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Adhesion measurements for tungsten dust deposited on tungsten surfaces

G. Riva[a], P. Tolias[b,*], S. Ratynskaia[b], G. Daminelli[a], R. Donde[a], M. De Angeli[c], E. Vassallo[c], M. Pedroni[c]

[a] Institute of Condensed Matter Chemistry and Energy Technologies – Consiglio Nazionale delle Ricerche, via Cozzi 53,20125,Milan, Italy
[b] Space and Plasma Physics – KTH Royal Institute of Technology, Teknikringen 31,Stockholm,10044, Sweden
[c] Istituto di Fisica del Plasma – Consiglio Nazionale delle Ricerche, via Cozzi 53,20125,Milan, Italy

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ABSTRACT

The first experimental determination of the pull-off force for tungsten dust adhered to tungsten surfaces is reported. Dust deposition is conducted with gas dynamics methods in a manner that mimics sticking as it occurs in the tokamak environment. Adhesion measurements are carried out with the electrostatic detachment method. The adhesion strength is systematically characterized for spherical micron dust of different sizes and planar surfaces of varying roughness. The experimental pull-off force is nearly two orders of magnitude smaller than the predictions of contact mechanics models, but in strong agreement with the Van der Waals formula. A theoretical interpretation is provided that invokes the effects of nanometer-scale surface roughness for stiff materials such as tungsten.

1. Introduction

It has been recently recognized that adhesion plays a pivotal role in various tokamak issues concerning dust [1,2]. For instance, upon dust-wall mechanical impacts, adhesive work is responsible for a significant part of the overall dissipation of the normal dust velocity component [3–5]. Moreover, during loss-of-vacuum accidents, dust mobilization occurs when hydrodynamic forces overcome the net adhesive force [6,7]. Furthermore, under steady state or transient plasma conditions, dust remobilization takes place when plasma-induced forces exceed the net adhesive force, also known as pull-off force [8,9]. Finally, the quantification of the pull-off force is an essential step towards the development of *in situ* dust removal techniques suitable for future fusion devices such as ITER [10]. Nevertheless, to date, there have been no pull-off force measurements for reactor relevant materials.

Experimental techniques that characterize the strength of dust-surface adhesion are generally based on exerting a well-known force in a controlled environment until mobilization is observed [11]. The colloidal probe method of atomic force microscopy (AFM) measures the cantilever deflection at the detachment instant, which after careful calibration can be converted into a spring force [12,13]. The centrifuge detachment method employs the centrifugal force arising from a rapidly rotating surface [14]. The electrostatic detachment method employs the electrostatic force resulting from the interaction between an externally imposed electric field and the contact charge it induces on the conducting dust surface [15]. The colloidal probe method is the most accurate, but it involves single grain measurements and thus acquiring statistics can be very time-consuming [11]. On the contrary, the centrifuge and electrostatic detachment methods are less precise but involve multiple simultaneous measurements.

In this work we report on the first pull-off force measurements for tungsten dust adhered to tungsten surfaces carried out with the electrostatic detachment method. The dust grains were adhered to the W surfaces in a manner that realistically mimics dust sticking as it occurs in tokamaks [8]. The strength of adhesion has been characterized for different micrometer-range sizes of W dust deposited on W surfaces of varying roughness. Comparison with theory revealed that contact mechanics models overestimate the pull-off force by nearly two orders of magnitude, whereas microscopic Van der Waals models provide pull-off force values very close to the experimental. It is argued that this is the consequence of nanoscale roughness; for stiff metals such as tungsten, even the small-

* Corresponding author.
E-mail address: tolias@kth.se (P. Tolias).

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est departure from atomic smoothness can remarkably reduce the surface energy due to the extremely short range of metallic bonding.

2. Theoretical aspects

Different expressions for the sphere-plane pull-off force can be derived by two complementary theoretical descriptions of the contact of solid bodies. The microscopic description is applicable to non-deformable solids and considers the overall effect of Lennard–Jones type interactions [16], neglecting chemical bonding. On the other hand, the macroscopic description is applicable to deformable solids and only considers the effect of short-range forces of chemical bonding nature in the contact zone. The macroscopic description is more appropriate for atomically smooth, *i.e.* zero roughness perfectly planar or spherical, solids. In what follows, we provide a brief presentation of the microscopic and macroscopic descriptions for smooth materials and discuss the multifaceted effects of surface roughness separately.

In microscopic descriptions of the contact, the pull-off force is calculated from simple balance considerations. When chemical bonding is negligible, the pull-off force needs to counteract the overall interaction between the instantaneously induced and / or permanent multipoles inside the bodies, which constitutes the attractive Van der Waals interaction. For a spherical dust grain of radius $R_{\rm d}$ in the proximity of a planar surface, the Van der Waals force is given by [16]

$$F_{\rm po}^{\rm VdW} = \frac{A}{6z_0^2} R_{\rm d} \,, \qquad (1)$$

where $z_0(\ll R_{\rm d})$ is the distance of closest approach between the two surfaces and A is known as the Hamaker constant. When considering the contact of two identical smooth metals, z_0 can be assumed equal to the lattice parameter $a(= 3.16\,\text{Å}$ for W [17]). The Hamaker constant is generally calculated on the basis of the Lifshitz continuum theory. For identical metals embedded in vacuum, neglecting the temperature-dependent entropic term and assuming a collisionless free electron permittivity $\epsilon(\omega) = 1 - \omega_{\rm pe}^2/\omega^2$ we acquire $A \simeq [3/(16\sqrt{2})]\hbar\omega_{\rm pe}$ [18]. The plasma frequency of W is $\omega_{\rm pe} \sim 7 \times 10^{15}$ rad/s [19] leading to the estimate $A \sim 10^{-19}$ J, which is close to the value recommended in the literature $A \simeq 4 \times 10^{-19}$ J [16]. Note that the Van der Waals force is not important for smooth metals in intimate contact ($z_0 = a$), since the interaction due to metallic bonding (owing to the sharing of the delocalized valence electrons) is dominant [20].

In macroscopic descriptions of the contact, the pull-off force is calculated by the contact mechanics approach [21]. The interaction strength is indirectly considered via the work of adhesion (per unit area) defined by $\Delta\gamma = \gamma_1 + \gamma_2 - \Gamma$, where γ_i denotes the surface energy, Γ the interface energy and in the case of identical metals $\Gamma \simeq 0$, $\Delta\gamma \simeq 2\gamma$ [22]. The surface energy is externally adopted either from first principle calculations [23] or from experiments [24], for tungsten $\gamma = 4.36\,{\rm J/m^2}$. When ignoring plasticity, established contact mechanics models, in spite of their different assumptions and validity ranges, lead to a pull-off force of the form [25]

$$F_{\rm po}^{\rm CMA} = \xi_{\rm a}\pi\,\Delta\gamma R_{\rm d} \,, \qquad (2)$$

with $3/2 \le \xi_{\rm a} \le 2$ a dimensionless coefficient [26]. The Johnson–Kendall–Roberts (JKR) theory leads to the coefficient $\xi_{\rm a} = 3/2$ [27], whereas the Derjaguin–Muller–Toporov (DMT) theory leads to the coefficient $\xi_{\rm a} = 2$ [28]. The aforementioned adopted value of γ incorporates metallic bonding in an automatic manner and the above expression is appropriate for metals in intimate contact. We point out that metallic forces are extremely short range and they can be considered to be effectively zero already for distances larger than 1 nm [29]. Consequently, as metallic dust approaches a smooth metal surface, the interaction is initially of the Van der Waals type and switches to the metallic type, which is stronger by orders of magnitude, only for distances close to the lattice parameter [29].

Surface roughness is known to significantly modify the pull-off force. Its presence alters many aspects of the contact and its effects can be categorized in the following manner: (I) Pure geometrical effects that occur due to changes in the local curvature of the bodies and their point-point separation. They have been considered in microscopic descriptions by decomposing the interaction into a contact term with the spherical asperity and a non-contact term with the underlying plane, where the statistically varying asperity parameters are expressed with the aid of measurable roughness characteristics [30,31]. (II) Deformation effects that occur due to the existence of different asperity heights, which lead to a competition between the compressive elastic forces exerted by the higher asperities and the adhesive forces exerted by the lower asperities. The former tend to detach the contacting bodies, effectively reducing the pull-off force [22]. Such effects have been considered in macroscopic descriptions by applying the JKR theory to individual asperity micro-contacts, assuming a Gaussian distribution for their height with respect to the average plane and summing up the force contributions [32]. They can be expected to be important for stiff materials with large elastic moduli. Refractory metals are characterized by a large Young's modulus and tungsten, in particular, has one of the largest values, $E \simeq 410\,{\rm GPa}$ in room temperature. (III) Bond switching effects that occur when the asperity dimensions are larger than or comparable to the range of interatomic forces. In this case, some parts of the bodies interact via weak Van der Waals forces and other parts of the bodies form strong chemical bonds.

Even mirror-polished tungsten surfaces are characterized by root-mean square (rms) roughness $R_{\rm q}$ that significantly exceeds the metallic bond range. Plasma exposed surfaces and tokamak-born dust can be expected to have $R_{\rm q} \gg 1\,{\rm nm}$. Therefore, we can safely assume that interaction via metallic bonding is limited in a very small fraction of the contact area and that it is further effectively reduced by deformation effects. This suggests that interaction via Van der Waals forces is dominant. Finally, for simplicity and as a crude approximation, we can neglect pure geometrical effects and employ Eq. (1) for the pull-off force.

3. Experimental aspects

The electrostatic detachment of micron-size metallic dust from metallic surfaces requires the application of strong fields that may lead to dielectric breakdown. Since low pressures can significantly increase the breakdown voltage, the experiments were conducted into a vacuum chamber with a pressure < 0.05Pa. This also eliminates humidity, known to affect pull-off force measurements [25]. The electrostatic field was generated by two parallel electrodes, see Fig. 1 for a schematic representation.

Electrostatic detachment. The configuration can be idealized as consisting of a rigid spherical conductor in contact with a grounded plane in the presence of a uniform normal electrostatic field. For this geometry, the Laplace equation for the potential can be analytically solved with the aid of degenerate bi-spherical coordinates. In cgs units, the contact charge of the sphere is given by the expression $Q_{\rm d} = -\zeta(2)R_{\rm d}^2E$ and the repelling normal electrostatic force acting on the sphere by $F_{\rm e} = [(1/6) + \zeta(3)]R_{\rm d}^2E^2$, where $\zeta(\,\cdot\,)$ denotes Riemann's zeta function [33]. The expression can be rewritten as

$$F_{\rm e} = kE^2R_{\rm d}^2 \;\; (\mu{\rm N}) \,, \qquad (3)$$

with $k = 1.52 \times 10^{-4}(\mu{\rm N\,mm^2})/({\rm kV^2\,\mu m^2})$, the field expressed in kV/mm and the radius in μm. Owing to $F_{\rm e} \propto E^2R_{\rm d}^2$ and $F_{\rm po} \propto R_{\rm d}$, force balance leads to $E \propto 1/\sqrt{R_{\rm d}}$ for the electrostatic field. Hence,