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Development and Study of Nb_3Sn Wires with High Specific Heat

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***Abstract*—Recently a new type of Nb_3Sn conductor was developed, which, by incorporating substances with high specific heat at 2-8 K in a proper design, has demonstrated significant improvement of minimum quench energy (MQE). This approach to improving conductor stability is promising to reduce Nb_3Sn magnet training. This work continues studying this type of conductor. Voltage-current (*V-I*) tests from 15 T to 0 T and voltage-field (*V-B*) sweeps at various currents were conducted to investigate the influence of increased specific heat on the intrinsic stability. MQE and normal zone propagation velocity (NZPV) were also measured. Finally, a scheme was put forward to introduce new types of substances with not only high specific heat but also high thermal diffusivity to Nb_3Sn conductors, which is expected to work more effectively.**

***Index Terms*— Nb_3Sn conductor; minimum quench energy; normal zone propagation velocity; specific heat; stability.**

I. Introduction

NB_3SN dipole and quadrupole magnets are well known to suffer from long training. Typically, the first quenches start at 60-70% of the short sample limits, and more than 20 quenches are needed to reach the magnet nominal fields [1], [2]. This leads to significant increases in the cost and magnet test time before installation in an accelerator. For a machine such as the proposed Future Circular Collider (FCC) that requires thousands of dipole and quadrupole magnets, reducing the number of training quenches is highly desirable.

On the other hand, significant (~50%) improvement of critical current density (J_c) of present state-of-the-art Nb_3Sn wires is also needed for 16 T dipoles [3], [4]. Recent development in the new artificial pinning center (APC) technique [5] has shown great promises to deliver such a high J_c [6]. However, higher J_c would inevitably lead to higher instability in conductors. Thus, a new technique is desired to stabilize the new high-J_c conductors.

The quenches of Nb_3Sn magnets are known to be caused by thermal perturbations due to conductor motion or epoxy cracking, etc. As the perturbation energy is larger than the conductor enthalpy margin in the minimum propagation zone (MPZ), a thermal runaway can occur, causing a magnet quench. This enthalpy margin is typically characterized by the minimum quench energy (MQE), which in the adiabatic condition equals to an integration of the conductor's specific heat in the MPZ over the temperature margin.

One way to reduce magnet training is certainly to understand the mechanism and reduce the perturbations (e.g., [7]). The other way is to increase MQE of conductors so that they are more tolerant to perturbations. Since increasing the temperature margin is difficult due to the $J_c(B,T)$ limitation, one promising way to improve conductor MQE is to improve their specific heat (C). This can be realized by introduction of high-C substances. This idea dates back to the 1960s [8], but the major obstacle has been implementation of this idea in a practical way. Keilin's group first demonstrated its effect in proof-of-principle experiments in 2004-2009 [9], but the used schemes were not feasible for long practical conductors. Recently at Fermilab we proposed a design compatible with present high-J_c multifilamentary Nb_3Sn wires. Taking advantage of the fact that billets of such wires are made by stacking a number of subelements and Cu hexagons into Cu cans, this new design simply replaces some Nb_3Sn subelements and Cu hexagons by Cu tubes filled with high-C powders. Since most high-C substances have low thermal diffusivity, we also proposed mixing them with Cu powder, so that Cu can provide a rapid path for heat diffusion from the outer Cu matrix into the whole high-C filaments.

The first Nb_3Sn conductor based on this new design was fabricated using tube type filaments [10]. This wire used Gd_2O_3 as the high-C material. However, the ratio of Cu powder to Gd_2O_3 powder in that wire turned out to be too low, and Cu only formed isolated islands inside Gd_2O_3. Due to the low thermal diffusivity of Gd_2O_3, it is expected that in a short duration only a thin layer of Gd_2O_3 at the high-C filament surface (i.e., that is in contact with the Cu matrix) was involved in heat absorption. Measurements showed that residual resistivity ratio (RRR) or non-Cu J_c was not negatively affected by addition of high-C filaments [10]. Despite unoptimized Cu/Gd_2O_3 ratio and high-C filament positioning [10], it was remarkable to see that the minimum quench energy (MQE) was already tripled due to increased specific heat [10].

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In this work we continue studying the properties of the high-*C* wire, including the voltage-current (*V-I*) stability at different fields and the voltage-field (*V-B*) stability at different transport currents as well as MQE and normal zone propagation velocity (NZPV), and compare them with those of the control wire.

Since Gd_2O_3 has low thermal diffusivity due to its ceramic nature, we are actively searching for alternate high-*C* materials with higher thermal diffusivity. There is a large inventory of substances with high specific heat below 10 K. Some of these substances, such as Re-Cu compounds (where Re is a rare earth element) [11], are intermetallics with much higher thermal diffusivity than Gd_2O_3. However, the key difficulty for using these substances is that they are not commercially available. In this work a scheme is proposed to form Re-Cu compounds in Nb_3Sn conductors.

II. Studies of the Wire with Gd_2O_3

The details of the high-*C* and the control wires can be found in earlier work [10]. Both wires have filament size of ~70 µm. The strands were heat treated on standard ITER barrels at 645 °C for 120 hours in flowing argon gas.

A. Study of V-I and V-B Stability

For the stability tests, the current and field ramp rates were kept the same for both samples. The results are shown in Fig. 1. The non-Cu J_c values of the two samples are more or less the same. For *V-I* tests the control wire had superconducting transitions at 12 T and above, but quenched at 10 T and lower fields. In contrast, the high-*C* wire still had a good transition at 10 T, but began to quench at 8 T. Below 8 T, the quenched current density (J_q) values of the high-*C* wire are clearly higher than those of the control wire. The *V-B* sweeps between 0 and 4 T also showed higher J_q for the high-*C* wire.

As mentioned earlier, the high-*C* wire has insufficient Cu powder for transporting heat from high-*C* filament surfaces inward, so the time constant for a high-*C* filament to absorb heat is large. By optimizing the Cu/Gd_2O_3 ratio so that heat can be quickly transported to Gd_2O_3 in the whole high-*C* filaments through continuous Cu paths, it is expected that the effect of stabilizing the conductors with transport current will be more pronounced. This will make this technique very useful for counteract the instability in high-J_c conductors.

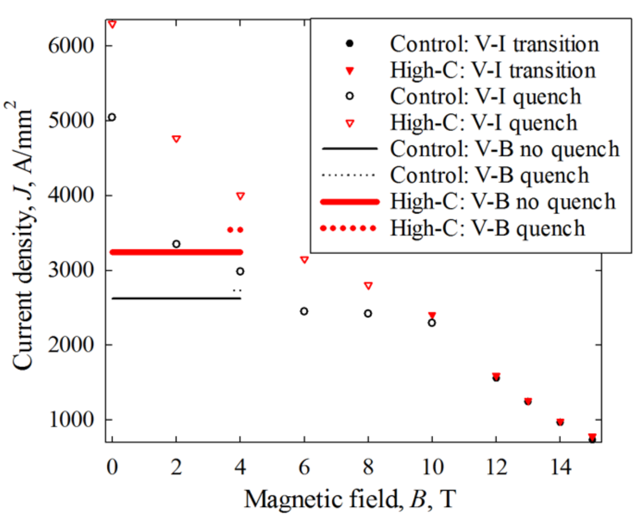

Fig. 1. Comparison of *V-I* and *V-B* test results of the control and high-*C* wires.

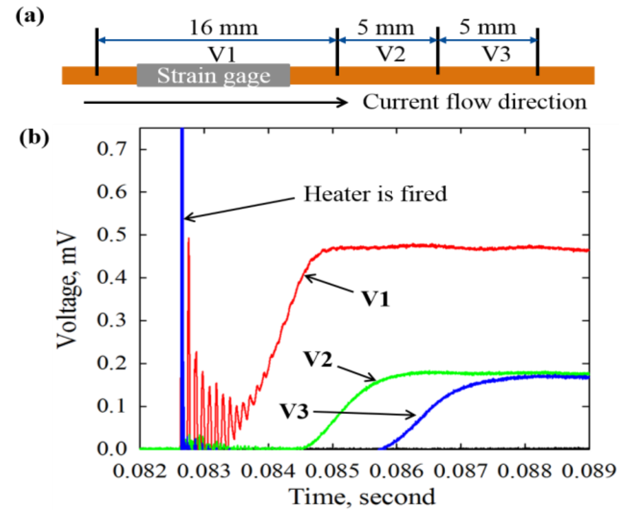

Fig. 2. (a) Schematic of a setup to measure MQE and NZPV of wires, and (b) a typical example of voltage development in the sample after a quench occurred.

B. MQE and NZPV measurements

A schematic of the experiment setup is shown in Fig. 2 (a). Strain gages of ~4 mm pattern length were used as the heaters, as described in detail in [10]. Three pairs of voltage taps were used to monitor the propagation of the normal zone, with V1 across the strain gage, V2 and V3 in the downstream of the current flow. For the measurements, first the I_c at a certain field (e.g., 15 T) was measured, and then the current was fixed at a certain I/I_c (in this work 40%, 60%, and 80% were measured), and then the power to the strain gage (with a duration of 200 µs) was increased gradually in small steps until the normal zone propagated (i.e., the sample quenched). From the input power, the MQE can be calculated.

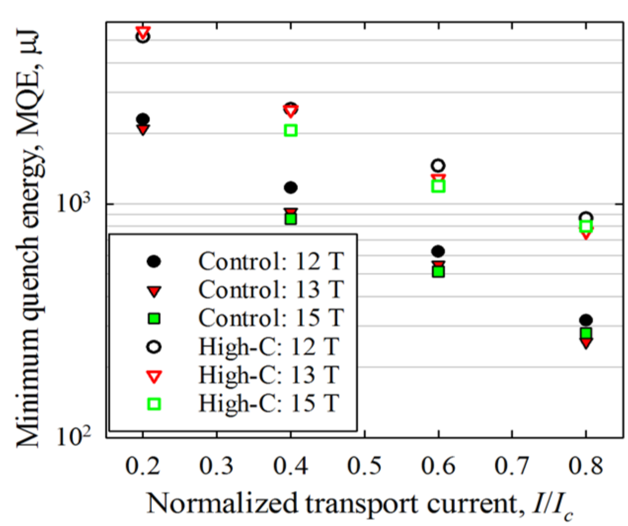

Fig. 3. MQE of the control and high-*C* wires at various fields versus the I/I_c fraction.

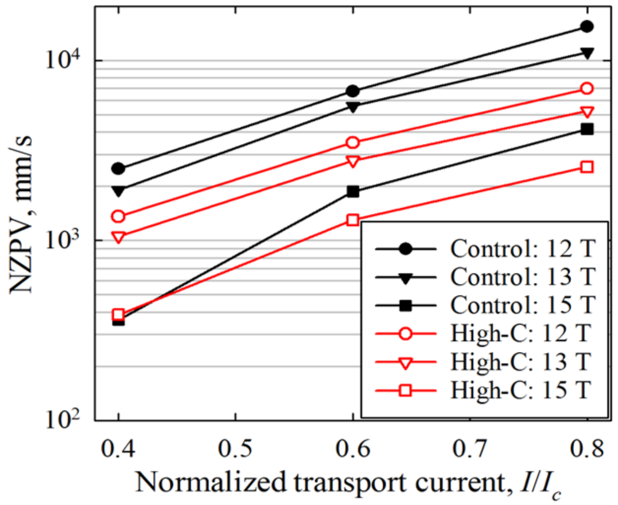

Fig. 4. NZPV of the control wire and high-*C* wires at various fields versus the I/I_c fraction.

As the thermal runaway occurred, first a voltage rise was seen in V1, and then V2 and V3. An example of the voltage development is shown in Fig. 2 (b). The propagation time between V2 and V3 is taken as the spacing between the middle of the transitions. The NZPV was calculated by dividing 5 mm by the propagation time between V2 and V3. The measured MQE and NZPV results versus I/I_c for the control sample and the high-*C* sample at different fields are shown in Figs. 3 and 4, respectively.

From Fig. 3 it is seen that the MQE is not sensitive to the field, perhaps because decrease in temperature margin and increase in specific heat as field increases are counter balanced. The MQE values of the high-*C* wire are dramatically higher than those of the control one, and the gain is higher at higher I/I_c, reaching a factor of ~3 at I/I_c of 80%, agreeing well with the previous results [10]. The increased MQE may help to reduce magnet training: e.g., in Fig. 3, with a perturbation of 1 mJ, the control wire quenched at I/I_c of ~40%, but the high-*C* wire survived until I/I_c reached ~80%.

From Fig. 4 it is seen that the NZPV was reduced due to increased specific heat. The reduction of NZPV was by ~50% at 12-13 T, but was less pronounced at 15 T: by ~30% for I/I_c of 60% and 80%. On the other hand, even though the NZPV was reduced by 30-50%, it was still on the magnitude of a few meters per second, which is still in the comfortable region for quench protection of magnets.

C. Inclusion of high-C substances into superconducting subelements

The above scheme of replacing outmost-layer subelements by high-*C* filaments is effective to intercept the heat from outside, and also helps to improve intrinsic stability. To further stabilize high-J_c wires, we also consider adding high-*C* substances into each Nb_3Sn subelements. One way is to integrate them into the subelement cores. This can be easily done for powder-in-tube (PIT) conductors, in which the high-*C* powder can just be blended with the Sn source powders (e.g., Sn+Cu) in the cores. After full heat treatments, low-Sn bronze with the high-*C* particles embedded is left in the cores, which can absorb heat quickly from the subelements. The effectiveness of this scheme will be studied.

III. A Scheme to Form Re-Cu Compounds

Taking advantage of the fact that Nb_3Sn wires require heat treatments at 625-700 °C for typically 50-300 hours to react Nb and Sn to form the superconducting Nb_3Sn phase, we propose using the commercially available Cu and rare earth metal powders, which can react to form Re-Cu compounds during the heat treatment. Similar to the structure in which a mixture of high-*C* powder (e.g., Gd_2O_3) and Cu powder is filled into a Cu tube to make a high-*C* filament, we can mix Re and Cu powders and fill the mixture into a Cu tube. Here an example of $HoCu_5$ is given, but other Re-Cu compounds such as $DyCu_5$, $ErCu_5$, or $CeCu_6$ can be formed using the same technique. According to the Ho-Cu phase diagram [12], Ho and Cu can form several line compounds, such as HoCu, $HoCu_2$ and $HoCu_5$, among which $HoCu_5$ is the most Cu-rich compound that can be formed. This means, if we select a Cu/Ho molar ratio larger than 5, they will form $HoCu_5$ with some Cu powder left, which can be used to transport heat among $HoCu_5$ particles. Another important fact is that Ho does not have solubility in Cu, an important feature assuring that RRR of the Cu matrix will not be affected due to Ho addition. This is also true for other rare earth elements. Another advantage of the above scheme is that it uses Ho and Cu powders, which are both metals with good ductility and can be elongated during wire drawing. Thus, this scheme is expected to lead to good wire drawing property.

One question with the above scheme is whether Ho and Cu can finish the reaction to form $HoCu_5$ within the heat treatment duration for Nb_3Sn conductors. In order to find this out, we fabricated a Nb_3Sn wire with Ho filaments, which were made by filling pure Ho powder (instead of the above-mentioned Cu and Ho powder mixture) into a Cu tube, with Ho particle size below 44 μm. Scanning electron microscopy (SEM) image of the unreacted wire is shown in Fig. 5(a). In this 61-restack wire, the six corners were Cu rods, and the rest of the outmost layer was occupied by 18 Cu-clad Ho filaments, and there were 36 Nb_3Sn filaments based on the tube type design. The billet for the wire was 19 mm in diameter, and was drawn to 0.7 mm without any breakage, demonstrating the good drawability of Ho powder. At the final size, in each Cu-clad Ho filament, the Ho core had a diameter of ~40 μm, and

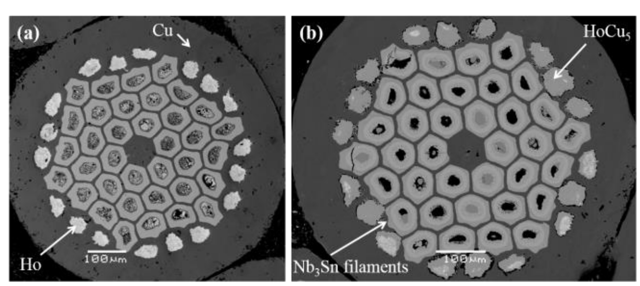

Fig. 5. SEM images of (a) the unreacted wire with Ho filaments, and (b) the wire after reaction at 640 °C for 120 hours.

the Cu/Ho area ratio is ~3.0, corresponding to a Cu/Ho molar ratio of 7.9. A reference wire was also fabricated, with the Ho filaments replaced by pure Cu rods.

Both wires were reacted in flowing argon at 640 °C for 120 hours (with a ramp rate of 50 °C/h), with SEM image of the reacted wire with Ho filaments shown in Fig. 5(b). It was found that the Nb_3Sn filaments were still not fully reacted due to insufficient reaction time: there was still Nb_6Sn_5 phase in the core, indicating that the wire was still in the early stage of reaction based on previous studies [13]. On the other hand, the Cu-clad Ho filaments had been fully reacted. Based on the energy-dispersive spectroscopy (EDS) measurements of the reacted Ho filaments, most of the Ho powder had turned into $HoCu_5$ (with the average composition of 19 at.%Ho-81at.% Cu). This indicates that the reaction time needed for Nb_3Sn formation is more than enough for the formation of $HoCu_5$. It is worth pointing out that if a mixture of Ho and Cu powders is used instead of pure Ho powder, the reaction time to convert Ho should be much shorter because the Ho-Cu diffusion distance is much shorter. It can also be seen from Fig. 5(b) that there are some voids around the formed $HoCu_5$, indicating that Cu is the primary diffusing species in the Ho-Cu diffusion reaction couple, which causes the volume of Ho to expand after turning into $HoCu_5$.

The heat capacity of a Cu-clad Ho filament after heat treatment was measured, and the calculated volumetric specific heat of $HoCu_5$ is shown in Fig. 6, along with the data of Cu and Gd_2O_3 [10]. It can be seen that although the specific heat of $HoCu_5$ is lower than that of Gd_2O_3 at 2 K, the former is several times higher than the latter at 6-8 K. Since the enthalpy is an integration of specific heat from the bath temperature (1.9 K or 4.2 K) to the current-sharing temperature (depending on the operational field and current, but is typically above 6 K), it is expected that the specific heat increase by using $HoCu_5$ is at least comparable to that by using the same volume of Gd_2O_3. It is also known from literature that the specific heat of $HoCu_5$ is not sensitive to magnetic field [11].

The magnetization versus field (*M-B*) loops of the control wire and the wire with Ho filaments after reaction were

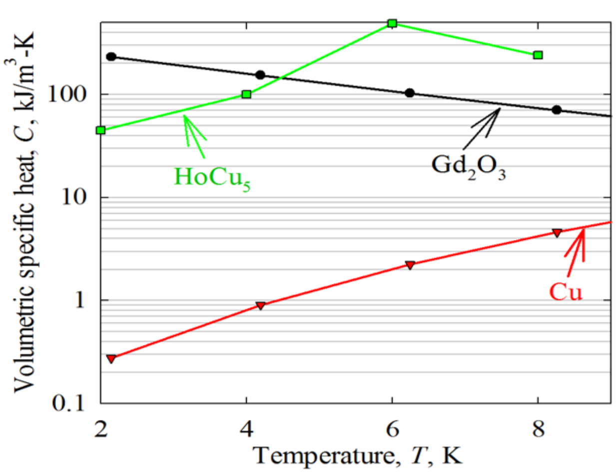

Fig. 6. Measured specific heat of $HoCu_5$ in comparison with Cu and Gd_2O_3.

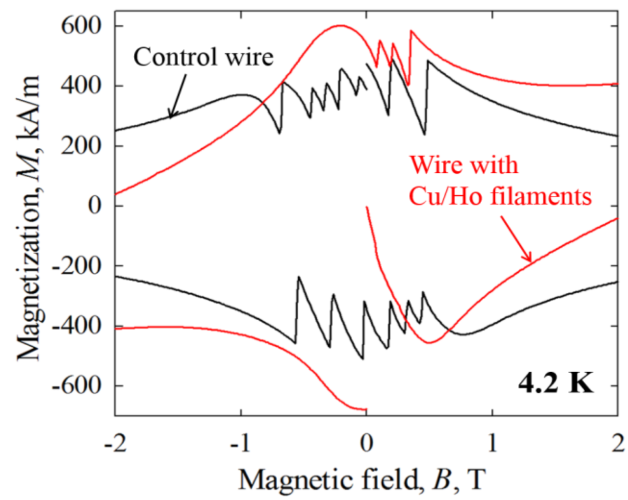

Fig. 7. *M-B* loops of the control wire and the wire with Ho filaments (with magnetizations normalized to the total volumes of Nb_3Sn filaments).

measured and are shown in Fig. 7. It can be seen that the control wire had wider flux jump ranges and slightly larger flux jump amplitudes. The *M-B* loop of the wire with Ho filaments is "tilted", most likely because $HoCu_5$ has a large paramagnetic susceptibility, which is not expected to affect wire stability. It was noticed that the control wire had 5-10% higher non-Cu J_c than the wire with Ho filaments. This could be due to additional magnetic fields generated by the paramagnetic $HoCu_5$ itself, because the $HoCu_5$ volume fraction in the wire was large, ~10 vol.%. Next, we will use mixture of Ho and Cu powders instead of pure Ho powder, aiming to reduce $HoCu_5$ fraction to 2 vol.%, which will improve the specific heat of a wire by 2-3 times at 2-6 K.

IV. Summary

The *V-I* and *V-B* tests show that increased specific heat can clearly improve intrinsic stability of Nb_3Sn wires. The MQE of the high-*C* wire was improved by nearly 3 times, whereas the NZPV was reduced only by 30-50%. To further stabilize high-J_c wires, we also consider adding high-*C* substances into each Nb_3Sn subelements by integrating them into the subelement cores. A new design is also proposed to form Re-Cu compounds as high-*C* substances in Nb_3Sn wires, which has an advantage of higher thermal diffusivity than Gd_2O_3. The feasibility of this scheme has been demonstrated by preliminary studies.

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