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Mechanical Properties of Polycrystalline Copper and Single-Crystal LiF Initial Components for Composite System Cu/LiF

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Abstract—The paper deals with an investigation of mechanical properties and deformation features of polycrystalline copper and single-crystal LiF under dynamic nano/microindentation. It is shown that the values of hardness and Young's modulus depend on the magnitude of the applied load ($P_{\max}$): when the load is increased, H and E decrease. General regularities of the indenter penetration process in a wide range of loads are revealed: the appearance of a "pop-in" effect at the initial stage of the loading process, the formation of more pop-in steps with the growth in load, and the formation of pileups around the indentations. Such a nature of deformation is the result of sequential activation of different dislocation mechanisms with indenter deepening. Along with a great similarity in the specificity of deformation, some differences are noted at the unloading stage. The results serve to compare the mechanical properties of Cu and LiF individual components with similar parameters of the "coating/substrate" composite systems (CS Cu/LiF) produced on their basis.

Keywords: mechanical properties, specificity of deformation, copper, LiF, dynamic indentation

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INTRODUCTION

Thin metal layer/substrate composite systems (i.e., composite structures) (CSs) are widely used in modern electronics for production of integrated circuits, magnetic and optical devices, strain microsensors, etc. In this case, the thin microstructure of films and their mechanical properties become critical parameters that determine the accuracy and service life of manufactured instruments. Copper compounds are now often used in microelectronic devices in place of aluminum compounds since the conduction of copper is higher and its electromigration properties are better in comparison with aluminum. This fact is very important in the context of decreasing the dimensions of devices and energy consumption [1–5].

With the substitution of copper for aluminum, not only the electrical properties but also the mechanical properties of each microchip and the whole integrated circuit change since both the hardness and the elastic and adhesive behavior of these two elements are quite different. For this reason, in recent years, researchers have been studying mechanical properties in small volumes such as microelectromechanical systems (MEMSs) [1, 5, 6]. The task is not only to improve the reliability of devices. This problem is also fundamental because the mechanical microscale properties differ from the bulk properties of a material as a result of the size effect and the surface effect [1, 7].

Thus, nowadays, intensive studies of layer/substrate CSs are under way with the aim to ascertain the behavior of these structures when an external local load affects them, in particular, with micro- and nanoindentation [4, 8–14].

With reference to the aforesaid, when designing a layer/substrate CS, the mechanical properties of the initial components for a future layer/substrate pair must be studied to estimate what properties of the basic components determine the durability parameter and plastic nature of a new composite material. Different pairs for CSs are considered in the literature depending on the spheres of their application [11, 13, 15–17]. For instance, if the applied layer is used as a hardening or protective coating, one looks for a hard wear-resistant and chemically stable substance, and the layer/substrate pair will be a hard layer/soft substrate system. In optoelectronics, when designing MEMS devices, coatings of rather soft materials applied to a hard substrate are necessary, so a soft layer/hard substrate system is produced. Pairs with similar structural and plastic properties, i.e., hard layer/hard substrate and soft layer/soft substrate systems are interesting as well.

However, at present, the mechanical parameters of materials and possible pairs for CSs need to be studied and compared [8, 9, 11, 13]. Thus, the aim of this work is to study the mechanical properties of two separate substances, namely, polycrystalline copper and single-crystal LiF, and to create Cu/LiF CS on their basis. The results will be compared, and the expected properties of a future composite structure will be estimated.

EXPERIMENTAL

The initial components Cu and LiF under investigation have a great deal in common: they have cubic structure, low and quite similar values of microhardness ($H_B \approx 0.8$ GPa for Cu and $H_B \approx 1.1$ GPa for LiF), and fairly high plastic properties. This implies that Cu/LiF CS based on them will be a soft layer/soft substrate system. The formation of rosettes of dislocation around the indentations in the LiF substrate at T_{room} allows the analysis of a substrate response in relation to the size of load to the indenter at dynamic indentation of the Cu/LiF CS.

The mechanical properties were studied by the dynamic indentation method using a Nanotester-PMT3-NI–02 device fitted with a Berkovich indenter. The surface microstructure was investigated by the optical microscopy (OM) method using XJL-101 and Amplival devices, a Linnik MII-4 interferometer, and a Nanostation II atomic force microscope.

The following stages were carried out for each specimen during the tests for nano- and microindentation:

- The loading–unloading process for 18 maximum loads, $P_{\max} = 2$–900 mN according to the following pattern: loading for 20 s, holding at maximum load ($P_{\max}$) for 5 s, unloading for 20 s. Five indentations were applied for each load. The results were calculated as an average value of five tests.
- Then, for each specimen, the following curves were plotted: load versus depth of indentation, $P(h)$; Young's modulus as a function of load, $E(P)$; and hardness against load, $H(P)$. The curves of nano- and microindentation were analyzed and the morphology of the deformed zones around the indentations was studied; the specific behavior of the elastoplastic deformation of the materials under investigation was evaluated.
- Calculations were carried out automatically using the Oliver–Farr method [18] with the help of the device software.

Dynamic micro- and nanohardness (H) were computed from the formula

$$H = \frac{P_{\max}}{A_{\max}}, \tag{1}$$

where $P_{\max}$ is the maximum load to the Berkovich indenter and $A_{\max}$ is the projected indention area. Young's modulus (E) is determined according to the formulas

$$\frac{1}{E_r} = \frac{\left(1-\nu^2\right)}{E} + \frac{\left(1-\nu_i^2\right)}{E_i}, \tag{2}$$

$$E_r = \frac{\sqrt{\pi}S}{2\beta\sqrt{A_p}\,(h_c)}. \tag{3}$$

Here, E_r is the reduced elastic modulus, which is determined from formula (3); E and ν are the elastic modulus and the Poisson ratio of the specimen, respectively; and E_i and ν_i are the same coefficients for the indenter. The parameter $S = dP/dh$ is called contact rigidity, which is determined as a slope ratio of the curve $P(h)$ at the origin of the unloading area; β is the King correction factor for the Berkovich indenter, which is 1.034.

An ordinary selective etching reagent (aqueous low-concentration solution of $FeCl_3$) was used to develop dislocation structures around indentations on the LiF single crystals.

RESULTS AND DISCUSSION

Mechanical Properties and Behavior of Polycrystalline Copper at Nano/Microindentation

A specimen of Cu 10 × 10 × 2 mm in size was cut from a large high-purity copper ingot. The specimen surface was chemically polished in strong HNO_3 to remove a defective layer because of the mechanical processing and to develop a fine-grained structure, which indicated the absence of mechanical defects and internal stresses in the specimen.

The measured value of Young's modulus (E_{Cu}) slightly depends on the load, and it is about 125 GPa. As for hardness, it appears to be more sensitive to the change in load. Figure 1 presents hardness versus load, $H_{\text{Cu}}(P)$, for the copper specimen. It is seen that, with the decrease in load from 900 to 5 mN, the hardness increases by more than a factor of two with a knee on the curve within the load range from 100 to 80 mN, which is in agreement with work [1]. Such behavior of the hardness values with the change in the applied load is apparently due to the indentation size effect (ISE), which is typical of a large number of materials. The study of indentation structure showed the plastic behavior of deformation within the whole load range (Fig. 2). Natural evacuation of material to the surface in the form of hillocks at the center of sides makes the indentations slightly convex and causes bending in interference fringes, most noticeable at high loads (Figs. 2d, 2g).

The dependences $P(h)$ presented in Fig. 3 show that the process is rather homogeneous, particularly, at high loads, and demonstrate the weak relaxation of the material when the load is removed: the curve segment at the unloading stage runs nearly perpendicu-