NIOBIUM-TITANIUM: WORKHORSE SUPERMAGNET MATERIAL*

T.G. Berlincourt

WHY SUPERMAGNETS?

In medical magnetic resonance imaging (MRI) patients are placed in intense magnetic fields provided in nearly all instances by superconducting electromagnets (supermagnets). Why supermagnets? Because electric current flows through the superconducting windings of a supermagnet without electrical resistance. As a consequence, the only power needed to maintain the current and magnetic field of a supermagnet is the amount needed to keep the supermagnet refrigerated to a few degrees above absolute zero. In contrast the copper windings of a conventional electromagnet present a resistance to the flow of electric current. The power needed to counter that resistance greatly exceeds the power required to refrigerate a supermagnet. Without supermagnets MRI imaging would be an order of magnitude more expensive. Similar advantages accrue in the use of supermagnets to bend charged-particle beams in giant high-energy particle-beam accelerators such as the Tevatron and the Large Hadron Collider (LHC).

FROM AMONG THE MANY THOUSANDS OF SUPERCONDUCTORS, HOW DID NIOBIUM-TITANIUM HAPPEN TO EMERGE AS SUPERMAGNET WORKHORSE?

When cooled to sufficiently low temperatures almost all metals become superconducting, i.e., they present no resistance to the flow of an electric current. This phenomenon was first observed in 1911 in Holland by Kamerlingh Onnes when he cooled a frozen-solid mercury specimen to within a few degrees of absolute zero. Early attempts to take advantage of this phenomenon to make supermagnets were frustrated when it was found that, for the metals studied during that era, a high current, or a modest magnetic field, or a combination thereof destroyed superconductivity. Stated another way, temperature, electric current, and magnetic field are all enemies of superconductivity. So, ideally, what is required for realization of a useful supermagnet? The answer is (a) a metal that remains superconducting at a temperature high enough for refrigeration to be convenient and economical, (b) a metal that can support large electric current densities without dissipation in the presence of a high magnetic field, (c) a metal that is ductile and easily fabricated, and (d) a metal that is affordable. Fifty years were to pass before something approaching this ideal would emerge. Then on January 9, 1961, *Physical Review Letters* (*PRL*) received a manuscript from Bell Telephone Laboratories (BTL) researchers Kunzler, Buehler, Hsu, and Wernick. In that manuscript they reported that the compound Nb_3Sn was capable of sustaining enormous electric supercurrent densities (100,000 amperes/cm^2) without resistance in very high magnetic fields (at least as high as 8.8 tesla).

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*A layman-level account of the Nb-Ti saga transmitted to Case Western Reserve University at its request. The last section "Correction of the Historical Record on Niobium-Titanium Alloys" is for the benefit of the fact-checking editors who could become hopelessly confused by referring to the scientific, popular, or internet literature. That section is not to be included in any publication which might result. Comments are welcomed at <tgberlincourt@aol.com>.

Not yet aware of that spectacular BTL discovery, Atomics International (AI) researchers Berlincourt and Hake were at that time also experimenting on superconductors of possible interest for application to supermagnets. Earlier, in 1957, Berlincourt had observed superconducting critical fields in uranium-molybdenum alloys high enough to eclipse a record that had stood for 27 years. Within two years that new record had been toppled by Hake in an investigation of titanium-molybdenum alloys. Now on January 12 and 13, 1961, the AI pair performed critical supercurrent density measurements on a number of superconductors in magnetic fields up to 3 tesla. Included were Nb_3Sn and the alloys tantalum-titanium and vanadium-titanium. The observed critical-supercurrent densities ranged up to 560 amperes/cm^2 at 3 tesla. Current densities of that magnitude are typical in copper-wire electromagnets, and so those results were viewed as mildly encouraging. But, curiously, the results did not vary appreciably from specimen to specimen, nor did they increase appreciably with decreasing temperature. This suggested that the contacts between the specimens and the copper wires feeding current to them were faulty. Not having anticipated enormous critical-supercurrent densities, Berlincourt and Hake had not made adequate provisions for measuring them. While efforts were underway to rectify this deficiency, the February 1, 1961, issue of *PRL* arrived at AI with news of the BTL breakthrough.

That BTL breakthrough revealed that Nb_3Sn fulfills most of the ideal requirements listed above. It also revealed that Nb_3Sn is inconveniently brittle and difficult to fabricate. No matter; it ushered in the supermagnet era. Although the BTL discovery appeared to many to be a bolt out of the blue, there had been many hints of what was to come, and, as noted above, other researchers had been close on BTL's heels. The Nb_3Sn advance intensified the search for additional useful supermagnet materials.

Subsequently, on April 24, 1961, BTL filed applications for patents on alloy superconductors. Based on critical-supercurrent-density measurements in magnetic fields up to 8.8 tesla, Matthias filed for niobium-titanium alloys, and Kunzler and Matthias together filed for niobium-zirconium alloys. Unaware of these new BTL developments, Berlincourt and Hake had independently made critical-supercurrent-density measurements on the very same alloys (as well as numerous others) in magnetic fields up to 3 tesla. With their current-contact problem behind them, they measured niobium-titanium alloys on April 17, 1961, and niobium-zirconium alloys on April 19, 1961. At both institutions the very-tough-and-difficult-to-fabricate niobium-zirconium alloys supported very high critical-supercurrent densities, whereas the observed critical supercurrent densities for the ductile-and-easy-to-fabricate niobium-titanium alloys were discouragingly low. In light of those results AI filed a patent application for the niobium-zirconium alloys but not for the niobium-titanium alloys. A contentious patent interference battle ensued over niobium-zirconium, with the patent ultimately being awarded to BTL in 1966. AI was left with a non-exclusive, royalty-free license. Small niobium-zirconium supermagnets soon became commonplace in research applications, and niobium-titanium faded into the background, seemingly destined for obscurity.

Ironically, prior to the Nb_3Sn breakthrough, the fundamental theoretical physics describing this remarkable new class of superconductors had already been developed by USSR physicists, Ginzburg, Landau, Abrikosov, and Gor'kov (GLAG). However, neither they, nor apparently anyone else, realized that those theories might account for the extremes of critical magnetic field and critical supercurrent density observed for Nb_3Sn. Indeed, the remarkable BTL observations were initially interpreted universally in terms of the now-discredited Mendelssohn sponge model. Then, several months after the BTL revelation, Goodman (Institut Fourier, Grenoble) reported remarkable agreement between GLAG theoretical predictions and the critical magnetic fields Berlincourt had measured years earlier for uranium-molybdenum alloys. Goodman's observation was ignored by most superconductivity researchers, but Berlincourt and Hake regarded it seriously and began a series of measurements on a multitude of transition-metal alloys. In this round of investigations they were equipped with a pulsed, copper-coil magnet allowing experimentation in magnetic fields up to 16 tesla, i.e., well above the 8.8-tesla field available at BTL. The new AI investigations firmly established GLAG as the applicable theory for the new high-magnetic-field materials. As might have been anticipated, however, publication of the AI findings was delayed for months by scientific-journal referees still enamored of the sponge model. Forty-one years later, in 2003, Ginzburg and Abrikosov were awarded Nobel Prizes for their contributions to GLAG.

Significantly, of all the transition metal alloy superconductors Berlincourt and Hake studied in the new round of experiments, niobium-titanium alloys exhibited the highest critical magnetic field (followed ever so closely by tantalum-titanium alloys and trailed by niobium-zirconium alloys). The high critical magnetic field for the niobium-titanium alloys, 14.5 tesla, was a clue that they might after all be useful for supermagnet windings despite the discouraging critical-supercurrent-density results obtained earlier at both BTL and AI. Accordingly, armed with the greater basic understanding provided by GLAG, encouraged by knowledge of the high critical magnetic field of niobium-titanium alloys, and tantalized by their highly-ductile, easy-to fabricate nature, Berlincourt and Hake reasoned that, with appropriate metallurgical treatment, niobium-titanium alloys might be made to support large superconducting critical current densities of practical interest. They soon found that extremely severe cold working was highly effective and resulted in superconducting critical current densities thirty times greater than had been observed in the earlier studies at BTL and AI. Thus, Berlincourt and Hake demonstrated conclusively the suitability of niobium-titanium alloys for supermagnets capable of generating magnetic fields greater than 10 tesla. All of this was reported at the April 1962 Washington, DC meeting of the American Physical Society. Notified of these advances, AI's patent counsel did a patent search. It was only then that the AI researchers learned of the pending Matthias niobium-titanium patent.

Almost immediately, niobium-titanium alloys became the workhorse supermagnet materials, and they have since remained so. However, it should be emphasized that this is largely a consequence of their ductility and ease of fabrication. Numerous uncooperatively-brittle materials have much-superior superconducting properties. For example, Nb_3Sn has a superconducting transition temperature twice as high (easing refrigeration requirements), has a critical magnetic field twice as high (enabling fabrication of higher-magnetic-field supermagnets), and is capable of supporting higher critical-supercurrent densities (facilitating more-compact supermagnet windings). In addition, there is now a whole new class of brittle ceramic "high-temperature superconductors" discovered in 1986 by Bednorz and Muller (IBM, Zurich). Superconductors of that class have since been shown to have superconducting transition temperatures fifteen times that of niobium-titanium alloys and are thought to have critical magnetic fields approaching 100 tesla. Unfortunately they are extremely difficult to prepare and fabricate into supermagnets, and to date their demonstrated ability to support critical supercurrents has been very disappointing, severely limiting their application. And so niobium-titanium continues to reign as the billion-dollar tonnage champion. But for how long is anyone's guess.

CORRECTION OF THE HISTORICAL RECORD ON NIOBIUM-TITANIUM ALLOYS*

In the scientific and popular literature, and on the internet, the role of Westinghouse in niobium-titanium alloy research is often misrepresented. While it is true that, by April 19, 1961, Hulm and Blaugher, at Westinghouse Research Laboratory (WRL), had also independently engaged in research on niobium-titanium alloys, their experiments were confined to measurements of superconducting transition temperatures. Because they made no determinations of critical supercurrent density as a function of magnetic field, they had no way to judge the potential of niobium-titanium alloys for application in supermagnets. Nevertheless, once proof of principle had been established at AI, Westinghouse was among the first to commercialize niobium-titanium alloys and was the first to achieve 10 tesla in a supermagnet with niobium-titanium windings.

Paternity for niobium-titanium alloys is also often erroneously attributed to developers of high-energy-particle accelerators and detectors, or to manufacturers of MRI imagers. In fact, those sectors made no significant contributions during the discovery phase of high-magnetic-field, high-critical-supercurrent-density superconductivity. However, to their great credit they figured most prominently in the nearly fifty years of highly-sophisticated optimization and engineering that followed. Today's Tevatron, Large Hadron Collider, and MRI imagers are monuments to their heroic accomplishments. Also, it should be noted that the Atomic Energy Commission (AEC), which was the major funder of high-energy physics in the United States in 1961, was also the major funder of the AI superconductivity research effort through a contract directed toward exploration of the basic electronic properties of nuclear reactor materials. Ironically, this research contributed more to the advancement of high-energy-particle physics than to the advancement of nuclear reactors. Additional funders of the AI superconductivity research effort included the Air Force Office of Scientific Research, and the Independent Research and Development Program of AI's parent company, North American Aviation, Inc.

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Based on information available at this writing, the significant events in the establishment of the suitability of niobium-titanium alloys for use in high-performance supermagnets are as follows:

1. Observation of superconductivity in niobium-titanium alloys:
    AI April 17, 1961
    BTL Before April 24, 1961
    WRL Before April 19, 1961

2. Measurement of superconducting transition temperatures for niobium- titanium alloys:
    WRL Before April 19, 1961

3. Measurement of discouragingly-low, critical-supercurrent densities as a function of magnetic-field strength in mildly-cold-worked niobium-titanium alloys:
    AI April 17, 1961
    BTL Before April 24, 1961

4. Measurement of the very-high, superconducting critical magnetic field of niobium-titanium alloys:
    AI Before April, 1962

5. Achievement of very high critical-supercurrent densities at very high magnetic fields for severely cold-worked niobium-titanium alloys, thereby confirming their suitability for application in-high performance supermagnets:
    AI Before April, 1962