. The indigenous insulation material is subjected to fast neutron fluence of $10^{17} - 10^{19}$ n/m² (E>0.1 MeV) in FBTR and KAMINI Reactor, India. TRIM software has been used to simulate similar kind of damage produced by neutrons by ion irradiation with 5 MeV Al ions and 3 MeV protons. Fluence of the ions was adjusted to get the same dpa. We present the test experiment of neutron irradiation of the composite material (E-glass, S-glass fiber boron free and DGEBA epoxy). The test results of tensile, inter laminar shear and electrical breakdown strength as per ASTM standards, assessment of micro-structure surface degradation before and after irradiation will be presented. MCNP simulations are carried out for neutron flux, dose and damages produced in the insulation material.

1. Introduction
In order to find a suitable insulation system for the superconducting magnets of future fusion Tokomaks, an indigenous development activity towards the insulation materials under high neutron fluence has been initiated. The epoxy based glass fiber reinforcement plastics composite of G-10 CR grade is applicable for the fabrication of electrical insulation breaks, structural material, laminate form for electrical isolation. Due to D-T fusion process, the neutron degrades the mechanical performance of the material which overall effect the performance of fusion machine. Due to neutron irradiation the resin chemical structure changes in formation of radicals and bond breakage at low temperature. This increases the mobility of larger gas molecules CO, CO₂, CH₄ which degrade the insulation materials in form of swelling, increased porosity and decrease resin bond strength. The collaborative effort with IGCAR, Kalpakkam, India, the selected indigenous materials material has been irradiated with fast neutron of > 0.1 MeV energy at 300 K in 30 kW U-233 fuelled Fission Research Reactor up to the neutron fluence of $10^{17}$ n/m². The mechanical and electrical properties as tensile, interlaminar shear
strength and breakdown strength were investigated for the degradation in properties after irradiation. The radiation dose was measured during and after irradiation experimentally and also calculated with using neutron codes.

The aim of this work is to develop an insulation material with high neutron resistance for future superconducting fusion machine. The GFRP material has been taken for the evaluation of irradiation effect. Liquid modified Bisphenol A resin system with Liquid aromatic amine hardener shows the better performance compared to the conventional epoxy resins.

2. Experimental details, design, and test parameters

2.1 Materials

The investigated insulation material system was fabricated by using the high pressure hydraulic press [2] method to sheet form. The tube form of same material fabricated using filament winding method. The details of insulation material are listed in table 1.

| Glass fiber | Resin system |
|-------------|--------------|
| E-glass fabric of yarn 18.80 (Warp/cm) X 12.60 (Weft/cm), 64-68 % wt | Resin: 32.3 6% |
| Density: 203 g/m2 ,Thickness: 0.18 mm ± 5% | Liquid modified Bisphenol Epoxy A with liquid aromatic amine hardener |
| Tensile strength (yarn): 2189 x 1751 | Mixed viscosity: 1400 mPa.s |
| Warp (N/5cm) X Weft (N/5 cm) | Shear strength : 20 Mpa |
| Weaving pattern: Plane (Warp: EC 9 68 1 x 0) | Tensile strength: 48 Mpa |
| (Fill : EC 9 68 1 x 0) | Heat deflection temp: 58 º C |
| Class F insulation IS-10192 | Specific gravity: 1.15 -1.20 |
| Composition:SiO2: 54% wt, AL2O3: 14% wt, CaO + MgO: 22% wt., B2O3: <10% wt., Na2O+K2O: < 2% wt, | Epoxy value: 5.25-5.40 eq/kg |

3. Irradiation and test procedure

The neutron irradiation was done in U-233 fuel based KAMINI fission 30 kW Reactor at 300 K to fast neutron up to 1.0 x 10^{17} n/m^2 neutron fluence (E>0.1 MeV) which corresponds to the Gamma dose at the time of irradiation of 6x10^8 R/h.. The samples were irradiated inside the dry tube locations 1 and 2 by hanging with nylon threads. The samples were fixed on the of 1S nuclear grade aluminum former as shown in figure 1a and 1b. The irradiation was done for 12 hours for breakdown strength samples and 18 hours for tensile and shear strength samples at dry tube location 1 and 2 respectively for equalizing the neutron fluence ~1.0x10^{17} n/m^2 in all samples as shown in the table 2. The experimental set up and interview of Reactor is shown in figure 2 and figure 3.

| Insulation Samples | Flux (n/cm^2.S) DT-1^a | Neutron Fluence (n/m^2) DT-1 DT-2 | Distance (cm) DT-1 DT-2 |
|--------------------|------------------------|------------------------------------|-------------------------|
| Sample 1(Breakdown strength) | 2.40x10^8 | 1.03x10^{17} | 18 |
| Sample 2 (Tensile strength) | 5.50x10^7 | 1.00x10^{17} | 92 |
| Sample 3 (Shear strength) | 5.50x10^7 | 1.00x10^{17} | |
| From reactor core | | | |
| ITER design fluence | 1.0x10^{22} | | |

^a DT-1-dry tube location 1 where samples 1 irradiated
^b DT-2-dry tube location 2 where samples 2 and 3 irradiated
4. Simulation with neutron codes and its results

SRIM and TRIM neutron codes programs were used to simulate similar kind of damage produced by neutrons by ion irradiation with 5 MeV Al ions and 3 MeV protons. Fluence of the ions was adjusted to get the same dpa. 10 dpa from 5 MeV Al ion will require a fluence of 3.2e16 ions/cm\(^2\) will take around 3 hours for 1cm\(^2\) area with a beam current of 0.5 micro Amp in the range 3.9 micron. For 3 MeV proton, range will be 90 micron of 1dpa itself will require a fluence of 5.04e18 ions/cm\(^2\), It may take a long nearer to 10 days. From the ion irradiation simulation results it shows the less damage occurs comparing with neutron irradiation. The MCNP neutron code was used for calculating the flux in all insulation samples with respect to the neutron energy at the time of irradiation as shown in table 3.

| Neutron Energy       | Flux at 20 kW (N/cm\(^2\). S) |
|----------------------|-------------------------------|
|                      | DT-1 | DT-2 |
| Up to 0.021 eV       | 9.42E+07 | 1.72E+07 |
| 0.021 eV > E <1 eV   | 1.46E+08 | 3.77E+07 |

Figure 1. Samples arrangement for irradiaition in Reactor (a), GFRP material samples (b)

Figure 2. Schematic of neutron experimental set up

Figure 3. Reactor internal view
4.1 Preliminary, Post Examination and Measurement on Irradiated Samples

The insulation material sample has irradiated in powder form of 20 mg for 5 minutes at 1 kW reactor power to know the behavior of material reaction with other materials and impurity level in reactor. The gamma radiation dose was measured at the time of irradiation, after irradiation in all samples as shown in figure 4. The gamma energy spectrum European activation system code ‘EASY’ (uses fish pact) was used for estimating the presence of isotopes and their activation, dose rate of isotopes present and in each samples. The isotopes presence activation and dose rate in all samples and isotopes is presented in table 4 and table 5 and respected graphs as shown in figure 5.

| Energy Range          | Activation (Bq) | Dose rate (mR/hr) |
|-----------------------|-----------------|-------------------|
| 1 eV > E < 10.7 eV    | 1.23E+05        | 1.55E+05          |
| 10.7 eV > E < 96.1 eV | 5.73E+03        | 6.03E+03          |
| 96.1 eV > E < 111 keV | 5.94E+01        | 1.32E+02          |
| 111 keV > E < 1 MeV   | 2.02E+00        | 6.23E+00          |
| E > 1 MeV             | 4.10E-01        | 1.35E+00          |
| <0.1 MeV              |                 |                   |
| >0.1 MeV              |                 |                   |

Table 4. Isotopes activation and dose rate.

| Isotopes presence | Activation (Bq) at (irradiation time) | Cooling (Irradiation time) | Dose rate (mR/hr) Cooling |
|-------------------|---------------------------------------|-----------------------------|--------------------------|
|                   | 6 hr 12 hr 1 days 4 days 8 days       | 6 hr 12 hr 1 days 4 days 8 days |
| Na 24             | 2.33x10^7 3.09x10^7 1.02x10^9 3.61x10^10 4.22x10^10 | 2.61x10^7 3.47x10^8 1.14x10^9 4.06x10^10 4.57x10^10 |
| K42               | 4.51x10^7 5.68x10^7 1.48x10^9 2.61x10^10 1.20x10^11 | 3.15x10^9 3.97x10^10 1.03x10^11 1.82x10^12 8.37x10^10 |
| Ca 45             | 5.63x10^7 1.07x10^11 1.07x10^11 1.05x10^11 1.04x10^12 | 8.05x10^15 1.61x10^14 1.60x10^14 1.58x10^14 1.55x10^14 |
| Ar 37             | 3.14x10^7 6.21x10^7 6.09x10^9 5.74x10^10 5.30x10^11 | 1.12x10^10 2.21x10^10 2.1x10^10 2.04x10^-10 1.89x10^-10 |
| SC 47             | 7.75x10^0 6.65x10^3 1.51x10^5 2.57x10^9 2.63x10^9 | 7.61x10^11 6.52x10^10 1.49x10^9 2.52x10^-9 2.31x10^-9 |

Table 5. Dose rate in irradiated samples.

| Irradiated samples | Dose rate (mR/hr) |
|--------------------|-------------------|
|                    | (Irradiation time) | Cooling |
|                    | 6 hr 12 hr 24 hr 48 hr 96 hr 120 hr |
| Breakdown sample 1 | 67.99 70.24         | 8.73 2.99 0.98 0.11 0.04 |
5. Mechanical, Electrical Performance Tests and Results

All tests were carried out of unirradiated and irradiated samples at 300 K as per the ASTM standards at Electrical Research & Development Association (Accredited by the National Accreditation Board for Testing and calibration (NABL), Govt. of India), Vadodara, India The tests were performed on three numbers of unirradiated and irradiated samples.

5.1 Ultimate tensile Strength

The tests were performed by using Universal testing machine 6074 of 100 kN capacity with speed 5mm/min as per the ASTM D638 standard as shown in figure 6. For the tensile test [1], samples in figure 7 were cut in 90° direction; in this direction the influence of radiation damage is more. From the test result only 2% degradation is observed as shown in figure 8.
Interlaminar shear strength

The wrapping procedure the materials have anisotropic properties, therefore the shear strength specimens were cut parallel (0°) and perpendicular (90°) to the winding direction of the reinforcing glass fiber [4]. The test was performed by using tensile test machine subjected to the test piece to a shear load by means of a circular punch and die shown in the figure 9 and figure 10 as per the standard ASTM 4248-1967. The result indicates that no significance degradation was observed for the shear property of used insulation system for the design neutron fluence as shown in table 7.

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**Table 6. Result of tensile strength test**

| Samples      | Breaking load (N) | Dimension Length x Breadth x thickness (mm) | UTS (MPa) | Displacement (mm) |
|--------------|-------------------|---------------------------------------------|-----------|-------------------|
| Unirradiated | 8460              | 115x19x5                                    | 256       | 3.66              |
| Irradiated   | 8379              | 115x19x5                                    | 253       | 3.36              |

**Table 7. Result of Interlaminar shear strength and breakdown strength test.**

| Samples       | Breaking load (N) | Dimension Length x Breadth x thickness (mm) | Interlaminar Shear Strength (Mpa) | Breakdown strength kV/mm |
|---------------|-------------------|---------------------------------------------|-----------------------------------|--------------------------|
| Shear sample  |                   |                                             |                                   | Air                      |
| Unirradiated  | 34251             | 100x20x5                                    | 50x50x5                           | 168.5                    |
| Irradiated    | 33127             | 100x20x5                                    | 50x50x5                           | 163.0                    |
| Breakdown     |                   |                                             |                                   | Oil                      |
| Unirradiated  |                   |                                             |                                   | 10.9                     |
| Irradiated    |                   |                                             |                                   | 10.7                     |
5.3 Breakdown strength
The test was performed by using 150 kV high voltages set up using 6 mm brass electrodes were used as per the standard ASTM D 149-2009. The tests were performed in air as well in oil medium at 300 K as shown in figure 11a and figure 11b. The flashover was observed in air medium at > 10.9 kV in samples but no puncture seen. In air medium due to some moisture content over the insulation surface with deposited dust on the insulator surface produces a conducting path. As a result the flash over the chance of flash over becomes more in air medium compare to oil. Due to space constraint in reactor for irradiation experiment the sizes of the samples were reduced from 200 x 200 x 5 mm to 50 x 50 x 5 mm, this could be the one of main reason for flash over. No significance degradation in breakdown strength of unirradiated and irradiated samples was reported as shown in figure 12.
Summary
From the test results no significant degradation is observed in tensile, shear and breakdown strength properties of the used insulation material. In this work modified epoxy system with high toughness and E-glass show higher resistance than commercial epoxy system investigated at 300 K, 77 K for $10^{14}$ n/m² neutron fluence [3] which was reported degradation up to 26% and 9% in tensile and shear strength respectively. This indicates that the resin system needs more innovation development for the higher radiation environment. The indigenous developed GFRP insulation system shows the potential and experimentally validated their neutron resistance up to $10^{15}$ n/m² fluence and radiation resistance up to a dose of $6 \times 10^4$ R/hr. For future application, where higher radiation levels are expected, further study and experimental plan for the innovation of new high radiation resistance cyanate ester base resin system reinforced with boron free ‘S’ and ‘R’ glass fiber, irradiation for long dose time, fast neutrons with high energy level (E>0.1 MeV) and neutron fluence up to ~ $1.0 \times 10^{22}$ n/m² (ITER design fluence level).

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Figure 12. Result of breakdown strength test.