Recent Progress and Tests of Radiation Resistant Impregnation Materials for Nb_3Sn Coils

R. Bossert, S. Krave, G. Ambrosio, N. Andreev, G. Chlachidze, A. Nobrega, I. Novitski, M. Yu and A.V. Zlobin

Technical Division, Fermi National Accelerator Laboratory, P.O. Box 500, Batavia, IL, 60510 U.S.A.

Abstract. Fermilab is collaborating with Lawrence Berkeley National Laboratory (LBNL) and Brookhaven National Laboratory (BNL) (US-LARP collaboration) to develop a large-aperture Nb_3Sn superconducting quadrupole for the Large Hadron Collider (LHC) luminosity upgrade. An important component of this work is the development of materials that are sufficiently radiation resistant for use in critical areas of the upgrade. This paper describes recent progress in characterization of materials, including the baseline CTD101K epoxy, cyanate ester blends, and Matrimid 5292, a bismaleimide-based system. Structural properties of "ten stacks" of cable impregnated with these materials are tested at room and cryogenic temperatures and compared to the baseline CT-101K. Experience with potting 1 and 2 meter long coils with Matrimid 5292 are described. Test results of a single 1-m coil impregnated with Matrimid 5292 are reported and compared to similar coils impregnated with the traditional epoxy.

Keywords: Potting, matrimid, cyanate ester, epoxy, insulation.

INTRODUCTION

The final step in the fabrication of Nb_3Sn coils is an impregnation to give the coil a precise size, an adequate structure, and protect the fragile Nb_3Sn strands from degradation during assembly and operation. The traditional epoxy material that has been used provides all these features. However, it is not clear that epoxy has a radiation resistance adequate for use in future accelerators, like the LHC. Two alternative materials (Matrimid 5292 [1], a bismaleimide based material) and a cyanate ester blend (CTD425) [2] can provide higher radiation resistance, but have not been tried in actual Nb_3Sn magnets. This paper compares these materials to the traditional epoxy (CTD-101K). Manufacturing issues and results of mechanical and electrical tests are reported. Examples of impregnated cross sections, comparing the "adequacy of fill" of the various materials are also shown.

MANUFACTURING ISSUES

Each of the materials being compared have different potting and curing processes, with the different requirements affecting the fabrication procedure. The baseline CTD101K epoxy allows for a long 24 hour pot life with a reasonably low viscosity of 100cP at 60°C. CTD101K does not pose any significant handling challenges. Matrimid poses some challenges with handling. Compared to the other two materials, it has significantly higher processing and curing temperatures and shorter pot life. It is also very sensitive to temperature fluctuations in terms of viscosity and pot life. CTD425 offers excellent potlife and good processing temperatures, however it does pose issues as exposure to water vapor or volumes of neat resin may lead to exotherm, resulting in overheating, during cure. Table 1 lists the temperature, viscosity and pot life for each material.

TABLE 1. Features of Potting Materials

Material	CTD101K	Matrimid 5292	CTD425
Family	Epoxy	Bismaleimide	Cyanate Ester Blend
Initial Viscosity	100cP	10cP	70cP
Potting Temperature	60°C	125°C	60°C
Max Cure Temperature	135°C	200°C	150°C
Pot Life	24 hours	60 minutes	100 hours

Operated by Fermi Research Alliance, LLC under Contract No. De-AC02-07CH11359 with the United States Department of Energy.

After initial validation of Matrimid on one meter long cable samples, both a one and two meter coil were potted [3,4]. They are shown in Fig. 1. Each coil was vacuum impregnated while being maintained at the proper potting temperature by an oven. Vacuum and resin transfer lines were heated and insulated exterior to the furnace to prevent solidification of the Matrimid. Coil potting occurred with unrestricted inlet flow to minimize the amount of time it would take to pot each coil. This strategy contrasts that of potting coils with epoxy, which are typically throttled to allow slow and even fill of the coil while utilizing the long pot life. Resin flow direction was along the length of the coil in a similar fashion to the impregnation of coils using CTD101K.

Resin volume was observed during impregnation and vacuum was released when resin was observed flowing out of the coil. Time elapsed from start of potting until release of vacuum was 15 minutes for the 1m TQ coil and 60 minutes for the 2 meter 11 Tesla coil potted with Matrimid. A series of 3.4 m long coils potted with CTD101K required 6 hours to fill. These times are consistent with the assumption of a coil as porous media where time to flow a distance could be approximated by

$$t = L^2 \frac{n}{k} \frac{\mu}{\Delta P}$$

Where t is time, L is the coil length and k is the coil constant, n is porosity, ΔP is the pressure differential, and μ is viscosity [5]. If coil fabrication remains the same, n, ΔP, and k stay constant and μ is a function of the resin. A rough approximation of the longest coil that could be potted can be found by setting t equal to pot life. When compared to the expected values, and using CTD101K coils as a reference, impregnation occurred slower than expected with the Matrimid, indicating an effectively higher viscosity than expected. To determine the maximum coil length able to be potted using each material, the calculations for CTD101K and Matrimid used a fit to actual coil fabrication data, while the CTD425 calculation used the fit for CTD101K and the published viscosity and pot life values as shown in Table 1. Approximate maximum coil length can be found in Table 2. Coil length values are extremely sensitive to changes in viscosity.

TABLE 2. Maximum Coil Length for Different Potting Materials

Material	Time (minutes)	Maximum Coil Length (m)
CTD101K	1440	6.8
Matrimid 5292	90	2.4
CTD425	6000	16

If simple vacuum impregnation will not provide a sufficiently fast potting time, such as for a Matrimid coil longer than 3 meters, several manufacturing options are available. The direction of flow could be changed, the pressure differential could be increased or multiple inlets and outlets in the impregnation fixture could be used. The need to pot coils of greater length will be the primary challenge for the viability of using the Matrimid material.

(a) **(b)**

FIGURE 1. 1-meter Quadrupole coil (a) and 2-meter Dipole coil (b) potted with Matrimid.

CABLE STACKS

Mechanical measurements of cables were performed on small stacks made of 10 cables each. Stacks are made by the same manufacturing process used for coils, consisting of a pre-cure with CTD 1202X binder at 10 MPa, a reaction at temperatures up to 650C, and an impregnation which varies with the material used, as described above. The stacks were then instrumented with strain gauges. All stacks were made identically except when differences in the potting material made variations in the impregnation process unavoidable.

The cable and cable insulation used for the stacks was identical to that used in TQ and LQ (90 mm bore Quadrupoles) manufactured for LARP (LHC Accelerator Research Project). The cable consisted of RRP 108/127 strands of .7mm diameter. Each cable was insulated with a 125 um thick sleeve made of S-2 glass. Cable and insulation information can be found at [6].

Of course, mechanical and electrical measurements of the stacks are dependent on the cable type and insulation, and this study measures only one type of cable/insulation combination. This study therefore will yield acceptable relative values of the three materials, but these values may be different if other cable designs are used. The TQ cable was chosen because it has been widely used in many short and long models [7-12], with a great deal of fabrication and test data available.

Mechanical Measurements

The cable stacks for mechanical measurements are shown in Fig. 2. There are two kinds of stacks, "azimuthal" and "radial", stacks. They are constructed identically, but are pressed in different ways, and instrumented accordingly. Type (a) are pressed "azimuthally" and type (b) are pressed "radially". The azimuthal and radial designations refer to the direction that the preload is applied to the cable in a typical cosine theta coil, as shown in Fig. 3.

Four strain gauges are applied to each stack. Two of them read the strain in the direction of the force applied, while two gauges read the strain in the transverse directions.

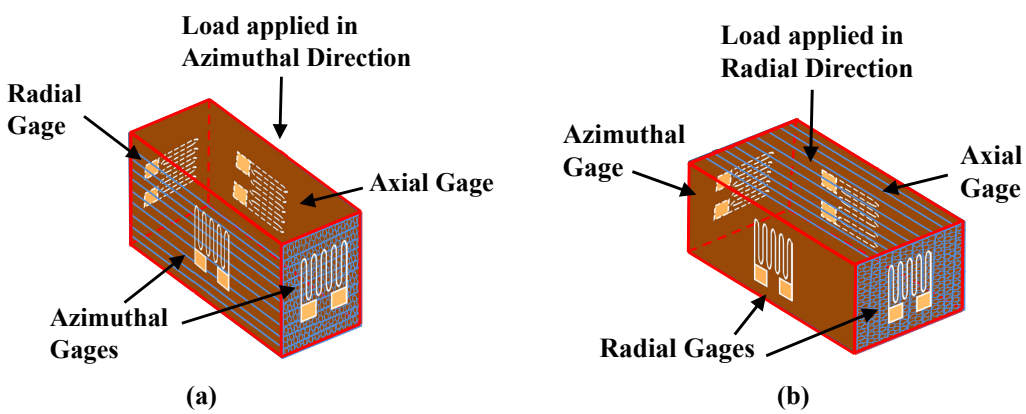


FIGURE 2. Instrumented cable stacks, showing the gauge configuration. Stack (a) is pressed in the azimuthal direction, and stack (b) is pressed in the radial direction. in both the azimuthal and radial direction.

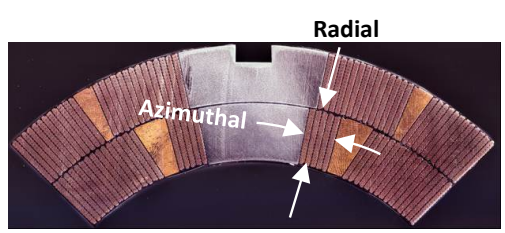


FIGURE 3. Magnet cross section with azimuthal and radial preload directions described.

To date, 3 azimuthal and 3 radial stacks each of CTD101K and Matrimid have been measured. 3 cycles to 100 MPa or 140 MPa and back to zero were done on each stack. Mechanical testing of Cyanate Ester Stacks as well as cryogenic testing of all stacks has not yet been completed, and will be deferred until a later study.

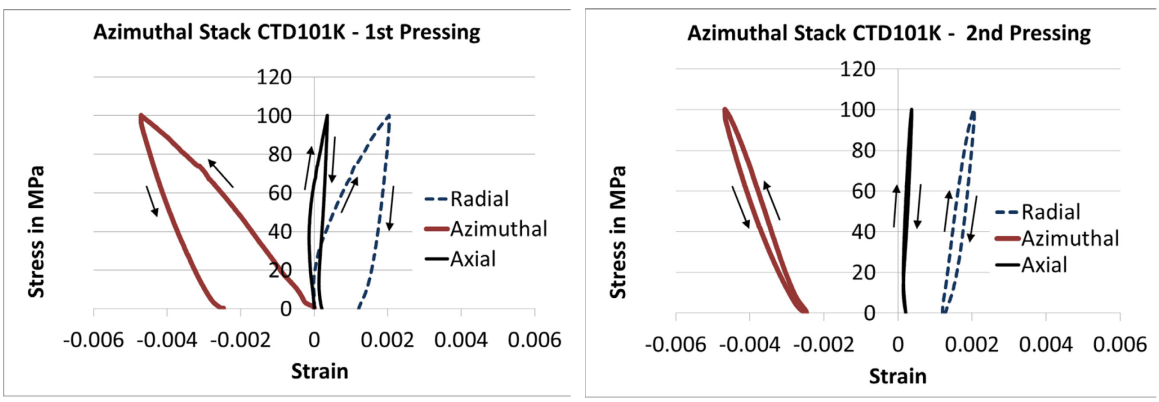
The typical shape of stress-strain curves for initial and subsequent pressings in azimuthally and radially pressed stacks is similar for both CTD101K and Matrimid, and are shown in Figs. 4 and 5 respectively (CTD-101K stacks are shown). Note that the material typically yields after the first pressing, while the Young's modulus of the cable in the azimuthal direction increases by approximately a factor of two between the first and subsequent pressings. The behavior in the radial direction is more complex, with very non-linear behavior. The stack size returns to nearly zero after each pressing, but on subsequent pressings the slope clearly changes at about 30 MPa. This effect is true of the radial stacks in both the CTD101K and the Matrimid materials.

Table 3 gives the data gathered from all stacks measured so far. Young's modulus was measured in both the azimuthally and radially pressed stacks. Poisson's ratio in both transverse directions was measured and is shown for the azimuthally pressed stacks, but is not reported for the radially pressed stacks.

Young's Modulus in both the azimuthal and radial direction for CTD101K are consistent with expectations. Values used in past analysis of Nb_3Sn TQ magnets were 20 GPa/40-44 GPa for first and subsequent azimuthal pressings and 25 GPa/50-52 GPa respectively for radial pressings [13]. The values found for the region of 30-100 MPa are consistent with the radial modulus being used in analysis.

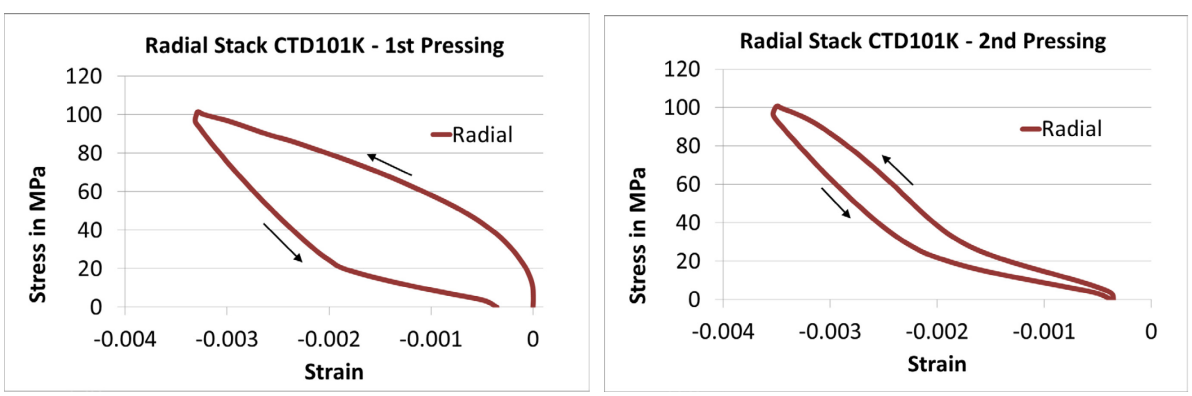
Measurements of Poisson's ratio in the radial direction when pressing azimuthally are reasonably close to the value of .3 used in previous analysis, but the axial value found here is significantly smaller.

A previous study [14] which looked at Young's modulus only, showed values significantly smaller than those in Table 3. This is likely due to the fact that the stacks used in the previous study used different cable, different and much thicker cable insulation, and had not undergone the reaction process before impregnation. The stacks shown here are therefore more realistic with respect to the cable actually used in magnets. Since they have an identical system to that used in the TQ and LQ magnets, direct comparisons can be made to the behavior of those coils.



(a) **(b)**

FIGURE 4. Stress vs strain of cable stacks pressed azimuthally. Initial pressing (a) and subsequent pressings (b).



(a) **(b)**

FIGURE 5. Stress vs strain of cable stacks pressed radially. Initial pressing (a) and subsequent pressings (b).

TABLE 3. Young's Modulus of Elasticity and Poisson's Ratio of stacks pressed azimuthally and radially.

Potting Material	Stack Type (Pressing direction)	Parameter	Initial Pressing	Subsequent Pressings
CTD101K	Azimuthal	Young's Modulus Az	17	37
		Poissons ratio radial	.34	.37
		Poisson's ratio axial	.12	.20
	Radial	Young's Modulus Rad	30	33/46*
Matrimid 5292	Azimuthal	Young's Modulus Az	12	31
		Poissons ratio radial	.27	.39
		Poisson's ratio axial	.11	.14
	Radial	Young's Modulus Rad	22	21/51*

*33/46 and 21/51 represent Young's Modulus (between 0-100 MPa/between 30-100 MPa)

FIGURE 6. An electrical stack.

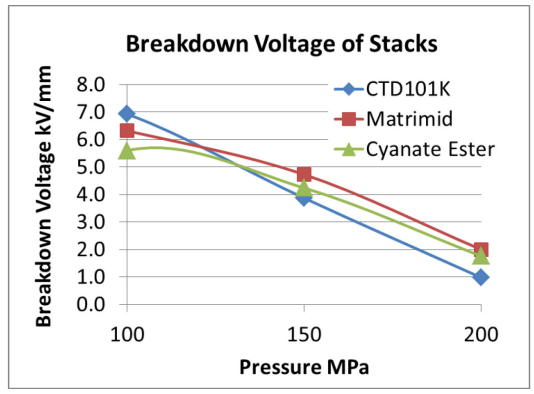


FIGURE 7. Breakdown Strength as a function of Pressure

Electrical Measurements

A series of stacks were also fabricated to evaluate the electrical integrity of each material. Figure 6 shows a picture of a stack and the method of wiring. Each stack is 14 cm long, and the total area being pressed on each stack was approximately 14 square centimeters. 5 stacks were measured, two potted with CTD101K and Matrimid and one with Cyanate Ester. Further stacks will be completed to gain more statistical data. Current leakage and voltage breakdown was measured at voltages up to 2500 volts and pressures to 200 MPa.

No leakage current for any stack exceeded 60 nA before breakdown, which is well within the criteria necessary for current Nb_3Sn magnets. This is similar to the leakage reported previously for similar insulation systems [15] using CTD101K.

Figure 7 shows the breakdown voltage for the CTD101K, Matrimid and Cyanate Ester stacks. Breakdown voltage is slightly lower than reported in [14] for CTD101K and Matrimid, which may be due to the different cable insulation system used (S-2 glass here vs. ceramic in the previous study). However, the relationship between the CTD101K and Matrimid (with Matrimid having a higher dielectric strength) is similar. At the benchmark pressure of 150 MPa, dielectric strength for the CTD101K is 3.8kV/mm while Matrimid is 4.8kV/mm.

Thermal Contraction Coefficient

The integrated thermal contraction coefficient of each stack was measured in the azimuthal, radial and longitudinal directions. Each stack was immersed into 77K liquid nitrogen, and measured using the strain gauges bonded to the stack. Measurements were made using a strain gauge technique [14,16]. The temperature induced strain in a sample due to a change in temperature ΔT is given by:

$$\varepsilon_s = \frac{\Delta R_s}{RF_G} = \left[\frac{\beta_G}{F_G} + (\alpha_s - \alpha_G) \right] \Delta T \tag{1}$$

Where $\Delta R/R$ is the unit resistance change, β_G is the thermal coefficient of resistance of the grid material, $(\alpha_s\text{-}\alpha_G)$ is the difference in thermal coefficients between the sample and the grid respectively and F_G is the gauge factor. If the same type of gauge is installed in a standard reference material with a known thermal coefficient α_R, then:

$$\varepsilon_R = \frac{\Delta R_R}{RF_G} = \left[\frac{\beta_G}{\mathrm{F}_G} + (\alpha_R - \alpha_G) \right] \Delta T \tag{2}$$

Subtracting the above two equations and rearranging, we get:

$$(\alpha_s - \alpha_R) = \frac{(\varepsilon_s - \varepsilon_R)}{\Delta T} \tag{3}$$

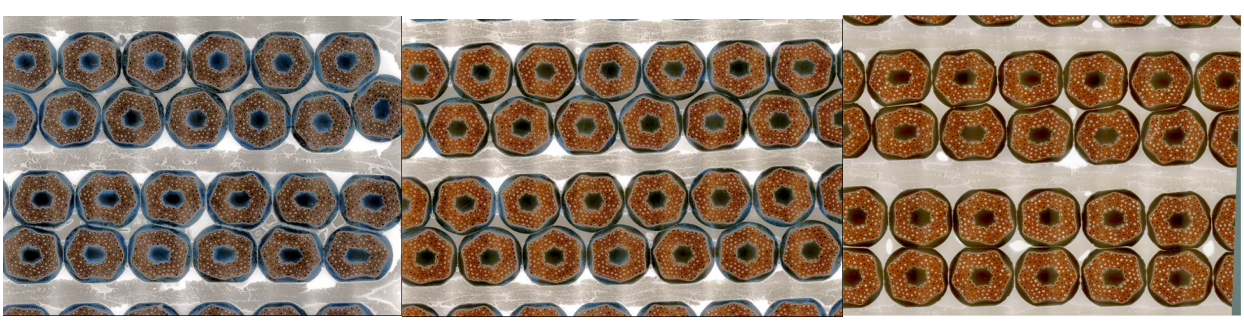
Knowing α_R, εs and ε_R for a particular change in temperature, we can compute αs, the integrated thermal contraction coefficient of the sample. A disk of quartz was used as the reference sample, since it has a thermal coefficient of nearly zero, and an instrumented aluminum block was added for further verification that the measurements were accurate. Measurements have been initiated, but are still ongoing, with only measurements between 290K and 77K completed. Results are shown in Table 4. The CTD101K measurements are consistent with previous measurements [14].

TABLE 4. Thermal contraction coefficients from room temperature to 77K (mm/m)

Material	CTE azimuthal	CTE radial	CTE axial
Quartz Reference sample	0	0	0
Aluminum test sample	.0038	.0038	.0038
CTD-101K	.0032	.0022	.0019
Matrimid 5292	.0033	.0017	.0017
Cyanate Ester 425	.0029	N/A	.0018

IMPREGNATION QUALITY

Two stacks of each material were polished and observed under a microscope to assess the degree of fill around and within each turn. Figure 8 shows each stack. Gray areas are completely filled with the impregnation material while white areas represent small voids. Impregnation quality of the stacks of the three materials is adequate, with the cyanate ester stack having slightly better fill than the other two. The quality of impregnation compares well with stacks and coils made in past tests [5].



(a) (b) (c)

FIGURE 8. Sections of impregnated stacks. CTD101K (a) Matrimid 5292 (b) and Cyanate Ester CTD425 (c)

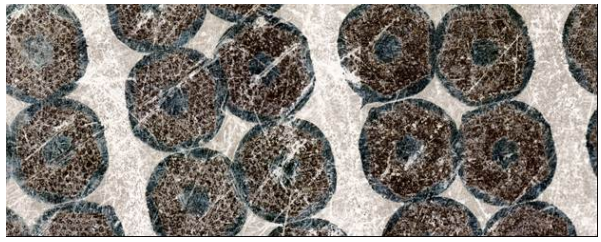


FIGURE 9. Section of 11T dipole coil cross section potted with Matrimid 5292.

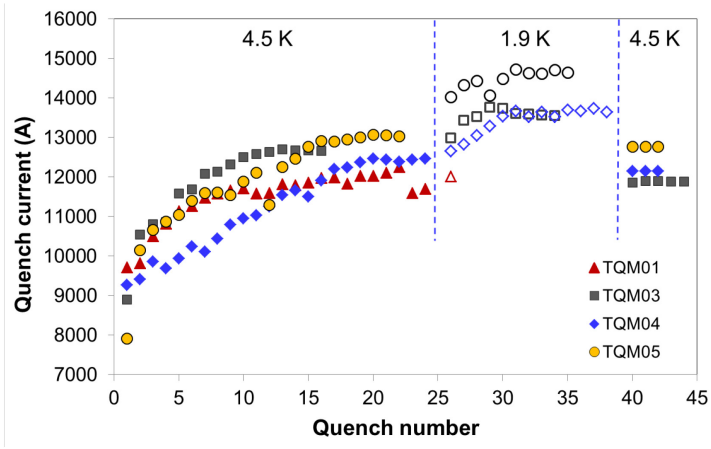


FIGURE 10. Quench plot of Matrimid mirror coil vs CTD101K coils.

In addition, a cross section of the 2 meter long coil (11T dipole) impregnated with Matrimid is shown in Fig. 9. All areas between strands are completely filled with the potting material. The figure demonstrates that the coil potted with Matrimid under actual production conditions has fill characteristics that are as good or better than the samples shown above.

COIL TESTS

The TQ coil potted with Matrimid was tested in a single coil test (mirror) structure. Detailed test results are shown in [4]. Results compare favorably with similar coils made with CTD101K. Figure 10 shows quench results of the Matrimid coil (TQM05) with respect to previous coils made with CTD101K (TQM01-4) tested in the same structure. To date, no coil made with Cyanate Ester CTD425 has been fabricated and tested.

NEXT STEPS

The processes that have been developed and the measurements taken here have allowed some preliminary conclusions to be drawn about the acceptability of each of the alternate potting materials. A series of further measurements will complete this study. More Young's Modulus and Poisson's ratio measurements are needed, even at warm temperatures. The measurements presented here give a reasonable estimate of the values, but are not sufficient in quantity to give a reliable statistical result.

Warm mechanical measurements of Cyanate Ester stacks and an equivalent set of measurements of all three materials at cryogenic temperatures will be completed. Shear and tensile measurements of joints made of the three materials will be added, both warm and cold. Thermal contraction measurements of stacks of the three materials will be completed at helium temperatures. Eventually, stacks made with other cable types and other insulation materials can be added if data concerning specific designs is needed.

More coils will be potted, both with larger cross sections (bigger bore size) and greater length (up to 4-6 meters long). Potting of much longer coils and adapting the potting procedures to achieve good quality will present new challenges. Some will be tested in mirror structures and eventually in full magnets.

CONCLUSIONS

The baseline potting material, CTD101K, has proven to be an acceptable material for impregnating coils, but higher radiation environments in the future may make the use of other materials necessary. This paper represents a first step in an attempt to qualify two additional materials for use in Nb_3Sn accelerator magnets for LARP. Alternative impregnation processes have been used for Rutherford cable and cosine theta coils. Mechanical, electrical and thermal testing of samples has begun. One coil potted with Matrimid has been successfully tested.

The results of this study indicate that either of the alternate materials can be qualified for use in future Nb_3Sn accelerator magnets. Further tests of samples and coils can verify these preliminary conclusions.

ACKNOWLEDGMENTS

The authors would like to thank Marty Whitson, Jesus Alvarez, Lisa Ruiz, Sean Johnson, Armand Bianchi, James Karambis, Steve Gould, Marianne Bossert, Tom Wokas and Robert Jones for technical work preparing and measuring stacks, fabricating coils, potting stacks and compiling data.

REFERENCES

1. Huntsman, "Matrimid 5292A Resin, Matrimid 5292B Hardener," Matrimid A/B Datasheet, Feb. 2005, https://www.huntsmanservice.com/WebFolder/ui/browse.do?pFileName=/opt/TDS/Huntsman%20Advanced%20Materials/English%20US/Long/Matrimid_5292A-B_us.pdf
2. N.A. Munshi, J.K. Walsh, M.W. Hooker, H.K. Babcock, A.H. Haight, S.R. Durso, A. Kawaguchi, P. Hough, "Radiation Resistant Electrical Insulation Qualified for ITER TF Coils," *Applied Superconductivity, IEEE Transactions on* , vol.23, no.3, pp.7700104,7700104, June 2013
3. R. Bossert, et al., "Fabrication and test of 90-mm Nb_3Sn quadrupole coil impregnated with MATRIMID – bismaleimide based material*",* in *IEEE Trans. on Appl. Supercond.*, Vol. 23, N 3, 2013, p. 400-405.
4. G. Chlachlidze et al., "Test Results of a Nb3Sn Quadrupole Coil Impregnated with Radiation Resistant Matrimid 5292", presented at IPAC 2013.
5. S. Krave, "Estimate of time required to vacuum impregnate an Accelerator Magnet Coil", Fermilab Technical Division Technical memo TD-13-007.
6. G. Ambrosio, et al., "Long Quadrupole Design Report", LARP report available at: https://dms.uslarp.org/MagnetRD/longquad/LQ_DR.pdf.
7. R. Bossert, et al., "Development and Test of LARP Technological Quadrupole Models of TQC Series, in *IEEE Trans. On Applied Superconductivity,* Volume 18, Issue 2, June 2008, pp. 175-178.
8. G. Chlachidze et al.,"Quench Performance and Field Quality of 90mm Nb3Sn Quadrupoles of TQC Series", proceedings of IPAC 2012, New Orleans Louisiana, 2012 and Fermilab-conf-12-170-td.
9. S. Caspi, et al., "Test Results of LARP Nb3Sn Quadrupole Magnets Using a Shell-based Support Structure (TQS)," *IEEE Trans. on Applied Supercond.*, vol. 19, no. 3, pp. 1221 – 1225, Jun. 2009.
10. G. Ambrosio, et al., Test Results and Analysis of LQS03 Third Long Nb3Sn Quadrupole by LARP", IEEE Trans. Applied Superconductivity, Volume: 23, Issue: 3 , Part: 2, pp. 4002204, June 2013
11. F. Nobrega, et al., "Magnetic Mirror Structure for Testing Shell-type Quadrupole Coils", presented at MT21, Hefei, China, Oct, 2009.
12. R. Bossert et al., "Fabrication and Test of 4m long Nb3Sn quadrupole coil made of RRP-114/127 Strand", in *AIP Conference Proceedings*, Volume 1434, pp. 869-876, 2012
13. Personnal communication, I. Novitski, FNAL and P. Ferracin, CERN.
14. D. Chichili et al., "Investigation of alternative materials for impregnation of Nb_3Sn magnets," *IEEE Trans. on Appl. Supercond.*, Vol.13, N 2, 2003, p. 1792.
15. R. Bossert et al., "Tests of Insulation Systems for Nb3Sn Wind and React Coils", in *AIP Conference Proceedings* Volume 986, pp. 161-168, 2008.
16. Strain Gage Technology: Thermal Expansion Measurement," Tcchnical Note, TN-513, MICRO MEASUREMENTS GROUP.