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Anisotropic layered media with microinclusions: Thermal properties of arc-evaporation multilayer metal nitrides

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ABSTRACT

This study aims at the theoretical examination of the anisotropy of thermal conductivity $F = \kappa_{||}/\kappa_{\perp}$ that could be engineered in hard multilayer coatings, prepared using arc-evaporation. High values of F and thereby high lateral heat dissipation can reduce detrimental thermal gradients that emerge during cutting and friction processes in hard coatings. As coating deposition is widely done using arc-evaporation that leads to inclusion of droplets of different shapes and material in the coating, it is further evaluated to which extent F is be affected by these inclusions. The ideal continuous anisotropic medium with inclusions can be described using the effective medium Maxwell Garnett Approximation (MGA). A deposited multilayer structure presents a particular case with a limited number of layers inducing anisotropy and is simulated using the Finite Element Method (FEM) and compared to the idealized MGA predictions. The results show that the effect of droplets on the anisotropy F is strongly dependent on droplet shape, material and orientation. For spherical droplets at concentrations that are usually observed in arc-evaporation coatings, the value of F decreases linearly, proportional to the droplet concentration and almost independent of droplet thermal conductivity for regularly experimentally encountered droplet materials. When ellipsoidal droplets are considered, F depends strongly on the material and orientation of the droplets. The effects of finite thermal interface resistance between individual layers and around the droplet inclusions are evaluated separately and are found to be generally beneficial. This study shows that creating multilayers with high anisotropy of thermal conductivity should be possible even in the presence of unavoidable droplet inclusions. Furthermore, controlling metal droplet formation upon arc-evaporation of hard coatings can be used as a tool to engineer the anisotropy of thermal conductivity in arc-deposited multilayer coatings.

1. Introduction

Hard coatings are used in many different areas, for example for protection of automotive parts, cutting and forming tools as well as for optical and decorative applications. Hard multilayer metal nitride coatings have risen to prominence due to improved hardness, low friction and wear resistance [1]. However, stacking different materials can have a sizeable effect on anisotropy of thermal conductivity, a fact that has so far received little attention in studies on hard coatings. It has been proposed that anisotropy of thermal conductivity in a coating can reduce thermal gradients on the wear-sensitive interface between coating and substrate [2]. Thermal conductivity of industrially relevant hard coatings varies over a wide range [3–6]. Proper combination of these materials in a multilayer structure allows for the theoretical description and targeted design of coatings with high anisotropy. Direct measurement of anisotropy of thermal conductivity of such coatings on substrates is difficult, because of severe limitations of existing methods [7].

The majority of industrial deposition systems for hard multilayer coatings use the arc-evaporation method because of stable operation, high deposition rate and excellent coating adhesion [8]. However, in contrast to magnetron sputtering, an arc rapidly erodes the target material. The immediate local heating within a few nanoseconds, and the enormous current densities in the filamentary arc discharge, frequently lead to the ejection of μm-sized metal particles from the target. The resulting incorporation of metallic

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droplets, or macroparticles, is well known and has been extensively studied [9–15]. It is of interest to know how these isotropic, and often highly conductive metal inclusions, affect the anisotropy of arc-evaporated multilayer coatings, aimed to possess designed anisotropy.

There have been studies on the effect of isotropic inclusions in an anisotropic matrix [16–18]. These studies consider electrodynamics but the theory can be applied to the problem of heat transport as well. It is unknown whether these theoretical descriptions are applicable to layered structures with interface resistance and where anisotropy is induced by layers of similar size as the inclusions. This work studies the effect of size, concentration, chemical composition and shape of highly conductive inclusions, such as metallic droplets, on the thermal conductivity of anisotropic multilayer materials.

The paper is organized in the following way. At first, the material properties of industrially relevant hard multilayer coatings are reviewed and it is discussed how to represent their room temperature thermal and microstructural properties in model calculations (Section 2). The anisotropy that can be created in a multilayer structure of these materials is then described using a thermal resistor model (Section 3). To describe the effect of inclusions analytical and simulation approaches are presented. The analytical approach for a homogenous anisotropic medium with inclusions is based on the Maxwell Garnett Approximation (MGA, Section 4). This is followed up by a model simulation using the FEM approach (Section 5). Finally, the results of the thermal resistor model, the MGA analysis and the FEM simulations are compared and discussed, with focus on the possible fabrication of engineered multilayer coatings with designed anisotropy of thermal conductivity (Section 6).

2. Material properties of hard arc-evaporation multilayer coatings and arc droplets

Properties are evaluated for use in analytical evaluations and FEM simulations. The multilayer system is supposed to represent an industrially relevant hard coating produced in an industrial scale arc-evaporation deposition system.

The materials of the individual layers of the intended multilayer structure are thought to be isotropic and to have a thermal conductivity close to their bulk value. This means that residual strain in the coating has to be minimized as it could have a significant effect on thermal conductivity [19,20]. Oriented grain boundaries, or columnar growth along certain directions within the coating interfere with the direction of thermal transport and thus can induce an anisotropy of thermal conductivity in thin films of otherwise isotropic materials [21,22]. Transition metal nitrides used in the field of hard coatings most often crystallize in the cubic B1 structure with no discernible intrinsic anisotropy of thermal conductivity.

Thermal conductance G_l of interfaces between individual layers on the one hand and between multilayer structure and droplet G_d on the other hand are also relevant due to its capability to affect the heat flow. The layer materials are thought to be deposited in one continuous deposition process resulting in an intimate and atomically clean interface between the individual layers. In spite of the fact that overall roughness of arc-evaporation layers is generally high, at the nanometer scale the interfaces can be very well defined [23,24]. If the adjacent layer materials are cubic nitrides, their similar lattice structure and Debye temperature can be assumed to result in similar phonon spectra. While it is difficult to calculate or measure thermal conductance of a single interface, a combination of similar materials can be estimated by comparison [25–28] to result in a range for thermal interface conductance at room temperature of about $G_l \approx 300\ldots10{,}000$ MW m^{-2} K^{-1}. Low values of G_l could increase anisotropy as only perpendicular heat flow is impeded by layer stacking while theoretically the parallel heat flow should be unaffected. Thermal interface conductance between arc-evaporation coatings and droplets G_d has never been reported but can be supposed to be lower due to imperfect contact and stronger material difference. We estimate $G_d \approx 10\ldots1000$ MW m^{-2} K^{-1}.

Choice of individual layer thickness is limited for different reasons. Total coating thickness in the tooling industry is optimized to ≈5 μm. The layers should be as thin as possible in order to closely approximate a homogeneously anisotropic medium and to make the most of interface resistance between the layers. On the other hand, roughness in the arc-evaporated coatings is usually in the order of $R_a \approx 200$ nm [29]. In order to provide a close resemblance of the geometries of parallel layers (to promote anisotropy) we use this roughness value as a guide for the lower bound of individual layer thickness and selected 250 nm as the layer thickness for many of the FEM simulations. At this order of magnitude, degradation of thermal conductivity due to nanoscale effects such as boundary scattering and increased defect densities is not supposed to play a major role [30–35]. It is instructive to consider the limiting case of very small individual layer thickness: This would lead to very high defect densities and high interface scattering, eventually resulting in an alloy or an amorphous material with low but isotropic thermal conductivity.

The incorporation of droplets may have several detrimental consequences for both monolithic and multilayer coatings: (i) reduced hardness and strength, (ii) increased surface roughness, (iii) chemical non-homogeneity and (iv) reduced adhesion to the substrate [11]. Additionally, the droplets may also facilitate diffusion and hence reduce thermal and oxidation durability of the coating. There have been efforts to reduce the amount of droplets that are deposited using diverse techniques, such as magnetic filters [36], arc steering [37] or current pulsing [38], but they have the common disadvantage of negatively affecting deposition rates. The droplet size and concentration in the arc deposition process are dependent on the melting point and vapor pressure of the target material, but can be influenced also by arc steering installations, deposition parameters (pressure, temperature, gas flow) and target design [9]. The droplets are supposed to be metallic but may feature surface nitridation [11]. Droplet morphology is generally considered as spherical, with sizes ranging from 0.2 μm (Zr in ZrN [9]) to 0.5–2 μm (Ti in TiN [11]), or as ellipsoidal droplets, oriented perpendicular to the growth direction, for example Al in $(Al,Cr)_2O_3$ with an average radius of 1 μm and an excentricity of up to 0.95 [15]. The volume fraction of the droplets in the deposited coating is affected by the deposition parameters and geometry and is usually in the range of 1–5% but droplet concentrations of up to 10% have been observed [11,39,40].

3. Thermal resistor model for the anisotropy of a periodic multilayer structure

Absolute thermal resistance R of a rectangular slab of material is defined as thickness x parallel to heat flow divided by area A perpendicular to heat flow and thermal conductivity κ:

$$R = \frac{x}{A\kappa} \tag{1}$$

Consider two stacked sheets of material. The resulting in-plane (parallel to the sheets) and cross-plane absolute thermal resistance $R_{||}$ and $R_{\perp}$ are given as