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The Influence of Microstructural Scale on the Combination of Strength and Electrical Resistivity in Copper Based Composites

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Introduction

There are many engineering applications in which composite materials are required to satisfy two or more criteria regarding physical and mechanical properties. An example is the development of two-phase wires or tapes for the fabrication of coils for high field magnets (WOOD et al., 1995). Simple diagrammatic representations can be developed which display the combinations of properties for a variety of materials in relation to design requirements. An example of such a diagram is shown in Figure 1.

For a simple solenoid, these diagrams are based on the development of functions to describe the response of the material in terms of the strength required to resist the Lorentz force and the conductivity required to minimize the ohmic heating in a given pulse of time Δt. Thus on this diagram we can indicate minimum acceptance criteria either in terms of the strength of the magnet in Teslas as horizontal lines or diagonal lines indicating maximum pulse durations. In general, materials for this application will be metallic composites or dispersion strengthened materials.

Further insight into the selection of materials can be gained by considering whether the properties of the composite are described by a suitable rule of mixtures or whether the scale and

distribution of the phases in the composite must be considered.

In this overview, consideration will be given first to drawn, filamentary composites in which attention must be given to the scale of the microstructure in relation to both the strength and the electrical conductivity. The results obtained on drawn, filamentary composites raise important fundamental questions regarding both the role of interphase interfaces in electron scattering and the magnitude and influence of internal stresses produced by co-deformation.

These concerns, particularly those related to electrical conductivity, can be addressed by reference to model systems of multilayers of copper and niobium prepared by vapour deposition. This portion of the study is based on the work of Dr. A. J. Griffin Jr. and Dr. M. Nastasi of the Los Alamos National Laboratory.

The final section of the overview will compare methods of using the appropriate laws of mixtures to develop fabrication methods for producing materials with suitable combinations of strength and electrical conductivity.

Drawn Filamentary Composites

Many two-phase materials containing plates or rods of the second phase can be deformed by wire drawing or rolling to yield fine-scale structures as shown in Figure 2. The codeformation of phases to produce fine-scale structures raises three fundamental issues: a) the morphological stability of the embedded phase during large strain deformation (macroscopic aspects of this problem has been examined in detail by STEIF, 1972); b) the range of validity of strengthening relationships such as the Hall-Petch formulation for ultra-fine-scale structures (CHRISTMAN, 1993) and c) the mechanism of energy storage at large strains in two phase systems.

In drawn, two-phase structures, the morphology of the phases can be very complex for the case where one phase is f.c.c. and one is b.c.c. due to the fibrous nature of the b.c.c. textures (HOSFORD, 1964). Thus prior to discussing the details of the validity of strengthening mechanisms and the energy storage processes, let us consider a simple description of the increase in strength due to imposed plastic strain in a system such as Cu-Nb.

If we assume that the strengthening is of the Hall-Petch type where the strength scales as

$\lambda^{-\frac{1}{2}}$ where λ is the spacing of the second phase fibres, the the data can be analyzed as shown in Figure 3 (from SPITZIG and KROTZ, 1988). It can be seen that at fibre spacings of the order of 30 nm the strength level approaches 2 GPa. This is of the order of E/80 where E is the elastic modulus of copper.

A very simple model of electrical resistivity can be developed by assuming that at large strains the dominant contribution to electrical resistivity is scattering at the Cu-Nb interfaces as the spacing of fibres becomes of the order of the mean free path of the electrons.

Following the work of DINGLE, (1950) and SONDHEIMER (1952), the following relationships were proposed to predict the resistivity:

$$\rho_f = \begin{cases} \rho_b [1 + \dfrac{3\bar{x}}{4\lambda}(1+p)] & \lambda \gg \bar{x} \\[2ex] \rho_b \left(\dfrac{1-p}{1+p}\right)\dfrac{\bar{x}}{\lambda} & \lambda \ll \bar{x} \end{cases} \tag{1}$$

where ρ_f is the resistivity of the fibrous material, ρ_b is the resistivity of the bulk material, $\bar{x}$ is the mean free path of the electrons, λ is the interphase spacing and p is the probability of the electrons being scattered elastically. Typical values of p have been found experimentally to be in the range 0.10 to 0.15 (FROMMEYER and WASSERMANN, 1975). The result of applying these equations is the prediction that, at large interphase spacings, the resisitivity is that of the bulk material, whereas at spacings which are comparable to the mean free path, the scattering at interfaces will dominate and the resistivity will be higher than that of the bulk.

Using this type of model, the dependence of electrical conductivity on the scale of the drawn microstructure can be predicted as in Figure 4. The physics of interface scattering has been discussed in detail by SKOMSKI et al. (1992). The resistivity of well controlled, Cu-Nb layered structures is discussed in relation to the model proposed by SKOMSKI et al. in a later section of this paper.

The simple laws for the dependence of strength and resistivity on the scale of the structure enables a basic initial framework to be developed to consider how these characteristics enter into the design of wire for magnets. Thus we can view the question of high-strength, high-conductivity composites from two viewpoints: a) the concept of the design of structures based on the simplified

models outlined above, and b) detailed consideration of the strengthening and resistivity mechanisms in fine-scale structures.

Fine-Scale Composites

If the strength of the composite increases as $\lambda^{-\frac{1}{2}}$ and the resistivity varies as λ^{-1}, then the combination of properties can be plotted on the design diagram as shown in Figure 5. The trajectory of increasing strength and decreasing conductivity will be determined by the <u>initial scale of the structure, λ_0, and the magnitude of the imposed strain in the drawing process</u>. The essential points of such a treatment are that structures with finer initial scale are advantageous because they can be processed with smaller imposed strains. However, the methodology has the drawback that both properties are linked to the same microstructural scaling parameter, namely the interphase interfacial spacing λ.

Macroscopic Composites

The properties of strength and resistivity can be effectively decoupled by considering a macroscopic composite in which the scale of the conducting phase (in this case copper) remains in excess of the mean free path of the conducting electrons while the second phase is chosen such that it is able to work harden rapidly.

Typical cross sections for such macroscopic structures are shown in Figure 6. Using this concept, we can use the fact that the copper stores dislocations as it work hardens but these contribute a resistivity of only 10^{-25} $\Omega \cdot m^3$ per unit length (BASINSKI and SAIMOTO, 1967) and hence the conductivity of heavily deformed copper is only reduced from the annealed copper standard by $1 - 2\%$ in the absence of any other scattering mechanisms. The incorporated second phase can now co-deform with the copper but strain harden at a much higher rate than copper. Typical examples would be pearlitic steel or some stainless steels which undergo a martensitic transformation upon deformation. The appropriate law of scaling is then:

$$\sigma_{comp} = \sigma_{Cu} V_{Cu} + \sigma_{EP}(\varepsilon)(1 - V_{Cu}) \qquad (2)$$

where V_{Cu} is the volume fraction of the copper matrix and $\sigma_{EP}(\varepsilon)$ is the strength of the embedded phase after an imposed codeformation of ε. The resultant increase in strength predicted for copper

containing 20% of various phases capable of work hardening to high imposed strains is shown in Figure 7. It can be seen that this method of decoupling the dependence of strength and electrical conductivity on the scale of the structure can be expressed as a new development trajectory on the design diagram as shown in Figure 8.

Fundamental Issues Relating to Heavily Deformed In Situ Composites

Let us consider first the problem of microstructual scale and conductivity. The recent work of SKOMSKI et al. (1992) considers the interface scattering in terms of the degree of substitutional disorder P_m and the interface thickness b as well as the scale of the microstructure λ. Thus:

$$\rho(\lambda) = \frac{1}{2}(\rho_A + \rho_B)\frac{4bP_m}{\lambda}\tanh\left(\frac{\lambda}{4b}\right)$$

(3)

where ρ_A and ρ_B are the bulk resistivities of phases A and B.

The data obtained for vapour deposited layers of copper and niobium by GRIFFIN et al. (1995) can be plotted in this form as shown in Figure 9. Despite the difficulty of obtaining regular layered structures less than 10 nm in wavelength, the data shows good agreement with the Skomski model providing an interface thickness of about 3 to 4 nm is assumed. A second interesting feature of this data is the observation that as the thickness of the vapour deposited niobium decreases the observed critical temperature for the superconducting transition in niobium is reduced as shown in Figure 10). The most likely explanation of this observation is that, in the fine-scale structure, superconductive pairing of the electrons is disturbed by the proximity effect, resulting in a reduction of the critical temperature.

Turning to the strengthening mechanisms in fine scale structures, it is of importance to note that several studies (eg. WOOD and EMBURY, 1995) have shown direct evidence of large elastic stress in the niobium filaments after codeformation. These elastic stresses need to be accounted for in the overall description of the detailed strengthening mechanism and it must be recognized that they cause the elastic-plastic transition to be rounded, making it difficult to define the yield stress and critical hardening rate. In addition to the elastic stress, the fine scale of the structure reduces the source length of the dislocations which may also contribute to the overall strength level. Also, it is important to note that the niobium phase is reduced in scale but that its cross section remains essentially uniform. Thus, the scale of deformation during codeformation at large plastic strains

must be extremely fine and it is of value to ask how energy is stored in these structures. As few dislocations are observed in either the copper or niobium, it is probable that storage at the interphase interfaces becomes an important process. This concept has been examined previously in models suggested by GIL SEVILLANO et al. (1981) and EMBURY (1992). Direct evidence of the process can be seen in the high resolution electron micrograph shown in Figure 11 where dislocations can be observed at spacings of the order of 7 – 10 atomic planes. Thus, as the scale of the structure is refined, the embedded phase eventually takes the form of whiskers which are developed by large strain co-deformation of the phses. In addition, large areas of interphase interfaces are created which are subject to tractions due to the compatability of the phases and dislocation storage at the interface.

These observations strongly suggest that in fine scale structures, new mechanisms of energy storage such as interfaces which are under traction due to the capture of dislocations must be examined in detail both by electron microscopy and by methods such as calorimetry and morphological and dimensional stability studies.

Conclusions

This paper emphasizes three essential aspects of fine scale composites:

a)that the scale of these structures can be incorporated in simple scaling laws which have direct relevance to component design in areas such as high field magnets;

b)that comparison of drawn in situ composites and macroscopic composites provide a useful way to decouple properties such as strength and conductivity which are normally linked to the same microstructural scale;

c)at interface spacings below 100 nm phenomena the interphase interfaces influence a range of processes such as electron scattering, superconductivity strengthening and energy storage and that systematic studies of these processes in relation to the specific combinations of materials represents a fruitful area for much future research.

Acknowledgments

References

BASINSKI, Z. S. and SAIMOTO, S. (1967) Resistivity of deformed crystals. *Can. J. Phys.* **45**, 1167–1176.

CHRISTMAN, (1993) Grain-boundary strengthening exponent in conventional and ultrafine microstructures. *Scripta Metall. Mater.* **28**, 1495–1500.

DINGLE, R. B. (1950) The electrical conductivity of thin wires. *Proc. Roy. Soc.* **A201**, 545–560.

EMBURY, J. D. (1992) Micromechanical descriptions of heavily deformed materials. *Scripta Metall. Mater.* **27**, 981–986.

FROMMEYER, G. and WASSERMANN, G. (1975) Anomalous properties of in-situ-produced silver-copper composite wires: I. Electrical conductivity. *Phys. Stat. Sol. A* **27**, 99–105.

GIL SEVILLANO, J., VAN HOUTE, P. and AERNOUDT, P (1981) Large strain deformation and textures. *Prog. Mater. Sci.* **25**, 69–412.

GRIFFIN JR., A. J., EMBURY, J. D., HUNDLEY, M. F., JERVIS, T. R., KUNG, H. H., SCARBOROUGH, W. K., WALTER, K. C., WOOD, J. T. and NASTASI, M (1995) Residual stress, mechanical behaviour and electrical properties of Cu/Nb thin-film multilayers. *in press.*

HOSFORD JR., W. F. (1964) Microstructural changes during deformation of [011] fiber-textured metals. *Trans. Met. Soc. AIME* **230**, 12–15.

SKOMSKI, R., ENRECH, M. and COEY, J. M. D. (1992) *Resistance and structure of metallic multilayers.* Nanostructured Materials, **1**, 337–345.

SONDHEIMER, E. H. (1952) The mean free path of electrons in metals. *Adv. Phys.* **1**, 1–42.

SPITZIG, W. A. and KROTZ, P. D. (1988) Comparison of the strengths and microstructures of Cu-20%Ta and Cu-2-%Nb in situ composites. *Acta Metall.* **36** (7), 1709–1715.

STEIF, P. J. (1972) On deformation instabilities in clad metals subjected to rolling. *J. Appl. Metalworking* **4** (4), 317–326.

WOOD, J. T., EMBURY, J. D., and ASHBY, M. F. (1995) An approach to materials selection for high-field magnet design. *In press.*

WOOD, J. T., and EMBURY, J. D. (1995) The development of ultra-high strength wires by the deformation of in-situ composites. *Proc. of ICCM-10: Vol. 2: Metal Matrix Composites,* (ed. A. POURSARTIP and K. N. STREET), pp. 99–106. ICCM-10 Society.
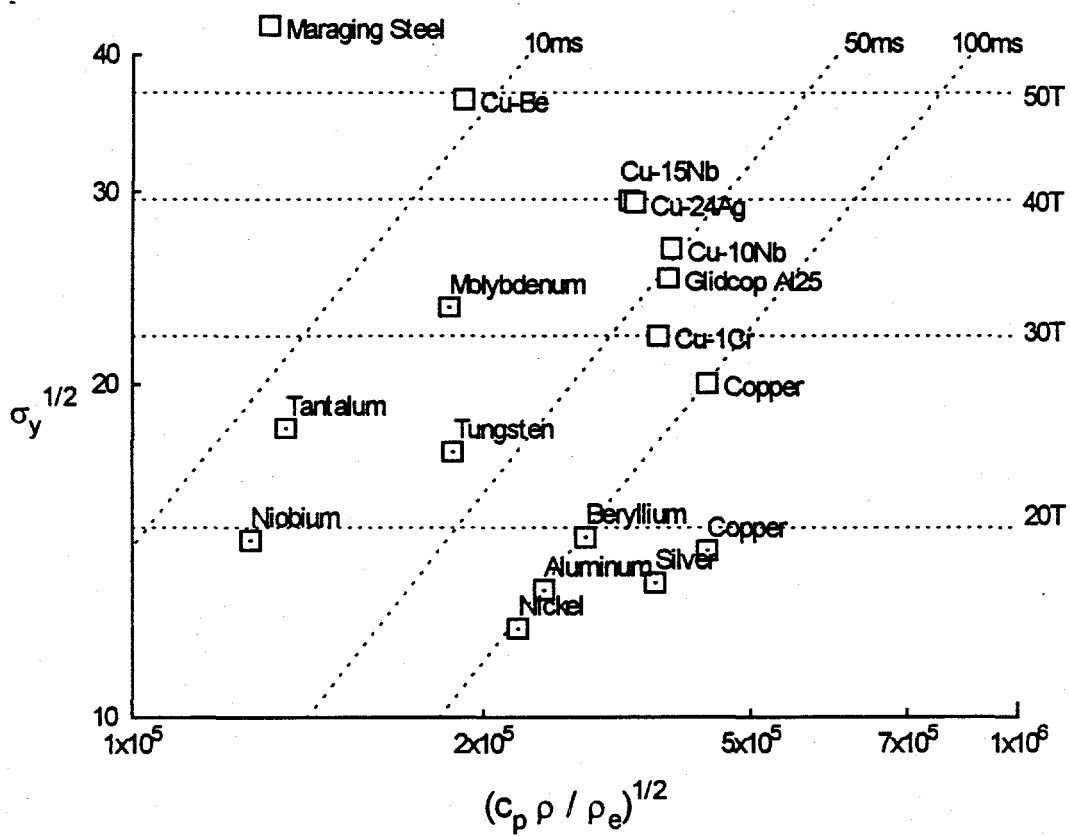
Figure 1.A materials selection chart for the windings of high-field, pulsed magnets.
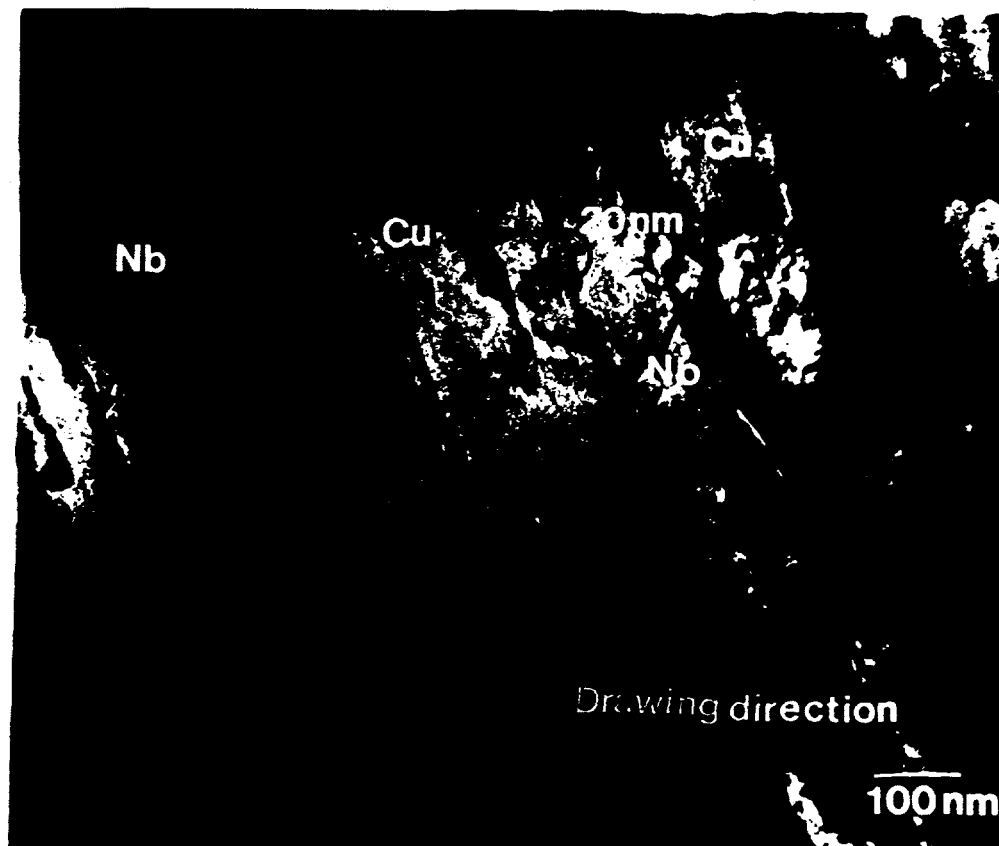
Figure 2.A TEM micrograph of a longitudinal section of copper-niobium wire after drawing.
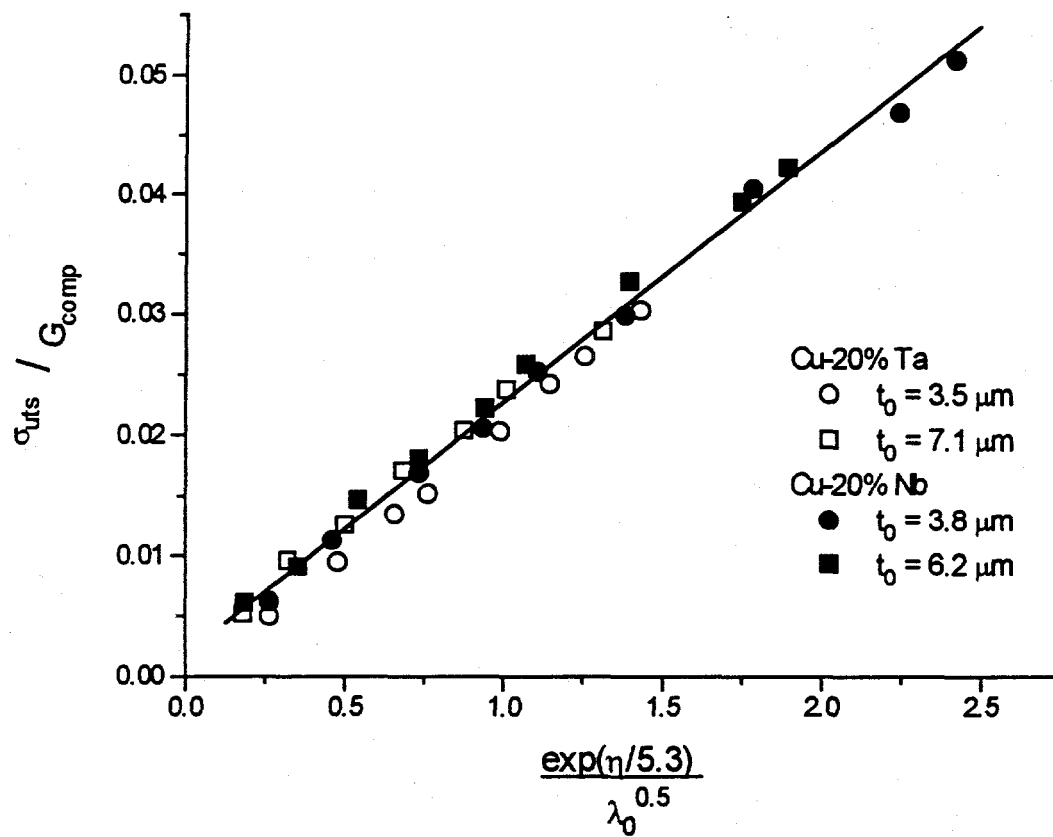
Figure 3.The relationship between normalized strength and interphase spacing for drawn copper-niobium wires. (Spitzig and Krotz, 1988).
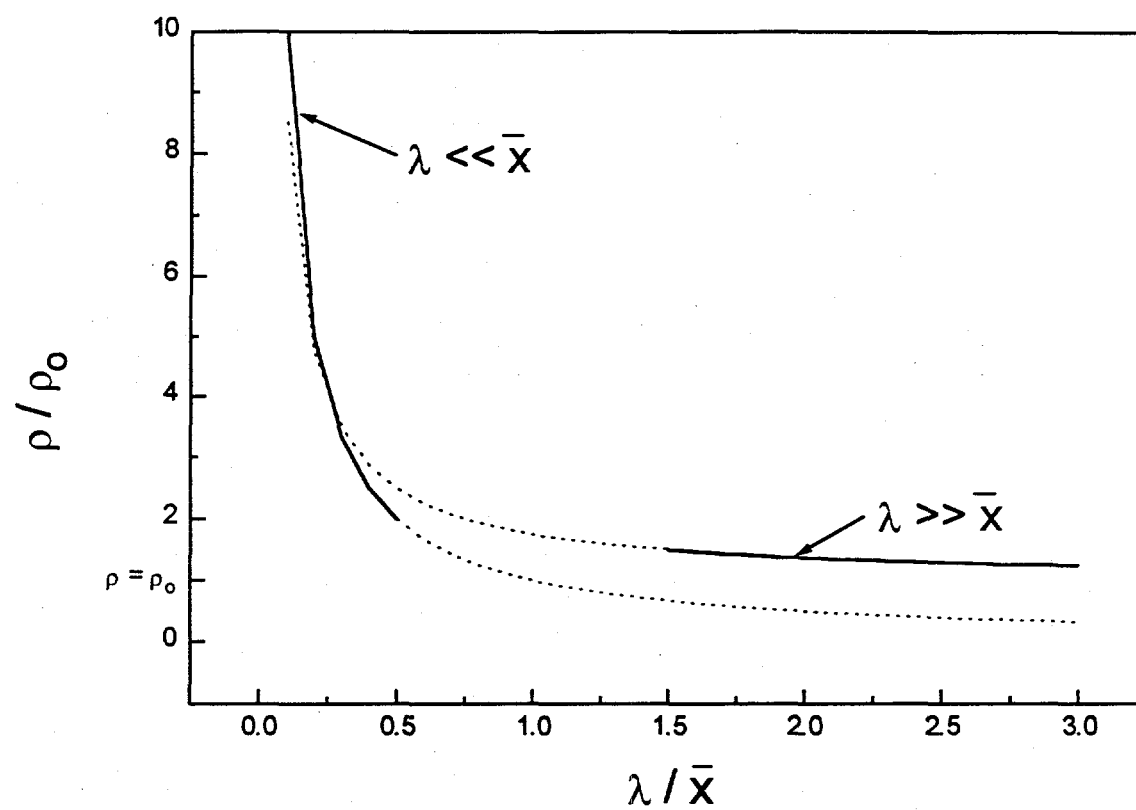
Figure 4.The relationship between electrical resistivity and interphase spacing for fine-scale, two-phase microstructures.
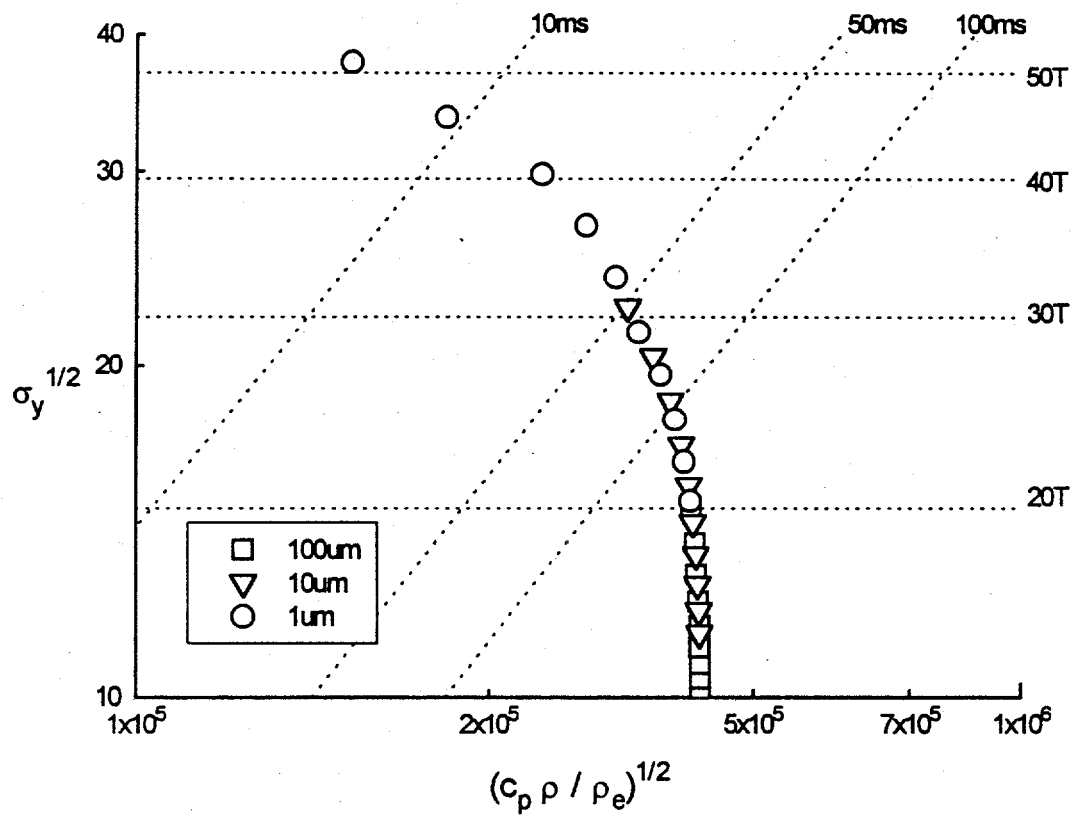
Figure 5. The pulsed magnet materials selection chart showing the evolution of properties of in-situ composites with strain for various initial interphase spacings.
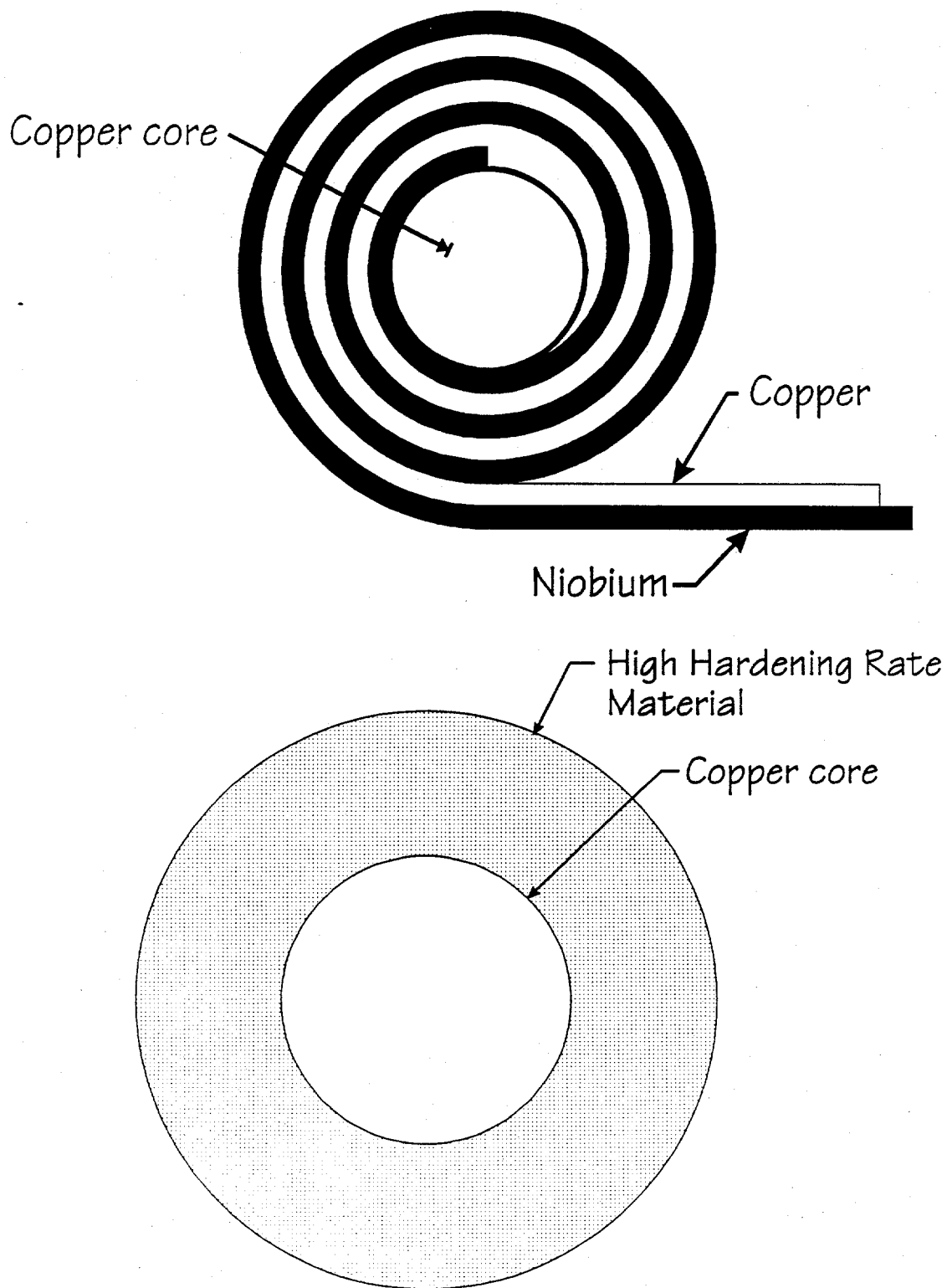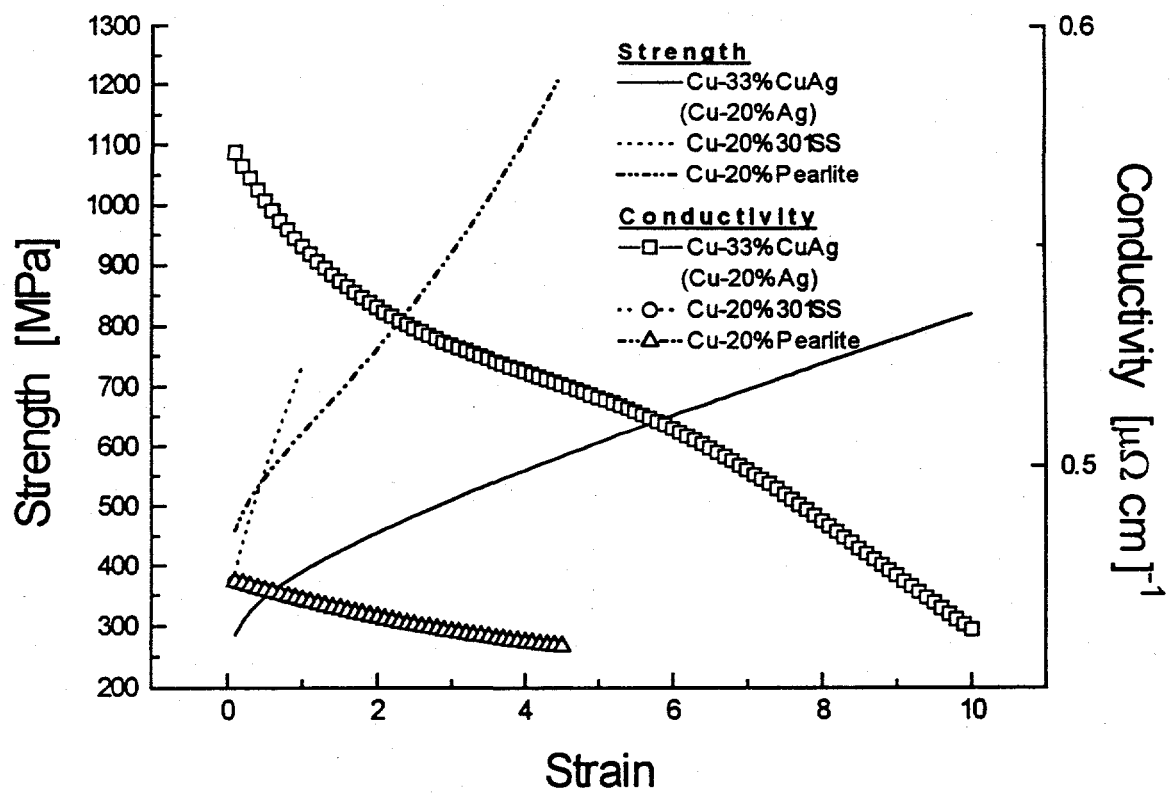
Figure 6.Schematic representations of macroscopic composite structures.

Figure 7. The calculated evolution of strength and electrical conductivity with strain for various macroscopic composite systems. (The conductivity data for Cu-301SS and Cu-Pearlite overlap.)
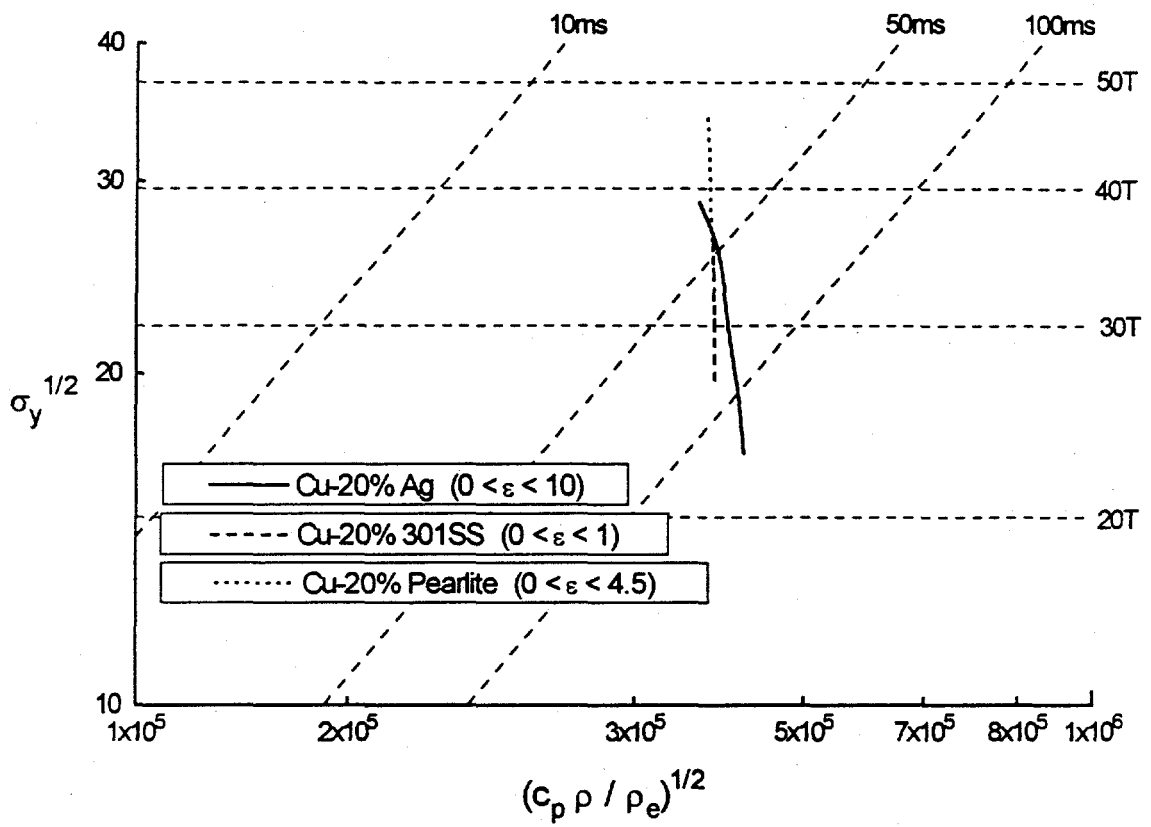
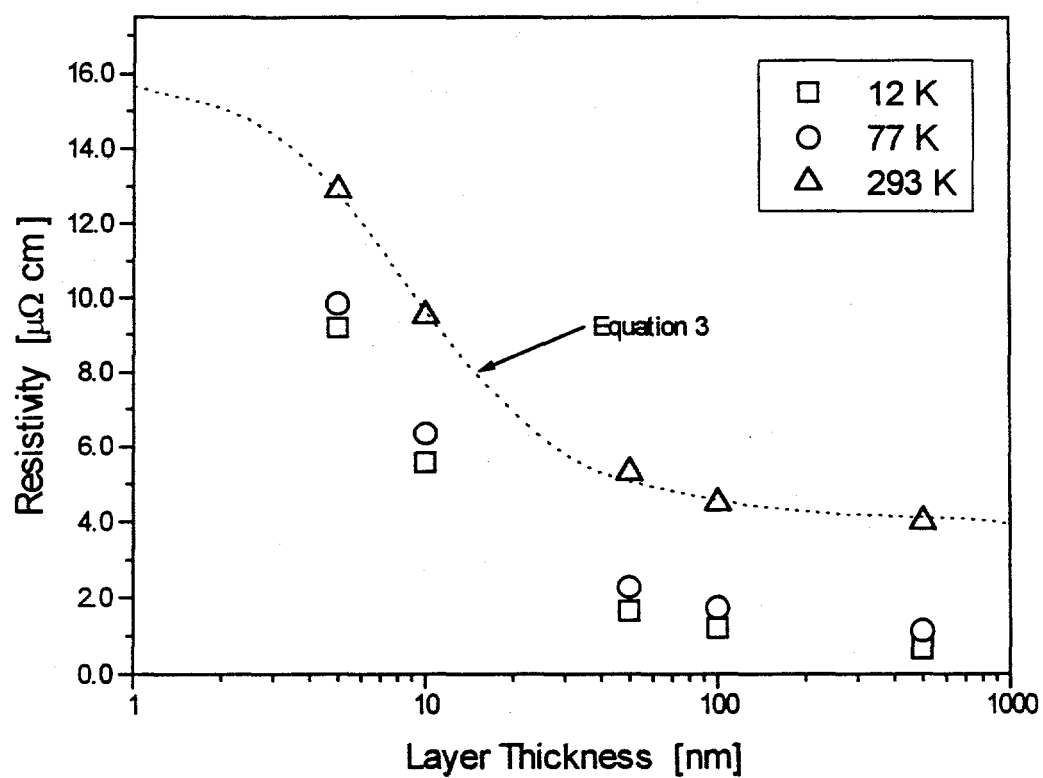Figure 8.Macroscopic composite data of Figure 7 plotted on the pulsed magnet materials selection chart.

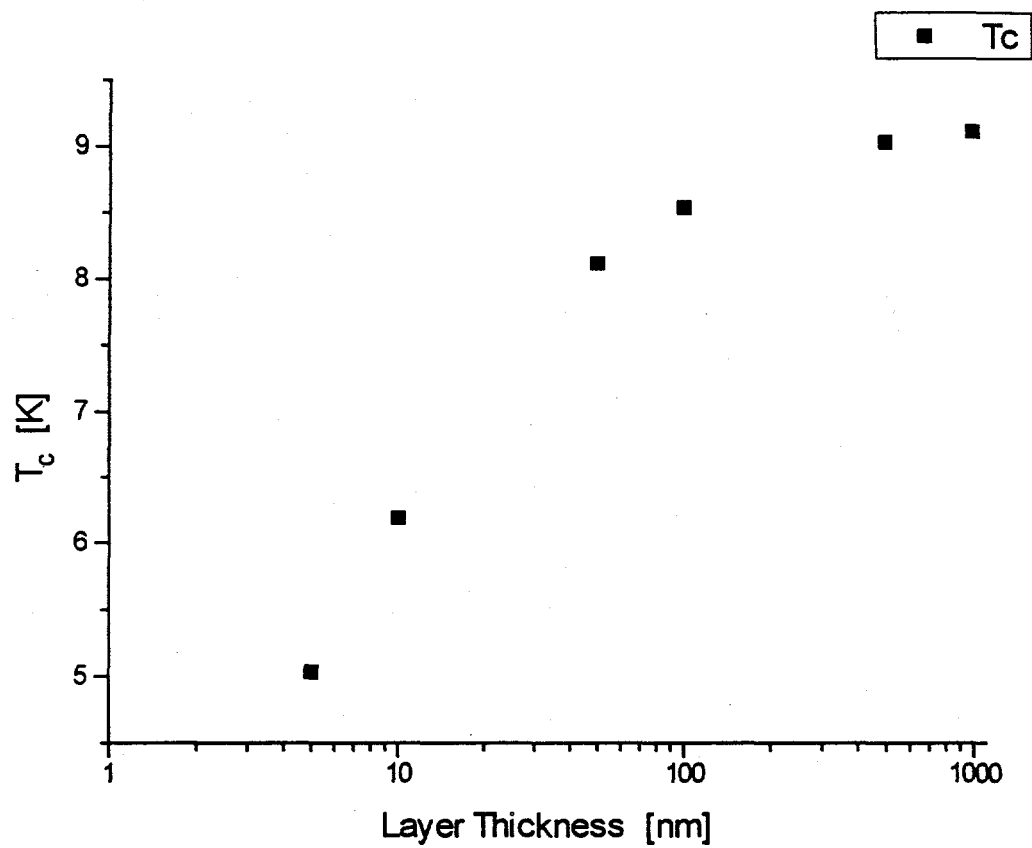Figure 9.The change in resistivity with layer thickness at various temperatures.

Figure 10.The decrease of superconducting transition temperature of niobium with the decreasing niobium layer thickness.
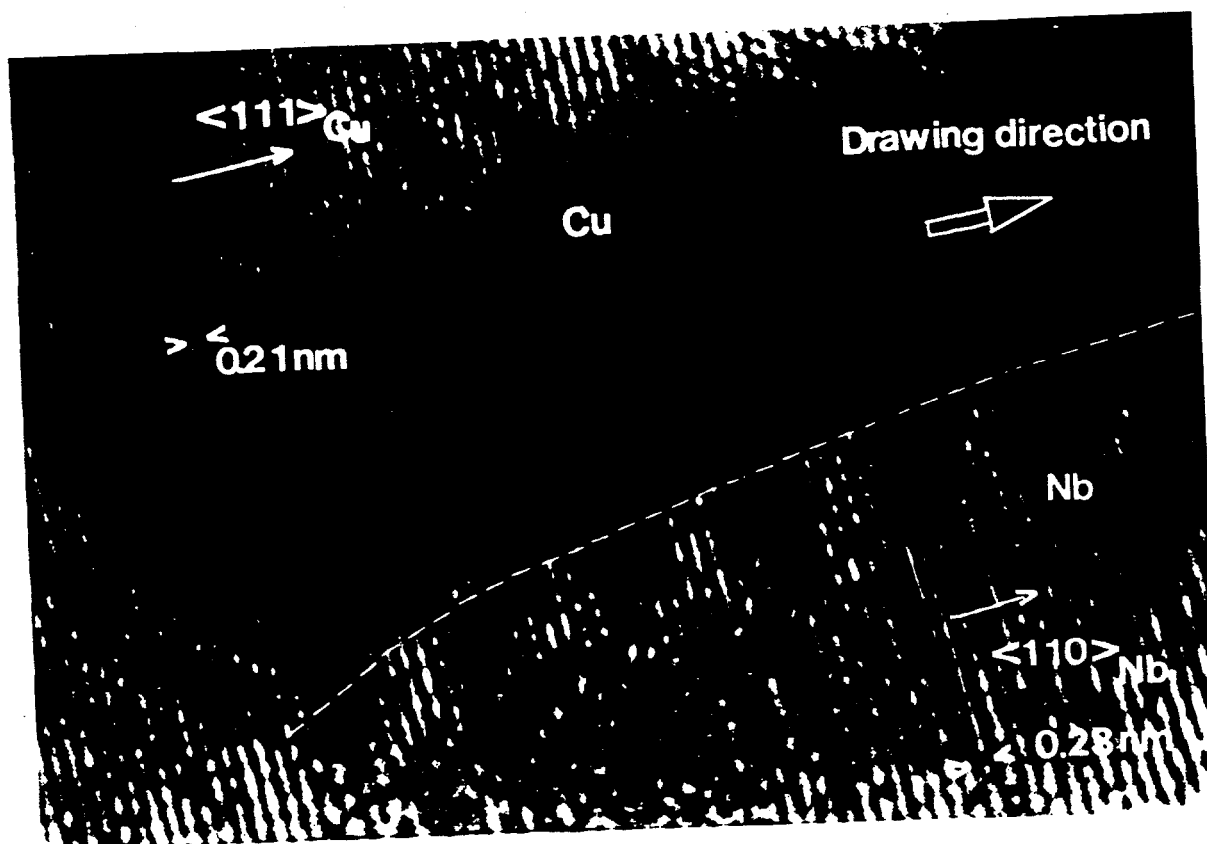
Figure 11.A high-resolution TEM image of the copper-niobium interface showing regularly spaced dislocations.