Superconducting and Mechanical Properties of Low-Temperature Solders for Joints

Yeekin Tsui, Ra'ad Mahmoud, Elizabeth Surrey, and Damian Hampshire

Abstract—All large superconducting applications require electrical connections between superconductors or between superconductors and other metallic parts of the systems. Such connections are essential components for these applications and are usually made by melting the solder (or soft soldering). In general, the melting point, electrical resistivity, mechanical and wetting properties, and toxicity are all important criteria when choosing the best solder for a specific application. If the solder itself is superconducting under the operating conditions of the application, then there is also the possibility of extremely low resistance joints. In this work, we report on the superconducting properties of six low-melting-point solders, namely, Bi₄₉Pb₁₈In₂₁Sn₁₂ (Cerrolow 136), Pb₂₀Sn₃₄Bi₄₆, Pb20Sn60Bi20, In52Sn48, Pb38Sn62, and Pb57Bi36Sb7, and the mechanical properties of three of the six solders (Pb₃₈Sn₆₂, Cerrolow 136, and $Pb_{20}Sn_{34}Bi_{46}$). We have found that all the solders except Pb₃₈Sn₆₂ and In₅₂Sn₄₈ have relatively high upper critical fields (higher than 2 T) at temperatures lower than 3 K. Although the good ductility and high maximum fracture elongation of Pb₃₈Sn₆₂ is useful when using low-temperature superconductors, other solders may be preferred with high-temperature superconductors because their lower melting temperatures mean they are less likely to delaminate them.

Index Terms—Coated conductor, solder, soldered joint, tensile test.

I. INTRODUCTION

S OLDERED joints are essential components of all large superconducting applications. These applications include superconducting power lines, high-field magnets for MRI machines, nuclear fusion reactors and particle accelerators. To ensure reliable operation of soldered joints, a detailed knowledge and understanding of all the relevant electrical, thermal and mechanical properties of the solder are required. As part of the first screening of solders for joints, we usually consider electrical resistivity, melting point, mechanical strength, wetting properties and toxicity. The importance of each of the solder's properties can differ from one application to another. If high-temperature superconducting coated conductors are involved then the melting point of the solders used must be low. A high

Manuscript received September 8, 2015; accepted February 16, 2016. Date of publication March 1, 2016; date of current version April 25, 2016. This work was supported by the RCUK Energy Program under Grant EP/I501045. The data are available at: http://dx.doi.org/10.15128/gh93gz495.

Y. Tsui, R. Mahmoud, and D. Hampshire are with the Department of Physics, Center for Materials Physics, Superconductivity Group, University of Durham, Durham DH1 3LE, U.K. (e-mail: yeekin.tsui@durham.ac.uk).

E. Surrey is with the European Atomic Energy Community (EURATOM)/ Culham Center for Fusion Energy (CCFE) Fusion Association, Culham Science Center, Abingdon OX14 3DB, U.K.

Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

Digital Object Identifier 10.1109/TASC.2016.2536806

TABLE I MAXIMUM TEMPERATURE USED TO MELT THE SAMPLES AND THE COOLING RATE USED TO FABRICATE THE SOLDER SAMPLES FOR MECHANICAL TESTS

Solder Alloy	Max. Temp. °C	Cooling Rate °C/second	
Pb ₃₈ Sn ₆₂	250	0.100	
Cerrolow-136	75	0.036	
$(Pb_{18}Sn_{12}Bi_{49}In_{21})$			
$Pb_{20}Sn_{34}Bi_{46}$	120	0.060	

soldering temperature can lead to delamination of the coated conductors and change of oxygen content in the superconducting layer. These effects will degrade the superconducting properties of the coated conductors [1]. However for joints using low temperature superconductors, the superconducting materials are much more chemically and structurally stable and melting point of the solder is not so important. The mechanical properties of commonly used solders such as Pb₃₈Sn₆₂ have been studied extensively [2]. However, the superconducting properties of many low melting point solders that are potential materials for joints have not yet studied in detail. In this paper we find that the cooling rate during the solidification of the solder strongly affects its mechanical properties, most probably because of the different microstructures formed [3]. Hence we present a study of both the superconducting and mechanical properties of six low melting point solders, so that with good control of sample preparation, we identify good candidate solders for superconducting applications.

II. EXPERIMENT

A. Sample Preparation

All the samples except the Cerrolow 136 were fabricated in-house at Durham. Cerrolow 136 was obtained from a commercial company. All the constituent elemental metals of each solder were placed into a crucible. The crucible was then heated to well above the melting point of the solder (cf. Tables I and II). Heat was removed when the molten solder was formed and the crucible was allowed to cool down to room temperature in air. The samples for mechanical tests were made by remelting the solders at temperatures well above their melting temperatures. Each sample was then cooled down to room temperature at a constant rate in air. The cooling process was monitored by two thermocouples to ensure there was no thermal gradient across the sample. We found that controlled cooling rates were essential to ensure reproducibility of the experimental results. Table I gives the maximum temperature and cooling rate used during the fabrication of the sample for

See http://www.ieee.org/publications_standards/publications/rights/index.html for more information.

Solder Alloy	Melting Point °C	ρ (room temp) $10^{-7} \Omega \cdot m$	$\rho (77 \text{ K})$ $10^{-7} \Omega \cdot \text{m}$	T _c K	<i>B</i> _{c2} (0К) Т
Bi ₄₉ Pb ₁₈ In ₂₁ Sn ₁₂ (Cerrolow 136)	58	9.4	8.1	6.4	3.3
Pb20Sn34Bi46	~ 96	5.5	2.6	8.4	2.3
$Pb_{20}Sn_{60}Bi_{20}$	~ 170	2.6	1.1	8.5	2.2
$In_{52}Sn_{48}$	118	2.6	1.3	6.4	0.34
Pb ₃₈ Sn ₆₂	183	1.5	0.48	7.3	0.30
Pb57Bi36Sb7	200 - 230	6.1	3.7	8.5	2.5

TABLE II Melting Point, Resistivity, and Superconducting Properties of the Solders Studied

the mechanical tests. The samples were formed as rectangular slabs with typical dimensions of $80 \times 4.4 \times 2.2 \text{ mm}^3$. For ac magnetic moment measurements, a small piece of each solder was cut and mounted in a plastic straw.

B. Experimental Procedures

All mechanical tests were carried out using a Mecmesin Multi-Test 2.5-d tensile & compression test system. A pair of Nyilas-type extensometers [4] were used to measure the elongation of each sample at different strain rates. These very sensitive extensometers consist of a copper-beryllium frame and are equipped with a Wheatstone bridge of strain gauges. The double extensometer configuration also has the advantage of mitigating against experimental errors resulting from sample distortion. Ac. magnetic moment measurements of all the solder samples were performed in a Quantum Design Physical Properties Measurement System in magnetic fields up to 2 T and at temperatures from room temperature down to 2 K. The resistivity of the solders was measured using the standard dc four-terminal method.

III. RESULTS AND DISCUSSIONS

A. Superconducting Properties

 $B_{c2}(T)$ of Cerrolow 136, Pb₂₀Sn₃₄Bi₄₆, Pb₂₀Sn₆₀Bi₂₀ and Pb₅₇Bi₃₆Sb₇ was determined from the ac magnetic moment measurements. The values of $B_{c2}(T)$ for these solders were obtained from the onset of the superconducting transition in decreasing temperature. Fig. 1 shows typical ac magnetic moment against temperature curves at different applied magnetic fields for one of the solders studied. $B_{c2}(T)$ of Pb₃₈Sn₆₂ or In₅₂Sn₄₈ was obtained from resistivity measurements. Typical resistivity data for $Pb_{38}Sn_{62}$ are shown in Fig. 2 $B_{c2}(0)$ and the transition temperature (T_c) were obtained by fitting the $B_{c2}(T)$ data with the Werthamer-Helfand-Hohenberg (W-H-H) equation [5] as shown in Fig. 3. Melting point, resistivity, $B_{c2}(0)$ and T_c of all solders are summarized in Table II. $Pb_{38}Sn_{62}$ or $In_{52}Sn_{48}$ have $B_{c2}(T)$ values almost 10 times lower than the other solders and relatively low T_c values. The solders with higher $B_{c2}(0)$ values open the possibility of very low resistance joints if the joints are located in low field regions for a given application. Some also have substantially lower melting points (i.e. all except Pb₅₇Bi₃₆Sb₇) which can ensure there is no degradation for example to HTS coated conductors during the



Fig. 1. Ac. magnetic moment against temperature in different applied magnetic fields for $Pb_{57}Bi_{36}Sb_7$. Lines are guides to the eye.



Fig. 2. Normalized resistivity of the $Pb_{38}Sn_{62}$ solder as a function of magnetic field at different temperatures. Lines are guides to the eye.



Fig. 3. Upper critical field as a function of temperature for $Pb_{57}Bi_{36}Sb_7$. Curves are fitted using the W-H-H equation.

soldering process. Equally if low temperature superconductors are being used and the operating conditions for the joints are at high magnetic field, then the soldering temperature need not be important and $Pb_{38}Sn_{62}$ may be a better solder to use because of its much lower resistivity and better mechanical properties.

B. Mechanical Tests

Fig. 4 displays a typical stress-strain curve for the samples measured in this work. The elastic modulus, tensile strength



Fig. 4. Stress–strain curve for $Bi_{49}Pb_{18}In_{21}Sn_{12}$ at a strain rate of 0.053 %/ second.



Fig. 5. Stress versus strain at different strain rates for Pb₂₀Sn₃₄Bi₄₆.

and fracture elongation were extracted from these curves at different strain rates for each sample (see Fig. 4). Fig. 5 shows the stress/strain curves for Pb₂₀Sn₃₄Bi₄₆ at different strain rates which shows the strain rate has a substantial effect on the solder's mechanical properties. The figure also demonstrates the good reproducibility obtained for nominally identical samples. The high strain rate dependence of mechanical properties is well established for both Pb-Sn and Pb-free solders in the literature [6]. Figs. 6 and 7 show the elastic modulus (E) and tensile strength (σ_T) against strain rate for all three solders. Among these 3 materials, Pb₃₈Sn₆₂ has the best mechanical properties then Cerrolow and finally Pb₂₀Sn₃₄Bi₄₆. The values of E and σ_T differ by ~30% for Pb₃₈Sn₆₂ and Cerrolow 136. Figs. 8 and 9 display the fracture elongation of the samples as a function of strain rate. In Fig. 8, the fracture elongation was measured using the whole length (\sim 44 mm) of the sample between the grips of the stress/strain measuring machine after failure. Whereas in Fig. 9, the maximum fracture elongation was measured in that part of the samples (25 mm in length) outside of the breaking point of the sample. Both of these figures show the fracture elongation of $Pb_{38}Sn_{62}$ is several times higher than that of Cerrolow 136. We conclude that the much lower values in Fig. 9 compared to Fig. 8 are much more relevant and useful for practical assessment of the best solder for joints. Consistent with our extensive experience making



Fig. 6. Elastic modulus versus strain rate for three solders on a log-log scale. Lines are guides to the eye.



Fig. 7. Tensile strength versus strain rate for three solders on a log-log scale. Lines are guides to the eye.



Fig. 8. Fracture elongation as a function of strain rate for three solders at room temperature. Elongation was calculated from measuring the full length of the broken sample (\sim 44 mm in length). Lines are guides to the eye.

soldered joints between HTS coated conductors and copperberyllium using both $Pb_{38}Sn_{62}$ and Cerrolow 136 and then straining the joints, we found Cerrolow 136 soldered joints sometimes came apart when they were cooled down to 4.2 K from room temperature and often failed at very low strain values at about 0.5%. Such behavior very seldom happened with $Pb_{38}Sn_{62}$ soldered joints.



Fig. 9. Maximum fracture elongation as a function of strain rate for three solders at room temperature. Elongation was measured in the middle section of the sample (25 mm in length) that was unbroken—outside the breaking point. Lines are guides to the eye.

IV. CONCLUSION AND FUTURE WORK

We have measured some of the electrical, mechanical and superconducting properties of some relatively high B_{c2} solders. These solders can provide good properties for applications requiring joints. We have reported that at room temperature the maximum fracture elongation and ductility of Pb₃₈Sn₆₂ is much greater than Cerrolow 136 and that a measurement of the maximum fracture elongation provides a good measure of the solder's mechanical properties for joints. In the future, we are going to extend the measurements to low temperatures, include wettability tests and expand the solder selection to contain other increasingly used lead-free solders such as Sn_{96,5}Ag₃Cu_{0,5} [7].

REFERENCES

- Y. Tsui, E. Surrey, and D. Hampshire, "Soldered joints—An essential component of demountable high temperature superconducting fusion magnets," *Supercond. Sci. Technol.*, to be published. [Online]. Available: http://dx.doi. org/10.15128/gh93gz491.
- [2] M. Fink, T. Fabing, M. Scheerer, E. Semerad, and B. Dunn, "Measurement of mechanical properties of electronic materials at temperatures down to 4.2 K," *Cryogenics*, vol. 48, no. 11/12, pp. 497–510, Nov./Dec. 2008.
- [3] K. S. Kim, S. H. Huh, and K. Suganuma, "Effects of cooling speed on microstructure and tensile properties of Sn-Ag-Cu alloys," *Mater. Sci. Eng.*, *A*, vol. 333, no. 1/2, pp. 106–114, Aug. 2002.
- [4] A. Nyilas, "Thermal contraction measurements of various materials using high resolution extensioneters between 290 K and 7 K," in *Proc. AIP Conf.*, 2004, vol. 711, p. 151.
- [5] N. R. Werthamer, E. Helfand, and P. C. Hohenberg, "Temperature and purity dependence of the superconducting critical field, H_{C2}. III. Electron spin and spin-orbit effects," *Phys. Rev.*, vol. 147, pp. 295–302, 1966.
 [6] W. J. Plumbridge and C. R. Gagg, "Effects of strain rate and temperature on
- [6] W. J. Plumbridge and C. R. Gagg, "Effects of strain rate and temperature on the stress–strain response of solder alloys," *J. Mater. Sci., Mater. Electron.*, vol. 10, no. 5, pp. 461–468, Jul. 1999.
- [7] H. Ma and J. C. Suhling, "A review of mechanical properties of leadfree solders for electronic packaging," J. Mater. Sci., vol. 44, no. 5, pp. 1141–1158, Mar. 2009.