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Modeling the electrical resistivity of deformation processed metal–metal composites

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Abstract

Deformation processed metal–metal (matrix–reinforcement) composites (DMMCs) are high-strength, high-conductivity in situ composites produced by severe plastic deformation. The electrical resistivity of DMMCs is rarely investigated mechanistically and tends to be slightly higher than the rule-of-mixtures prediction. In this paper, we analyze several possible physical mechanisms (i.e. phonons, interfaces, mutual solution, grain boundaries, dislocations) responsible for the electrical resistivity of DMMC systems and how these mechanisms could be affected by processing conditions (i.e. temperature, deformation processing). As an innovation, we identified and assembled the major scattering mechanisms for specific DMMC systems and modeled their electrical resistivity in combination. From this analysis, it appears that filament coarsening rather than dislocation annihilation is primarily responsible for the resistivity drop observed in these materials after annealing and that grain boundary scattering contributes to the resistivity at least at the same magnitude as does interface scattering.

Keywords: Composites; Electrical resistivity/conductivity; Size effects; Interfaces; Grain boundaries

1. Introduction

Deformation processed metal–metal composites (DMMCs) are a class of in situ composites that display high strength and high electrical and thermal conductivity [1]. They are produced by severe plastic deformation (i.e., rolling or extruding, swaging and wire drawing) of two ductile phases [1]. The first Cu–Nb DMMCs were developed by Bevk et al. [2], and other Cu refractory metal composites were studied by Verhoeven et al. [3]. The strengthening mechanisms in DMMCs have been studied extensively [4–8], but only a few papers have attempted to explain the electrical conductivity of DMMCs [9,10]. An early model of the electrical resistivity of DMMCs can be described by the rule of mixtures (ROM) [10]. However, the ROM considers only the resistivity and volume fractions of each constituent phase without accounting for any microstructural feature of the DMMCs.

Various resistivity models have been developed for macroscopic metal matrix composites to include the microstructural details (e.g. size, shape, orientation and spacing of constituent phases, dislocations and mutual solution) [11–18]. Most of these are empirical models that do not consider the physical mechanisms for resistivity, and they depend on many phenomenological parameters to represent microstructural features. Thus, they are best described as fitting models. Moreover, these models are all based on various simplifying assumptions of composite microstructures and are therefore restricted by the difficulty of

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accurately capturing the complexity of real microstructural features. The spheroidal model developed by Ondracek [11] considers two-phase composites in which discontinuous spheroidal particles are embedded in the matrix phase. This model is capable of predicting the electrical conductivity of two-phase composites over a range of compositions by considering the shape and orientation of both phases. However, this model does not account for the formation of a continuous network of the inclusion phase at a sufficiently high volume fraction, which will increase the conductivity drastically when a conducting inclusion phase is interconnected in an insulating matrix phase [12]. This percolation concept is well described by the general effective media (GEM) equation developed by McLachlan et al. [13]. Based on percolation theory [14], which is applicable only when the electrical conductivities of two phases differ substantially (conductivity of one phase tends to zero), the GEM equation incorporates the conductivities of both constituent phases, their volume fractions and two free parameters to explain different experimental results over an entire composition range. The percolation effect (i.e., the formation of an interconnected network of highly conductive inclusion phases) in this model is described by an asymptotic S-shaped curve that shows the effective conductivity dependence on the volume fraction of the high-conductivity phase. However, the major drawback of the GEM model is that the free parameters do not have a direct correlation with the real microstructural features and serve only as fitting parameters, which limits the applicability of GEM to that of a fitting model relying on experimental inputs [15].

Fan [16] introduced a new approach to topologically transform a two-phase microstructure into a body with three well-defined microstructural elements, which is microstructurally and electrically equivalent to the original microstructure. This model recovers the ROM for conductivity when both phases are perfectly aligned along the current direction (parallel configuration) and recovers the ROM for resistivity when both phases are alternately separated from each other along the current direction (serial configuration). The model considers the effects of the microstructural features (e.g., volume fraction, phase arrangement, phase continuity and phase resistivity ratio) on the electrical resistivity of two-phase composites, making it superior to ROM and Hashin and Shtrikman's bounds for electrical resistivity [17] that account only for the volume fraction and resistivity of constituent phases. The major drawback of Fan's model is that the two constants characterizing the phase topology are difficult to measure, rendering it phenomenological rather than physical. A comprehensive review of these macroscopic models was done by Lux [18].

In all the above resistivity models, the composites comprise a conducting metal inclusion phase and an insulating matrix phase, and the inclusion phase comprises mostly macroscopic particles. However, in DMMCs, both the reinforcement and matrix phases are metals, and the reinforcement phase comprises long (extremely high aspect ratio), almost continuous filaments with a thickness from tens to hundreds of nanometers. None of the above models would be able to predict the resistivity of DMMCs well for the following reason. It is well known that a size effect exists for the resistivity of thin metal films or wires when any one of the dimensions of metal specimens is comparable with the mean free path of free electrons in the bulk metal [19–25]. The mean free paths of free electrons in most metals are in the range of several to tens of nanometers [9,10,26], thus the size effect on the resistivity should be considered in DMMCs. The size-dependent resistivity can be attributed to the increasing restriction of the interface on the mean free path of electrons as film thickness decreases since the resistivity is inversely proportional to the mean free path of electrons. Thomson [19] first derived a formula for thickness-dependent resistivity of thin films by averaging all the free paths of a single free electron over all depths and all angles under the restriction that surfaces act as termination sites for the electron's free path. Fuchs [20] pointed out that it is necessary to consider the mean of all the free paths of all the electrons in the metal in order to obtain an accurate mean free path. By solving Boltzmann's transport equation under the diffuse or partial diffuse scattering boundary condition, he obtained a rigorous formula for thickness-dependent resistivity of thin films and derived approximate formulas for thin and thick film limits. An interface scattering factor was introduced in his paper to characterize the probability of elastic scattering. According to Fuchs, the interface scattering factor is directly related to the surface roughness. A perfectly smooth mirror surface tends to scatter electrons elastically, making the conductivity of a perfectly smooth thin film the same as its bulk counterpart. In contrast, a jagged, rough surface tends to scatter electrons diffusely and increases the resistivity of a thin film.

Fuchs found a good agreement between the theory and experimental resistivity data of a cesium thin film. Sondheimer corrected the approximation formula in the thick film limit of Fuchs [21]. Dingle [22], MacDonald and Sarginson [23] and Chambers [24] extended the thin film work to circular, square and arbitrary cross-section thin wires with two dimensions confined, respectively. In the above theories, grain boundaries are believed to have a negligible effect on the resistivity of the metals because the grain size is usually much larger than the electron mean free path in bulk metal so that the grain boundary contribution to resistivity is relatively small [25]. However, the average grain size in a polycrystalline thin film would be roughly equal to film thickness, which is comparable to the electron mean free path [25]. Therefore, grain boundary scattering also predicts a thickness-dependent resistivity similar to the Fuchs size effect caused by surface scattering. Mayadas et al. incorporated the grain boundaries as parallel, partially reflecting planes with delta scattering potential and solved the Boltzmann transport equation with the purely specular scattering at the external surface (i.e. no Fuchs