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MICROSTRUCTURE AND DAMPING PROPERTIES OF ULTRA FINE GRAINED Al WIRES REINFORCED BY Al_2O_3 NANOPARTICLES

R. Casati[1], M. Vedani[1], A. Tuissi[2], E. Villa[2], D. Dellasega[3], X. Wei[4], K. Xia[4]

[1]Department of Mechanical Engineering, Politecnico di Milano, Via La Masa 1, 20156, Milano, Italy
[2]CNR-IENI, Corso Promessi Sposi 29, 23900, Lecco, Italy
[3]Department of Energy, Politecnico di Milano, Via Ponzio 34, 20133, Milano, Italy
[4]Department of Mechanical Engineering, University of Melbourne, Victoria 3010, Australia

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Abstract

Commercial purity Al and Al based composite wires reinforced with alumina nanoparticles were produced via powder metallurgy route. Powder of Al and Al with 2 wt.% of nano-Al_2O_3 particles were processed by high-energy ball milling. The powders were consolidated by hot extrusion into billets and then rolled to wires down to a square section of 1 mm^2. Commercially pure Al and Al-2wt.% Al_2O_3 powders were successfully consolidated by hot extrusion at 400°C and cold rolled down to wires of 1 mm^2 in section without failures. The ball milling process led to the fragmentation of the native oxide layer that covers the Al powders into nano-sized particles and favored the embedding of these phases in the Al matrix. Then, also commercially pure Al wires took advantage of the strengthening effect of the hard particles. The nanocomposite exhibited higher hardness (about 10%) and higher internal friction than the Al sample.

Introduction

The damping capacity is a measure of the energy that is dissipated by a material during imposed mechanical vibration under cyclic loading. Vibrations generated in response to a dynamic loading can create high noise levels, premature fatigue failure and wear. For a large number of structural and functional applications, it would be of interest to search for new materials exhibiting high mechanical performance and good damping capacity. Most of the frequently used alloys for structural application, like steels, Mg alloys and Al alloys, exhibit a relatively low internal friction [1-2].

In the last few years, metal matrix nanocomposites (MMnCs) emerged as interesting materials to studied since they show higher strength than the corresponding base metals, retaining a good toughness [3-4]. MMnCs are made up of a ductile metal matrix reinforced with hard nanoparticles (NPs). The reinforcement phase is generally thermodynamically stable and apt for high temperature applications [3-4]. Such small particles can hinder the motion of dislocations and are responsible of the formation of geometrical necessary dislocations due to the mismatch of coefficient of thermal expansion (CTE) and of elastic modulus between the metal matrix and the NPs [5-8]. The low wettability and the high surface area of NPs do not allow the preparation of MMnCs by conventional casting methods, thus several alternative processes have been proposed for their production [9-15]. Besides, ultra-fine grained (UFG) materials, processed by severe plastic deformation methods such as equal channel angular pressing (ECAP) or high pressure torsion, have attracted growing interest because of their unique physical and mechanical properties [16]. The combination of properties conferred to the aluminum matrix by a UFG microstructure and by hard NPs would be particularly attractive for all those applications requiring low density and high mechanical properties. In this investigation, Al-based MMnCs wires reinforced by Al_2O_3 NPs and characterized by an UFG microstructure were produce by a powder metallurgy route. The powders were consolidated by hot extrusion into billets and then rolled to wires down to a section of 1 mm^2. This work is mainly aimed at exploring the damping behavior of Al/Al_2O_3 as candidate materials for all those applications in which the combination of good mechanical properties and high capacity to dissipate energy under cyclic load is considered as a fundamental requirement.

Experimental methods

Commercial purity Al powders with average size of 20 μm (supplied by ECKA Granules) and a isopropanol dispersion of alumina with particles average size of 50 nm (supplied by Sigma Aldrich) were mixed and dried in order to achieve a mixture of Al-2%wt. Al_2O_3 composite powder. High energy ball milling was then carried out with Al and composite powders by using a Vario-Planetary Mill Pulverisette 4 equipped by tempered steel bowls and balls (10 mm in diameter). 1.5% vol. of ethanol was added as process control agent (PCA) to avoid excessive cold welding and agglomeration. The bowls were packed and sealed in argon atmosphere into a glove box to prevent oxidation of the powder during the whole process.

High energy ball milling was performed for 16 hours with ball-to-powder weight ratio r = 10:1. Temperature rise was avoided by interrupting the procedure each 30 minutes for 10 minutes. The speed of the main disk was set to 250 rpm clockwise whereas the speed of the two planets was set to 200 rpm counter-clockwise. Powder compaction was executed by hot extrusion [11].

Cylindrical Al alloy cans (external diameter of 10 mm, inner diameter of 8 mm) were filled with the powder which was manually pressed by a plunger at several stages of the filling process. The containers were closed by means of a press-fit plug and then subjected to hot extrusion at 400°C. The extrusion system was heated by an induction coil and the temperature was monitored by a type K thermocouple. The starting billets were characterized by a diameter of 10 mm that were reduced to 4 mm after extrusion. After hot extrusion, the deformed cans were peeled off, and samples were cold rolled down to a square section

of 1 mm^2 by a caliber rolling mill. Intermediate annealing treatments were performed for 5 min at 400°C after area reductions of about 20%.
Microstructural analysis of rolled wires was carried out by scanning electron microscope (SEM) Zeiss Supra 40 equipped with high efficiency In-lens SE detector. SEM analysis was performed on samples etched with Keller's solution. TEM foils with final thickness of 100nm were prepared using a Nova Nanolab 200 focused ion beam (FIB) microscopy (Fig.1). The TEM foils were picked up using ex-situ lift-out method and placed onto a standard 3mm carbon copper grid. High angle annular dark field (HAADF) imaging by scanning transmission electron microscopy (STEM) was performed using a FEI Tecnai F20 transmission electron microscopy operating at 200 kV.

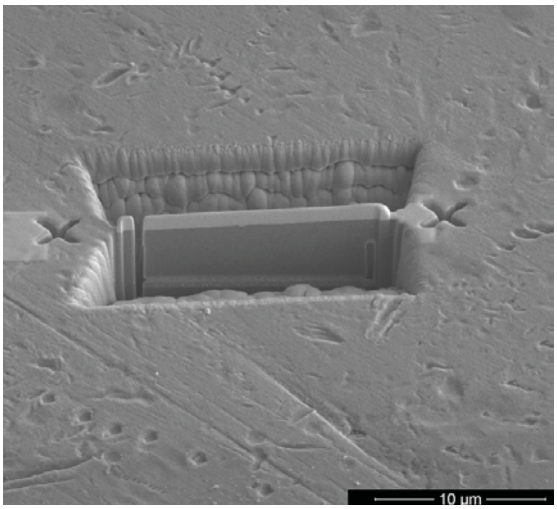

Fig.1 Secondary electron image of TEM foil after milling and polishing by focused ion beam.

Vickers microhardness (HV) was measured using Future Tech Corp. FM-700 tester applying 2 N load for 15 s loading time.
Internal friction tests were carried out using a DMA Q800 TA Instruments equipped by liquid nitrogen cooling system. The samples were tested in single cantilever configuration at two different frequencies, 1 and 10 Hz, in the temperature range from -130°C to 400°C with a temperature heating rate of 2°C/min.

Results

Commercially pure Al and Al-2%wt.Al_2O_3 nano-composite powders were successfully consolidated by hot extrusion at 400°C. The extruded bars exhibited a good workability, indeed they were cold rolled down to a section of 1 mm^2 without failures.
The microstructures of the materials investigated were analyzed by TEM (Fig.2 and Fig.3). Both the Al and nanocomposite wires showed an ultra-fine grained structure. The average grain size of the Al-2%wt.Al_2O_3 composite was slightly smaller than that of commercially pure Al sample (170 nm and 210 nm respectively, as measured by linear intercept methods according to ASTM E112 standard). TEM analysis also highlighted the presence of few nano-sized pores in the nano-composite sample.
The distribution of oxide particles within the matrix was better evaluated by SEM observations. The best results were achieved by etching the samples in Keller's solution. Commercially pure Al sample contains aluminum oxide in the form of fine particles (<70nm), which mostly appear as discrete and well scattered nano-particles in the metal matrix (Fig.4).

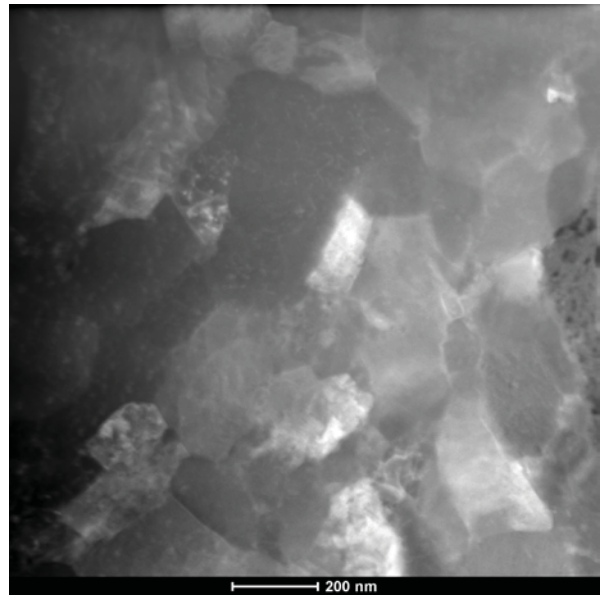

Fig.2 High angle annular dark field (HAADF) image of Al wire (transversal section).

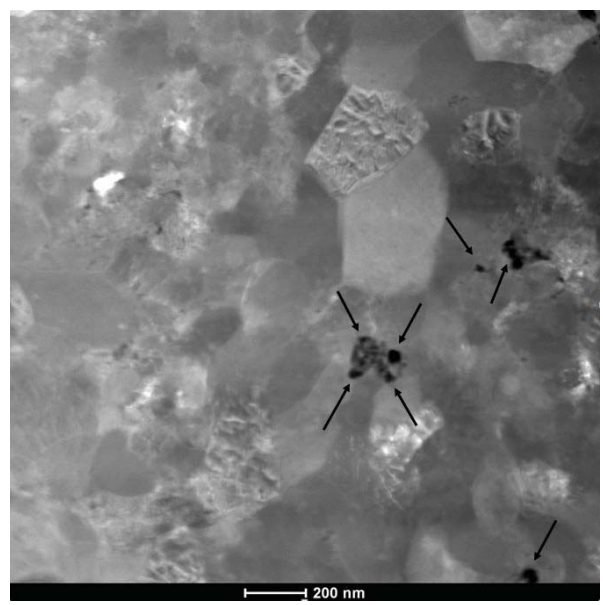

Fig.3 High angle annular dark field (HAADF) image of Al/Al_2O_3 MMnCs wire. The arrows indicate nano-sized pores (transversal section).

Their presence could be due to the ball milling effects that led to the fragmentation of the native oxide layer covering the Al powders into nano-sized pieces and favored their embedding in the Al matrix. On the contrary, the Al-2wt.% Al_2O_3 showed some small clusters of alumina, as depicted in the micrograph of Fig.5. A more severe ball milling might have facilitated an improved breaking up of these Al_2O_3 clusters. The difference in grain size between the Al and composite samples is very small likely

because of NPs pinning effect on grain boundaries which is not only activated in MMnCs sample but also in commercially pure Al wire.

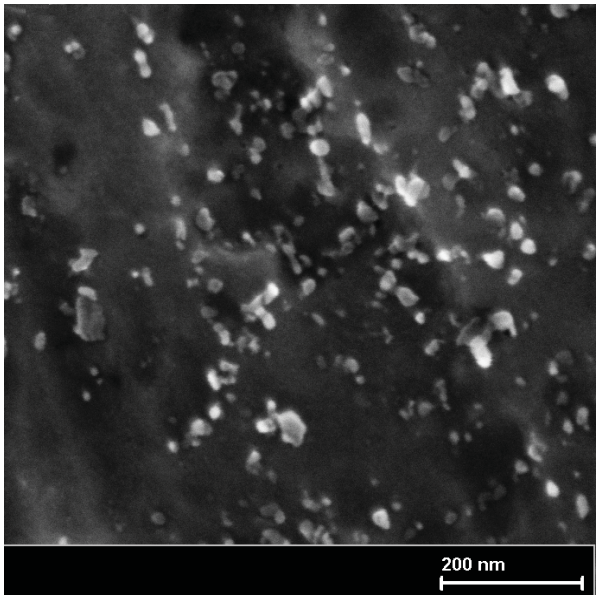

Fig.4 SEM micrograph of Al wire after etching

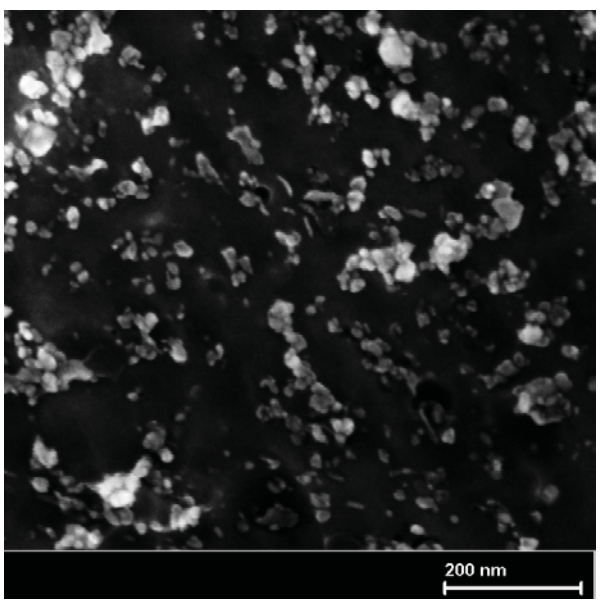

Fig.5 SEM micrograph of Al/Al_2O_3 MMnCs wire after etching

The mechanical properties of the wires were measured in terms of Vickers hardness. The MMnCs showed higher hardness than the Al sample (106±1 HVN vs. 96±1 HVN). The best mechanical performance is supposed to be due to the superposition of different strengthening effects which are more pronounced in the nanocomposite sample. In particular, Orowan, Hall-Petch and CTE mismatch contributions are responsible of the improved mechanical behavior of MMnC sample [7].

The internal friction, or damping, is a property related to time-dependent elasticity of a material. Metals and alloys respond to an applied load, not only by an time-independent elastic strain, but also by a time-dependent strain that lags behind the applied load. Because of the lag induced by the relaxation, the stress σ and strain ε can be expressed as:

$$\sigma = \sigma_0 \exp(i\omega t)$$
$$\varepsilon = \varepsilon_0 \exp(i\omega t - \delta)$$

where σ_0 and ε_0 are the stress and strain amplitudes, respectively; ω is the angular vibration frequency; and δ is the loss angle by which the strain lags behind the stress. By combining these two equations, the resultant complex modulus, E is defined as

$$E = \frac{\sigma_0}{\varepsilon_0}(\cos\delta + i\sin\delta) = E' + iE''$$

where E' is called storage modulus and E'' loss modulus. The storage modulus represents the material stiffness whereas the loss modulus is a measure of the oscillation energy transformed into heat. The ratio between the loss modulus and the storage modulus is the tanδ and it is commonly used as indicator of the damping capacity of a material [1]:

$$\tan\delta = \frac{E''}{E'}$$

In Fig.6, the results of damping tests are depicted in terms of tanδ as function of the temperature. At 1 and 10 Hz, a peak of the tanδ is well appreciable for the curves related to commercially pure Al and nanocomposite wires. The peak shifts to higher temperature increasing the loading frequency. This phenomenon is generally related to relaxation processes (relaxation type peak) [1-2, 17-18]. Nano-Al_2O_3 particles lead to better damping performance (higher tanδ peak) either at 1Hz, for temperatures higher than 70°C, and 10 Hz, for temperatures higher than 100°C. The tanδ (1Hz) at 25°C for monolithic commercially pure aluminum is about 0.001, while at 275°C it is about 0.007 [19]. Then, a significant improvement of the damping capability was achieved by both the materials presented as can be easily inferred comparing the above mentioned values with those extrapolated from the first graph of Fig.6. It is well known that reinforcing particles lead to the formation of interfaces (surface defects) with the matrix due to their lattice mismatch [1]. Moreover, they are surrounded by dislocation networks (line defects) because of the difference in coefficient of thermal expansion between them and the aluminum [5]. Damping behavior of materials is indeed very sensitive to the presence of defects. For instance, dislocations contribute to damping by the internal friction between the vibrating dislocation lines and their adjacent regions, while interfaces give their contribution by the mobility of the incoherent microstructure, when the bond of the two surfaces is strong, and by slip, when the bond is weak. Also grain boundaries play a role in damping through reciprocal viscous sliding [1,2]. Thus, the overall internal friction behavior of the materials object of this work is given by the contribution of surface and line defects. The expected higher density of defects in the nano-composite wire than in the Al one is in agreement with the increased measured damping behavior. Moreover, although the external applied stress is small, the internal stress concentration in regions of high density of defects may be enough to cause relative atomic sliding. At room temperature, the displacements are typically fractions of an atomic diameter whereas, at high temperatures, this sliding can be

extensive and lead to viscoelastic strain. For this reason the damping effects are more pronounced at elevated temperature [1,2].

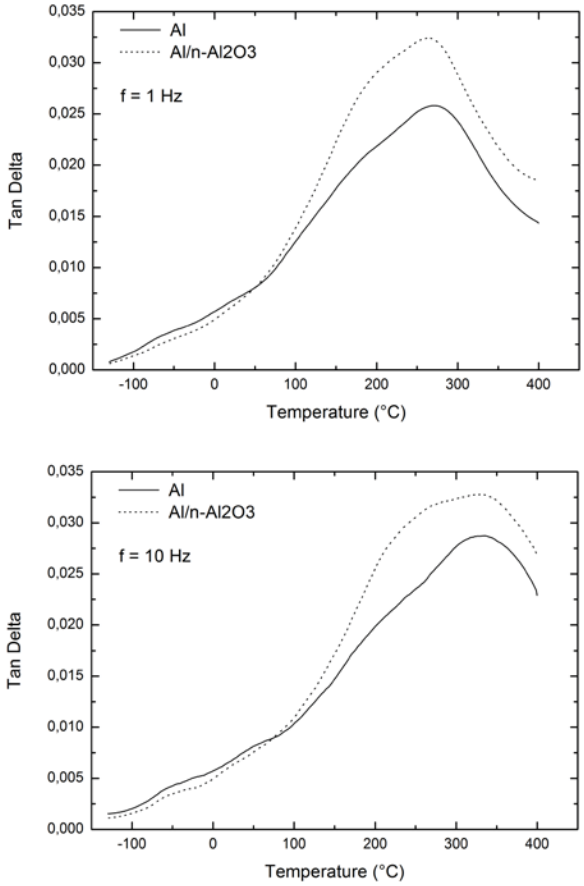

Fig. 6 The results of damping tests are depicted in terms of tanδ as function of the temperature. The tests were carried out at two different frequencies: 1 Hz (top) and 10 Hz (bottom).

Conclusions

The following conclusions can be drawn:

1) Commercial purity Al and Al-2wt.% Al_2O_3 powders were successfully consolidated by hot extrusion at 400°C and cold rolled down to wires of 1 mm^2 in section without failures.
2) Al and composite samples showed an ultra-fine grained structure (about 200 nm).
3) High energy ball milling led to the fragmentation of the native oxide layer that covers the Al powders into nano-sized particles and favored the embedding of these phases in the Al matrix.
4) Both Al and nanocomposite wires take advantage of the strengthening effect of ultra fine grained microstructure and of the hard particles. A further strengthening and contribution to internal friction is given by nano-Al_2O_3 particles added ex-situ to the MMnC sample.
5) The MMnCs exhibited higher hardness (about 10%) and higher internal friction than the Al sample. In particular, the materials analyzed in this work show a remarkable damping behavior at high temperature. The tanδ (1Hz) at 275°C for monolithic commercially pure aluminum is about 0.007 [19]. The equivalent value of tanδ for Al sample is 0.025, while for the Al with 2 wt.% of nano-Al_2O_3 particles is 0.033.

The good workability shown by the MMnCs object of this work makes this kind of materials interesting for new fields of applications. For example, since they can be drawn down to small diameters, they could be weaved or embedded in high performance fabrics and nets. Moreover, the improved internal friction could be exploited to reduce high noise levels, premature fatigue failure and wear in all those application in which the material is subjected to vibrations.

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