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Sputtering of sodium and potassium from nepheline: Secondary ion yields and velocity spectra

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ABSTRACT

Silicates are the dominant surface material of many Solar System objects, which are exposed to ion bombardment by solar wind ions and cosmic rays. Induced physico-chemical processes include sputtering which can contribute to the formation of an exosphere. We have measured sputtering yields and velocity spectra of secondary ions ejected from nepheline, an aluminosilicate thought to be a good analogue for Mercury's surface, as a laboratory approach to understand the evolution of silicate surfaces and the presence of Na and K vapor in the exosphere. Experiments were performed with highly charged ion beams (keV/u-MeV/u) delivered by GANIL using an imaging XY-TOF-SIMS device under UHV conditions. The fluence dependence of sputtering yields gives information about the evolution of surface stoichiometry during irradiation. From the energy distributions N(E) of sputtered particles, the fraction of particles which could escape from the gravitational field of Mercury, and of those falling back and possibly contributing to populate the exosphere can be roughly estimated.

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1. Introduction

Among the three main classes of solid materials in space (ices, silicates and carbon- based ones), silicates are the most abundant and present throughout the Solar System and the interstellar medium [1]. Silicates are materials with a large diversity in chemical composition and structural properties and play an important function in the cosmic life cycle of matter [2]. In several silicates as it is the case of nepheline, the Si atom is coordinated in a tetrahedron by four O atoms, and different structures are formed linking the radical $[SiO_4]^{4-}$ with different atoms which should compensate the negative charge. Silicates are found both in amorphous and crystalline phases [3].

In the Solar System, silicates are present in planets and their moons [4], in transneptunian objects, in asteroids and meteorites, and in comets [5,6]. Silicates are the dominant component at Mercury's crust, even more abundant than iron. Sputtered particles

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http://dx.doi.org/10.1016/j.nimb.2017.01.042 0168-583X/© 2017 Elsevier B.V. All rights reserved. can contribute to the formation of Mercury's active exosphere. The particle environment surrounding Mercury is complex and composed by i) thermal and directional neutral atoms originating via surface release and charge-exchange processes, and ii) ionized particles originating from photo-ionization and from surface release processes such as ion induced sputtering. Indeed, sputtering of the components of minerals present at the surfaces of Mercury and the Moon appears to be the most efficient source of sodium and potassium in their exosphere [7]. Na⁺ and K⁺ ions have been observed in Mercury's exosphere by the MESSENGER spacecraft [8].

Ion irradiation of solids can lead to physico-chemical changes. Among them are structural modification (e.g. amorphization) and sputtering of charged species [9,10]. These effects induced by energetic ions have been studied in the laboratory with the aim to simulate solar wind ions and cosmic ray induced modifications. For instance, ion implantation in silicates was studied by Strazzulla et al. [11] to simulate formation of molecules containing the projectile atoms. Sputtering of silicates was analyzed in order to investigate the importance of solar wind ions on this process [12] and to simulate the alteration of regoliths of outer Solar System bodies

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[13]. Nepheline is an aluminium silicate containing sodium and potassium with the chemical formula (Na,K)Al₄SiO₄. It is well suited for simulating the surface of several objects in our Solar System, like Mercury [14,15]. In addition, its composition and structure are well known [16] and thus nepheline is a good candidate to investigate the origin of and also the ratio between Na and K in the exosphere of Mercury.

Here, we report results obtained by irradiating nepheline samples with heavy ions accelerated to low (keV) and high (MeV) energies. Experiments with nickel and germanium beams, with atomic numbers close to that of iron, allow indeed a good simulation of effects induced by the heavy ion fraction (a few percent) of cosmic rays. Experiments with low energy xenon beams are a first step towards laboratory simulations of solar wind impact. Xenon is present in the solar wind, albeit in very small quantities ($\sim 10^{11}$ times smaller than protons), but it can be considered a template for heavy ions (essentially all of the ions heavier than He).

2. Experimental details

The experiments were performed at the *Grand Accélérateur* National d'Ions Lourds – GANIL in Caen. The experimental set-up called AODO was mounted successively on the "medium energy" beam line SME (*Sortie Moyenne Energie*) and on the low energy facility ARIBE. AODO is dedicated to the study of the secondary ions emitted from targets prepared *in situ*, in order to minimize surface contaminations, in ultrahigh vacuum (pressure $< 2 \times 10^{-9}$ mbar). This set-up has been described elsewhere [17–19].

The nepheline sample was prepared ex situ in a high vacuum chamber (10^{-6} mbar) by electron beam deposition method using natural mineral as target to be evaporated. The electrons emitted thermally by tungsten filament are accelerated at 8 kV and directed to the target by a controllable magnetic field. This allows the complete melting and evaporation of nepheline that occurs very quickly at about 1400 K. The nepheline vapor expands from the source until it condenses onto a cold substrate. Since the cloud is homogeneous, the uniformity of the thickness of the deposited film only depends on the distance from the source and on the solid angle of the cloud that is intercepted by the substrate. Thus, with this technique we can produce thin uniform films of the evaporated material with final stoichiometry identical to the target materials. The targets were heated in situ in the ultrahigh vacuum chamber during 48 h to desorb surface contamination [17–19]. The target thickness was of the order of 1 µm.

The experiments were performed with slow highly charged Xe¹⁵⁺ (225 keV) and Xe²⁶⁺ (390 keV) ions at ARIBE and with swift heavy Ge²⁸⁺ (690 MeV) and Ni²⁴⁺ (630 MeV) ions of charge state close to equilibrium at SME (see Table 1). Spectra obtained with Xe¹⁵⁺ (225 keV) and Xe²⁶⁺ (390 keV) are quite similar. The same is true for the ones obtained with Ge²⁸⁺ (690 MeV) and Ni²⁴⁺ (630 MeV). Therefore, for simplicity and avoiding repetition, only the spectra obtained with Xe¹⁵⁺ (225 keV) as a "low energy" example and the spectra with Ni²⁴⁺ (630 MeV) as a "high energy example" are shown here.

Table 1

Ion beams, projectile energy (E), electronic (Se) and nuclear (Sn) stopping power and projectile fluences. The stopping power was calculated with the SRIM software [21]. A density value of 2.64 g/cm^3 was adopted.

Ion Beam	E (MeV)	Se (keV/nm)	Sn (keV/nm)	Fluence (particles cm ⁻²)
Xe ¹⁵⁺	0.225	0.3	1.6	10 ¹² -10 ¹⁴
Xe ²⁶⁺	0.390	0.4	1.5	5×10^{12} to 5×10^{13}
⁷⁶ Ge ²⁸⁺	690	7.0	0.004	$5 imes 10^9$
⁵⁸ Ni ²⁴⁺	630	5.3	0.003	$10^{10}6\times10^{12}$



Fig. 1. Schematic representation of the XY-TOF-SIMS experimental set-up.

An outline of the XY-TOF-SIMS method is shown in Fig. 1. The ion beam impinges on the target inside a vacuum irradiation chamber. The sputtered ions are extracted by means of an electrostatic field and directed onto a position-sensitive Micro-Channel-Plate (MCP) detector. The induced electron avalanche at the position of the secondary ion impact generates a fast "STOP" signal. Also, the electron avalanche is collected by a XY-delay-line anode, from which the impact position (X, Y) can be determined. Secondary electrons emitted upon projectile impact on the Al foil placed in the beam for MeV/u projectiles, or a pulse applied to parallel plates to deflect the low energy ion beams (this latter case is shown in Fig. 1) provide a fast "START signal". From the two fast "START" and "STOP" signals, the time of flight of the secondary ions can be determined and converted into mass spectra. Together with the impact position, the complete velocity vector of emitted secondary ions is known and e.g. velocity distributions can be calculated.



Fig. 2. TOF mass spectra from nepheline by low-energy Xe¹⁵⁺ ion beams at 225 keV (bottom) and high-energy Ni²⁴⁺ ion beams at 630 MeV (top). Inset: mass spectrum obtained by the MESSENGER's FIPS device [8].

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