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Indentation pop-in as a potential characterization of weakening effect in coating/substrate systems

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ABSTRACT

Thin hard coatings are generally used for surface enhancement. However, it was theoretically predicted that thin hard coatings may have a weakening effect of reducing the resistance to onset of plasticity in coating/substrate systems. Such weakening effect may also deteriorate the wear property of a coated system. In this study, we report a direct experimental validation of the weakening effect. Spherical nanoindentations were performed on both tungsten coated and uncoated single-crystalline silicon. The statistical distribution of indentation pop-in load reveals that plastic yielding initiates at a lower load in the coated silicon than in the uncoated one. A cross-sectional transmission electron microscopy (XTEM) observation demonstrates that the plastic deformation area in the coated silicon is larger than that in the uncoated one. These evidences prove the existence of weakening effect. Utilizing the recently reported finite element analysis models, the plastic yielding bearing capacity of coated and uncoated silicon was calculated based on the von Mises criterion. Good qualitative agreement is found between experimental results and the theoretical models.

1. Introduction

Hard coatings are widely used for surface strengthening [1,2]. However, theoretical studies proposed that thin hard coatings may reduce the resistance to onset of plasticity in a coating/substrate system, which is defined as the weakening effect. Using finite element method (FEM), Komvopoulos [3] was perhaps the first to notice that indentation of thin hard coatings on soft substrates may cause higher stresses in the substrate compared to uncoated cases. It was further pointed out that such higher stresses have a detrimental effect on the resistance to plastic yielding [4]. This phenomenon has also been studied by Sun et al. [5] using FEM for elastic–plastic contact between a rigid sphere and compliant substrates coated with hard TiN coating. It was found that the critical load of plastic yielding inception of the coated system is smaller than that of the uncoated substrate, indicating that hard coating weakens the system by reducing its load carrying capacity. Recently, Goltsberget et al. [6] and Goltsberg and Etsion [7] studied the plasticity onset in coated spheres loaded by a rigid flat using FEM. The results showed that the additional stress in the coated system, which causes the weakening effect, is closely related to elastic moduli mismatch between the coating and the substrate. Song et al. [8] studied the corresponding case of a coated flat indented by rigid sphere and obtained similar results to Refs. [6,7]. In very thin hard coating systems, the coating can hardly shield the substrate from applied loads due to the limited thickness. As a result substrate plastic yielding can cause premature failure in such coated systems. Using FEM simulation, Michler and Blank [9] and Holmberg et al. [10] showed that substrate plastic yielding can introduce higher stresses in the coating in both indentation and friction process and facilitate the generation of cracks. Based on FEM and wear test, Jungk et al. [11] indicated that substrate plasticity results in premature failure of a coated system which causes cracking and delamination of the coating material. Therefore, the weakening effect of substrate plastic yielding could possibly undermine the wear performance of a coated system. Rebholz et al. [12] experimentally studied the wear rate of Ti–Al–B–N coatings on soft stainless steel substrate. The results showed that the relatively hard coating with large elastic modulus had a higher wear rate than a coating with moderate hardness and modulus. In our recent study [13] of friction and wear performance of TiN and TiAlN coatings on copper (Cu), high speed steel (HSS) and cemented carbide (WC_Co), we found that the wear performance of the harder and more rigid TiAlN coating on Cu and HSS substrate is poorer than that of the TiN coating. This could be caused by the more severe plastic deformation in the substrate beneath the TiAlN coating due to larger modulus mismatch between coating and substrate, which results in weakening effect

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and early plastic yielding in the substrate. Although the weakening effect phenomenon has been well modeled in thin hard coating systems, the experimental validation is still insufficient. Our previous work [14], qualitatively validated the weakening effect using the elastic–plastic loading (EPL) index, which is defined as the ratio between the dissipated energy due to plastic deformation and the total energy consumed in the indentation [15], and showed experimentally that thin hard coatings could reduce the resistance to plastic yielding inception for coated systems. However, a direct observation and convincing validation of the weakening effect, which is main goal of this paper, have not been reported yet.

Material plastic deformation can be generally defined as a phenomenon that includes every irreversible deformation caused by dislocation, planar defects, phase transformation and so on [16]. Single-crystalline silicon undergoes, under high pressure, a series of phase transformations [17], which fit the above definition of plastic deformation. Therefore, premature phase transformation in the presence of thin hard coating on silicon substrate may help identify weakening effect, provided that the phase transformation itself can be easily detected. A possible option is to use nanoindentation test to cause phase transformation in single-crystalline silicon [16–18] and study the reported discontinuity (pop-in) behavior observed on the loading-displacement curves of such a test [18,19]. The exact mechanism of silicon pop-in behavior is still under some debate [20,21]. Chang and Zhang [22] and Beake et al. [23] regard pop-in as a signal for the onset of phase transformation from cubic diamond structure (Si-I) to metallic β-Sn structure along with volume reduction of 22%. According to some other studies, the pop-in behavior occurs after phase transformation from Si-I to Si-II and is caused by the material flow of metallic Si-II [24,25]. In spite of the above different opinions regarding the exact mechanism of pop-in, they all support the idea that stress-induced phase transformation (i.e. plastic deformation) is somehow the prerequisite to pop-in behavior. Hence, the critical indentation load, corresponding to the appearance of a pop-in event, could possibly be considered an easy evaluation tool of resistance to plastic deformation [26]. The main goal of the present study is therefore to characterize experimentally the weakening effect in hard coating/silicon substrate system by investigating its correlation to the pop-in critical load and comparing the experimental results with the theoretical models in Refs. [6,7].

2. Experimental methods

Several (1 0 0)-oriented single-crystalline silicon wafer samples with the dimensions of $10 \times 10 \times 0.5$ mm^3 were used as substrates for coating deposition. Tungsten coatings having a thickness t of 700 nm were deposited on the silicon samples by ion beam assisted deposition (IBAD) system [27]. For the deposition process, the vacuum chamber was first exhausted to a base vacuum pressure of 3.5×10^{-4} Pa to remove impurity gases containing oxygen and water vapor. Following this, working gas Ar was introduced into the chamber and pressurized to 1.0×10^{-2} Pa to start the ion arc. The silicon substrate samples were bombarded by the assist ion source for 10 min for decontamination prior to the coating process. During the deposition process, the working current of the ion sources was 80 mA with energy of 2500 eV. In order to obtain uncoated specimens for comparison, some silicon samples were masked with aluminum foil and were placed adjacent to the samples to be coated. The thickness of the foil was about 20 μm and the foil was slightly pressed to keep closely attached to the silicon wafer. This was done to minimize possible heat induced differences between the coated and uncoated samples.

Spherical nanoindentation tests (CSM Instrument Nanoindentation Tester NHT2) were carried out on both the coated and uncoated silicon samples using a spherical diamond tip of radius $R = 20$ μm, maximum indentation load of 480 mN and loading rate of 240 mN/min. Since the pop-in event occurs randomly and the pop-in load is scattered, a sequence of indentation tests is needed to obtain its statistical distribution. Toward this end four coated and four uncoated specimens were used and the indentation test on each specimen was repeated 50 times to obtain a total of 200 repetitions for both the coated and the uncoated silicon. The 50 repetitions on each single specimen were done in a 5×10 matrix and 20 μm apart to avoid possible overlapping.

Following the completion of the indentation tests, focused ion beam (FIB) and transmission electron microscope (TEM) were used to further analyze the structure beneath the indentation for both the coated and uncoated cases. FIB system (TESCAN LYRA 3 XMH) was used to prepare the XTEM samples from beneath the indentation of one coated and one uncoated specimen. The conventional FIB-XTEM preparation method [28] was applied to prepare the XTEM samples. A protective layer of platinum was deposited prior to the ion milling to protect the specimen surface from ion etching. TEM (JEOL 2011) was used to observe the microstructure to confirm the occurrence of phase transformation during the indentation tests. The areas of transformed structures were compared between coated and uncoated specimens to verify the weakening effect.

Finally, the nanohardness and elastic modulus of the tungsten coating and the silicon substrate were measured on the above nanoindentation system by a pyramidal Berkovich tip with a force of 2 mN using the Oliver–Pharr method [29]. For these measurements 20 repetitions were carried out on each sample in a 4×5 matrix and 20 μm apart. The Poisson's ratio of both the silicon substrate and tungsten coating is assumed to be 0.3 in the modulus measurement. The elastic modulus and Poisson's ratio of diamond indenter is 1100 GPa and 0.07.

3. Results and discussion

3.1. Statistical distribution of pop-in load, displacement and EPL index

Fig. 1 shows typical load–displacement curves that were obtained from spherical nanoindentation tests for coated and uncoated silicon samples. Since the tungsten coating is relatively thin compared with the indenter radius, the loading curves of the coated and uncoated cases are quite similar below the load of 200 mN. When the indentation load exceeds 200 mN, the

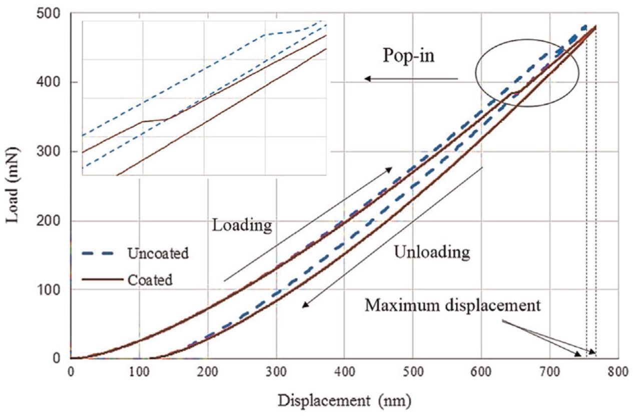

Fig. 1. Typical load–displacement curves of coated and uncoated silicon.