CRYOGENIC PROPERTIES OF METALLIC AND NON-METALLIC MATERIALS UTILIZED IN LOW TEMPERATURE AND SUPERCONDUCTING MAGNETS

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I. Introduction

Composite superconductors allow the transport of current densities at least one order of magnitude higher compared to room temperature normal conductors. The superconductor may be exposed to magnetic fields higher by a factor of ~ 2.5, than the peak field values in conventional or cryogenic experimental magnets. Thus superconductors and composites are subject to mechanical stresses which are a factor of about 30 higher than in room temperature magnets. Although the material strength is increased at cryogenic temperatures compared to room- or elevated temperatures, the gain is not nearly sufficient to compensate the strain imposed by the magnetomechanical forces acting upon the conductor.

Stresses on the conductor have various origins:

(i) conductor prestraining during winding

(ii) different thermal contraction coefficients of various materials being in contact with each other (e.g. components of the conductor, coil former, bandages, supports)

(iii) magnetomechanical forces (Lorentz-forces), when the magnet is in operation.

In general materials used in cryogenic or superconducting magnets are seldom stressed beyond their elastic limit. However if the material is strained periodically (thermal and magnetic cycling) the material exhibits fatigue and its yield strength will be reduced. In this case plastic creep may occur. On the other hand, if materials are cooled, their ductility is reduced at low temperatures and material embrittlement is observed.

We treat in this paper briefly: mechanical, thermal and magnetic properties of metals, mechanical and thermal properties of plastics at cryogenic temperatures in quantitative form. The data presented are of interest in the design of low temperature normal and superconducting magnets.[1]

II. Mechanical Properties

The most common tests to measure the strength of solids are tension tests yielding elongation. Data obtained are yield- and tensile-strength and the elastic modulus. In Fig. 1 the stress-strain relation of copper and aluminum is given at 4.2 K. For polycristalline metals the tensile strength may be two to five times that at room temperature. In Fig. 2 we represent the stress-strain relation of a number of materials, such as for Nb(50%)Ti, at room temperature and 4.2 K, and of a composite conductor with a copper to superconductor ratio of 10:1, annealed copper and glassfiber reinforced epoxies (F-MNA). It may be pointed out, that the effect of the fiber glass angle to the direction of the applied strain is decisive.

Structural materials for reinforcement of coils are few types of nonmagnetic stainless steels, beryllium coppers and unidirectional glass tapes. When some stainless-steels are strained at low temperatures they may become slightly magnetic, which for pulsed magnets (residual fields) and high precision magnets (field homogeneity) is a disadvantage. Unidirectional glassfiber tapes or beryllium copper are selected for reinforcement.

It is a well accepted criterion that the choice of material for reinforcement is not only based on high yield strength but also on impact strength and the resistance against fracture. The criterion for crack growth is given by the GRIFFITH-relation as the gradient of the stored elastic energy U over the fracture face A. Cracks grow when the surface energie γ fulfils the condition $\gamma = -\partial U/\partial A$. The crack in thermosettings used to impregnate coils, grows and propagates between coil pancakes, layers or even between turns. A network of cracks which is common in pure thermosettings without glassfiber reinforcement releases elastic energy, which in unsupported conductor sections yields conductor movement. The released elastic energy is by far sufficient to initiate premature quenching and thus coil training. Several possibilities have been explored to date: The use of very elastic thermosettings, which become hard when cooled down, but the stored elastic energy released is too small to produce cracks. Various types of wax have been employed to impregnate coils with good success. Another method is to use filled epoxies to match their thermal contraction to that of the composite conductor. As a filler inorganic materials such as Al_2O_3, quartz or glass rovings are used. The use of a filler has three distinct effects:
The thermal contraction can be matched to that of a metal or a composite.
The thermal conductivity is increased with the concentration of the filler.
The flexural-strength of the material is reduced, which affects the bond between insulation and conductor, as well as the ultimate strength of the complex coil, composed of several metallic and nonmetallic materials.

The mechanism of interaction between the resin and the filler or reinforcement is a simple one. When the impregnation is performed, the thermosetting must wet the fiber or filler thoroughly. It polymerizes or condenses around the fiber to form a rigid solid. During this process not only does the resin stick to the fiber but also shrinks on curing. The mechanism of adhesion is thus partially mechanical. It is however known that adsorption and diffusion are equally important. Finally chemical and physical effects have to be considered. The mechanical adsorption and diffusion processes involve intermolecular forces, the physical effect electrostatic forces and the chemical effect the chemical bond.

In order to improve the bond strength of the filled impregnant, wetting of inorganic surfaces by means of organic polymers (silicon chemicals) such as silan coupling agents has been very successful. When treating the E-glass cloth or the Al_2O_3 filler with the organic polymer and impregnating it with epoxy, the flexural strength of the system is increased by a factor of two to four. This improvement in mechanical strength permitts the filling of epoxies by more than 60% of its weight. As seen in Fig. 10 this complex mixture has almost the same thermal contraction characteristics as the composite conductor.

Material embrittlement is encountered when high strength materials are cooled to cryogenic temperatures. A classical example is low carbon steel (Fig. 3). As in actual magnet design the iron return yoke is cooled down to liquid helium temperature and the coils are supported by the iron return yoke (shell), the choice of the core materials is not only by means of a.c.-losses or relative permeability but also by their mechanical and fatigue properties.

When magnets are thermally cycled or pulsed (as in synchrotron - magnets), the conductor as well as structural materials undergo periodic mechanical strain. Due to fatigue the ultimate strength of materials is reduced. As illustrated in Fig. 4 and Fig. 5 the ultimate tensile strength of aluminum is reduced threefold and of copper at 4.2 K about 15%. Glassfilament structures are fairly persistent.

III. Thermal and Transport Properties

Under this cathegory we treat the specific heat, thermal and electrical conductivity and thermal contraction.

Specific heat

The specific heat of a number of metals is illustrated in Fig. 6. It is strongly dependent of temperature, the lattice part being proportional to T^3 for low temperatures below $\theta_D/10$ (θ_D= DEBYE-temperature) and the electronic part proportional

to T. It is seen that the experimental data for pure metals are in good agreement with DEBYE theory at temperatures below 10 K, where the quadratic DEBYE phonon spectrum is a good approximation.

For Nb_3Sn the jump in the temperature dependence of the specific heat near T_c due to the exponential behaviour of the electronic part below T_c, is close to the BCS prediction.

Thermal conductivity

Thermal conductivity of metals, superconductors and alloys is given in Fig. 7, for pure and filled epoxies in Fig. 8. For nonconducting crystalline materials thermal conductivity is governed by phonon-defect-interactions and phonon-phonon-"Umklapp"-processes. At low temperatures when the phonon wavelength becomes comparable to the defect dimensions, impurity scattering dominates and gives reason for a T^2 to T^3 increase of the thermal conductivity.

For metals the conduction electron scattering is usually more important than phonon scattering. The electronic contribution results from scattering at crystal imperfections proportional to T and the electron-phonon-interaction proportional to T^{-2} for temperatures below $\theta_D/10$. If the defect concentration increases (cold working, impurity alloying), the electronic thermal resistance due to imperfection scattering may become comparable to the thermal resitance in the phonon conductivity branch. In this case the lattice thermal conductivity cannot be neglected; its influence is manifested in a reduction of the conductivity and a suppression of the conductivity maximum. This behaviour is well demonstrated by the superconducting alloys in Fig. 7.

As an electronic transport phenomenon the thermal conductivity of metals is reduced by magnetic fields. Up to now only few experiments have been performed to study the thermal magnetoresistance.

Thermal diffusivity

Thermal diffusivity of materials, defined by the thermal conductivity and the heat capacity, is important in the study of stability criteria in superconducting magnets. Enthalpy stabilization requires that the thermal diffusivity be much higher than the magnetic diffusivity in order to confine the region of normality or cool down the normal area and restore superconducting conditions. As seen from Fig. 9 the thermal diffusivity of superconductors is at least three orders of magnitude smaller than that of commercially available aluminum. The thermal diffusivity of filled epoxies is even three times lower at 4.2 K than that of Nb_3Sn. It is therefore of interest to imbed superconductors in matrix materials with very high thermal diffusivity, but due

to eddy current losses and coupling between filaments, pure metals are not always adequate in multistrand composites. Using copper or aluminum alloys the thermal diffusivity is reduced about two orders of magnitude when compared to very pure metals.

Thermal contraction

Thermal contraction is an effect of non-harmonic lattice vibrations and is according to the GRÜNEISEN theory proportional to the specific heat. With decreasing temperature and therefore decreasing vibrational excitation the mean interatomic distance is lowered, the material contracts. The temperature dependence of several metals, alloys and thermosettings, filled and unfilled, is given in Fig. 10. As metioned before, loading the thermosetting with inorganic fillers the thermal contraction can be reduced considerably to match copper or even composite conductors. With fibre glass as filler the thermal contraction can be varied by the relativ amount and the angle of glass filament to the direction of the contraction considered. Fig. 11 shows the variation of thermal contraction versus the angle of roving (direction of glass filaments to the direction of contraction). It is interesting to note that the variation of $\Delta L/L$ at temperatures below 77 K could be an order of magnitude.

Electrical resistivity

The electrical resistivity is due to interactions of the conduction electrons with crystal defects and phonons. The lattice part proportional to T^5 may be neglected at 4.2 K, where only the temperature independent residual resistivity is important.
Under the influence of mechanical stress, impurity alloying or irradiation the defect concentration is increased thus reducing the resistivity ratio $r=\rho(293K)/\rho(4.2K)$ Fig. 12). For high purity polycrystalline bulk metals r may be in the order of several ten thousands. The resistivity ratio is reduced for thin films and wires, when electron mean free path and sample dimensions become comparable (FUCHS-SONDHEIMER-theory). Furthermore, the resistivity ratio is affected by the magnetoresistance. The relative decrease of resistivity $\Delta\rho/\rho=$ $(\rho(B;T)-\rho(0;T))/\rho(0;T)$ is according to KOHLER's rule a function of $B \cdot r_{B=0}$ only. This was found to be true, if only one scattering mechanism is acting. If there are two (or more) mechanisms of comparable influence as impurity and phonon scattering at T>4.2 K, KOHLER's rule is not applicable. $\Delta\rho/\rho$ increases with increasing purity and temperature (for aluminum in the range from 4.2 K to about 18 K). For some metals, e.g. aluminum, $\Delta\rho/\rho$ tends to saturation in high fields, whereas for others, as copper, a linear increase of $\Delta\rho/\rho$ with B has been observed up to 10 Tesla. This behaviour reflects the different electronic structure

represented by the shape of the FERMI surface. For very impure metals and alloys the magnetoresistance effect is often negligible. For Al and Cu the magnetoresistance behaviour is shown in Fig. 13.

Reference:

[1] H. Brechna, G. Hartwig, W. Schauer: KFK Nr. 1470 (1971)
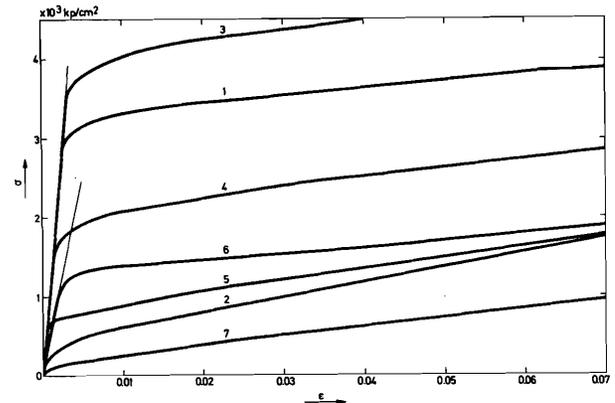In this paper all pertinent references are given.

Fig.1. Stress-strain relation for Cu and Al.
1. ETP-copper 1/4 hard; 2. ETP-copper annealed; 3. OFHC-copper hard; 4. OFHC-copper 1/2 hard; 5. OFHC-copper annealed; 6. commerc. Al; 7. Al annealed.
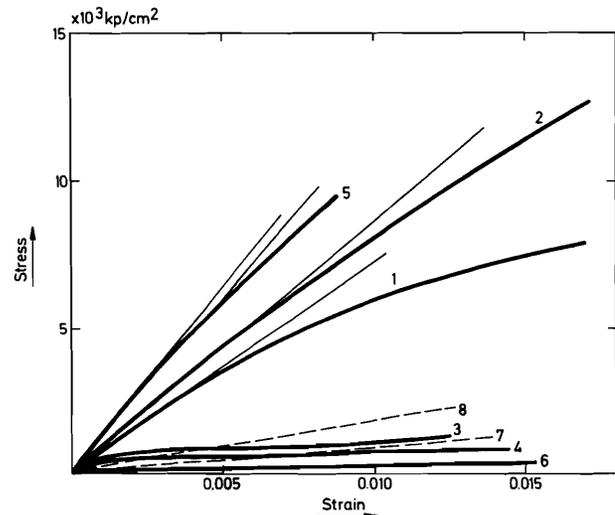

Fig.2. Stress-strain relation for various materials.
1. $Nb_{(x)}$ Ti wire, 0.025 cm diam. at 300 K; 2. $Nb_{(x)}$Ti at 4.2 K; 3. composite conductor: Cu: $Nb_{(x)}$Ti = 10:1; 4. annealed OFHC-copper; 5. Be(2%)Cu at 4.2K; 6.,7.,8.glassfiber-epoxy tubes, compression test, filament angle to tube axis: 6.:45°; 7.:30°; 8.:55°.
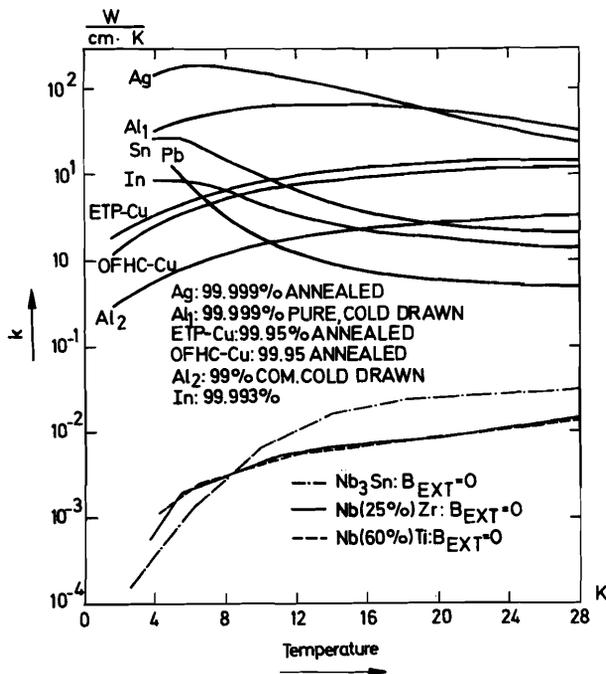
Fig.3. Ultimate tensile, yield and elonga-
tion of low carbon and Ni(9%)-steel
vs. temperature.

Fig.5. Fatigue properties of few structural
materials at various temperatures.

Fig.4. Fatigue data of Cu and Al at various
temperatures (NBS)

Fig.6. Specific heat of materials.
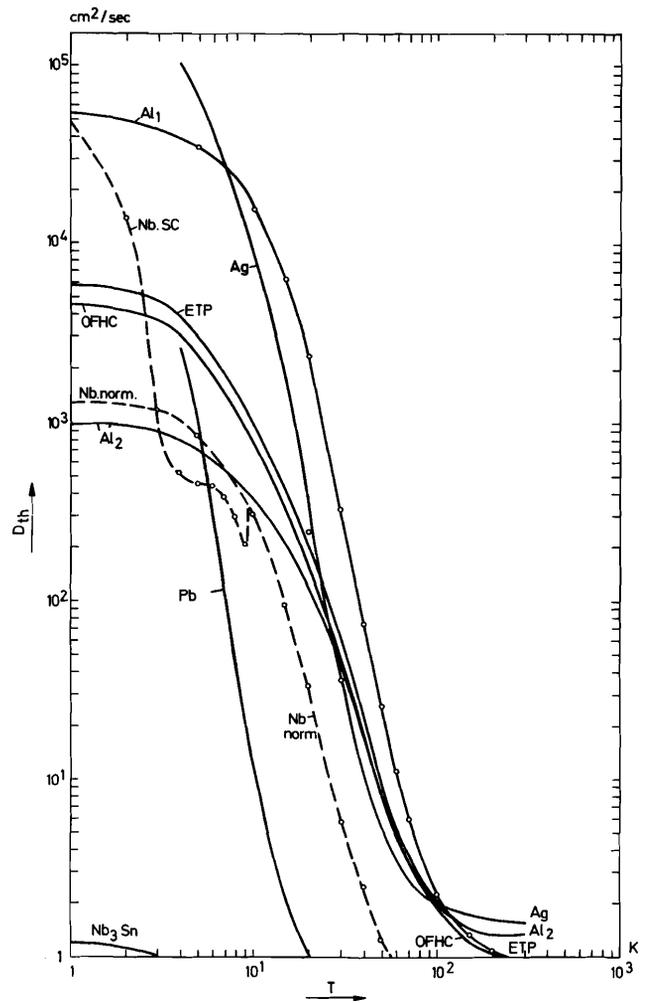
Fig.7. Thermal conductivity of metals and Type II superconductors.
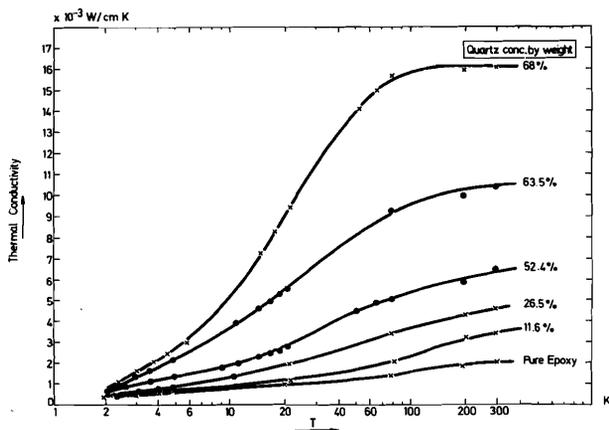

Fig.9. Thermal diffusivity of various materials.

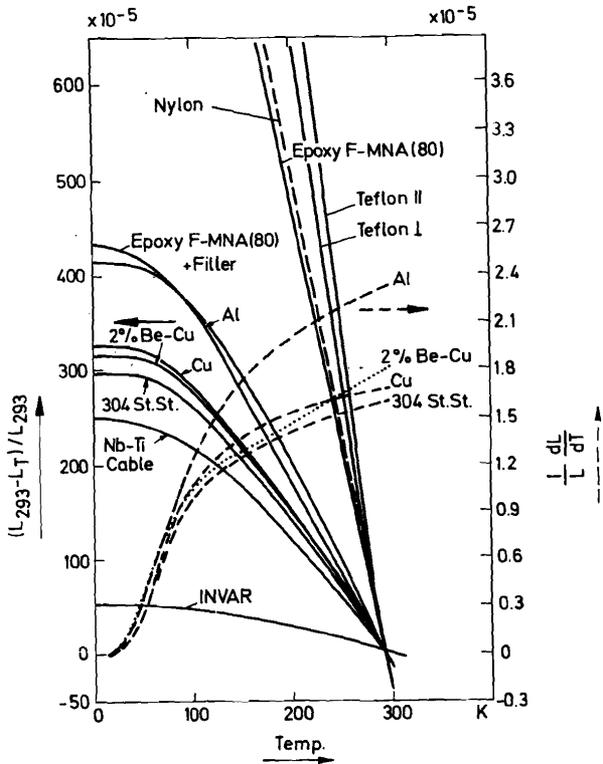Fig.8. Thermal conductivity of epoxies (F-MNA), pure and quartz filled (RHEL)
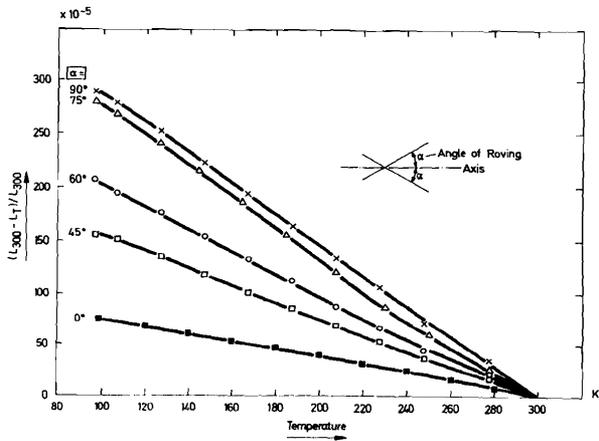
Fig.10. Thermal contraction of materials.

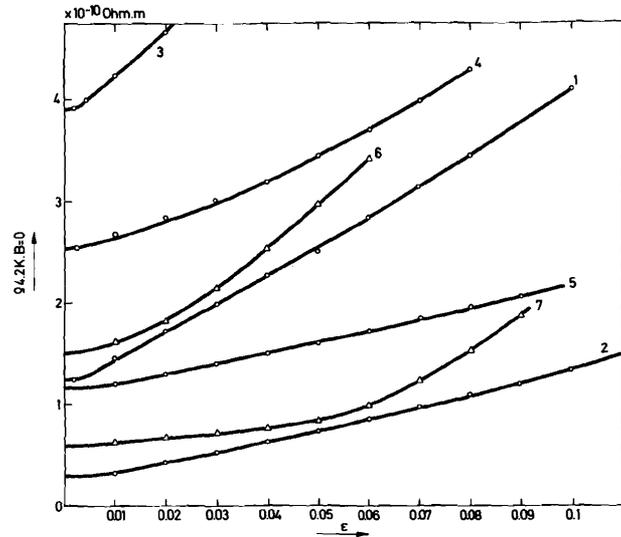Fig.11. Thermal contraction of fiberglass-
epoxy systems.

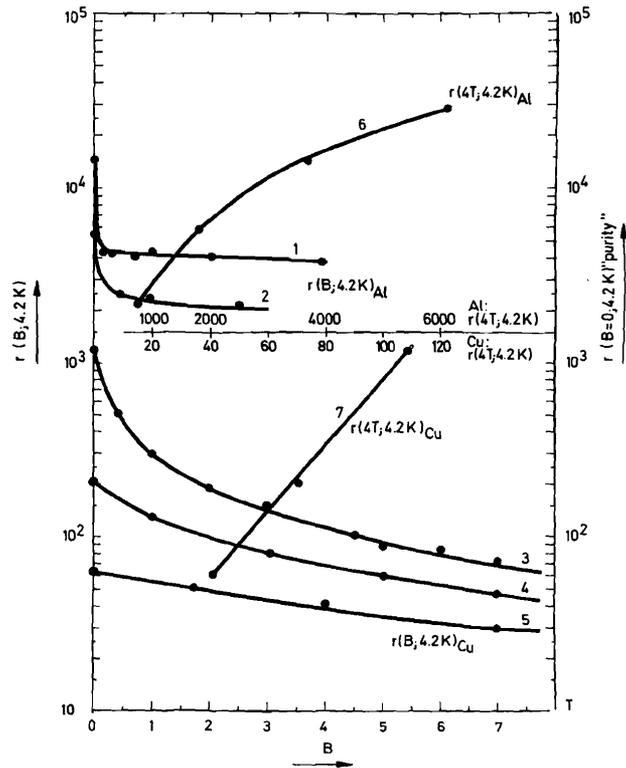Fig.12. Resistivity vs. strain for Cu and Al
(notations as in fig.1)

Fig.13. Magnetoresistance of Al and Cu at
4.2K. Resistivity ratio as function of
magn. field (1.-5.); resistivity ratio
at 4 Tesla, 4.2K as function of purity
(6.,7.). 1.,2. high purity Al (NBS;GfK
Karlsruhe); 3. 99.999% Cu; 4. ETP-
copper; 5. OFHC-copper (3.,4., 5.: Gen.
Elec.); 6. high purity Al 2,000 ≤ r
(B=0;4.2K) ≤ 28,000 (NBS); 7. copper,
purity as 3.,4., 5.