Study of the (010) olivine surface by Rutherford backscattering spectrometry in channeling geometry

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ABSTRACT

The (010) surfaces of forsterite (Fo100) and natural forsteritic olivine (Fo90) single crystals have been studied by Rutherford backscattering spectrometry (RBS). Spectra were collected either in channeling geometry with the incident beam parallel to the [010] zone axis, or in a random orientation. In both materials, two surface preparations were examined: (1) mechanical polishing and (2) chemical etching by hydrofluoric acid after mechanical polishing. Composition profiles extending to several hundreds of nanometers below the surface were probed by RBS in random mode. Simulations of all the spectra indicated constant major element compositions, equal to the bulk compositions of the crystals. Characteristics of the few top atomic layers were probed by RBS in channeling mode. The crystalline quality of the surface of chemically etched samples has been evaluated quantitatively and is shown to be much better than that of mechanically polished samples. Given the energy resolution, we estimate that the bulk crystalline quality is perturbed for more than 40 nm below the surface in the case of mechanical polishing whereas disorder is limited to a topmost layer thinner than 1 nm in the case of chemical etching. On average, for the chemically etched samples, less than one atom per [010] atomic row is displaced from its mean crystallographic position. The bulk stoichiometry is preserved in the topmost layer of pure forsterite whereas a slightly higher Fe/Mg ratio is detected at the surface of chemically etched Fo90. A method for the quantitative assessment of surface quality of olivine is thus proposed, constituting a useful preliminary step before any study of surface modifications of olivine subjected to various geological conditions.

INTRODUCTION

The reactivity of silicate minerals under geological conditions is largely conditioned by the structure and chemistry of interfaces in contact with surrounding solids or fluids. These interfaces are key parameters of many natural processes such as biotic or aqueous weathering of minerals, space weathering, and interstellar chemistry. They may also have played a role in prebiotic syntheses of organic molecules and in the origin of life (e.g., Banfield and Hamers 1997). A better fundamental understanding of mineral surfaces and interfaces requires, among other approaches, a model of the surfaces of silicates at an atomic scale.

The simplest possible mineral surfaces are made up of the few outermost atomic layers in which atoms are slightly displaced from their crystallographic positions in the bulk material. Quantum chemistry calculations provide first models of “ideal” mineral surfaces at an atomic scale (e.g., Parker et al. 1997; Watson et al. 1997). In comparison with surfaces of simple oxides such as MgO, Fe2O3, and SiO2, the surfaces of silicates are not easily amenable to numerical simulation because of the complexity of their chemical compositions, crystallographic structures and distributions of electric charges. Additionally, most natural crystals are covered by a thin reaction layer formed when the mineral evolves from the thermodynamic conditions of its genesis to those of its actual geological setting at the Earth’s surface.

Our goal was to prepare silicate samples of several millimeters square dimensions having a high quality crystallographic arrangement of the outer atomic layers and being as smooth as possible at the atomic scale. The preparation of millimeter-sized silicate surfaces requires initial substrates that are not commercially available. In this study, we chose to prepare the dense (010) olivine surface because olivine is an ubiquitous phase in the Earth as well as in extraterrestrial materials and its (010) faces are among the most prevalent in natural samples. In addition, single crystals of gem quality are easily available and olivine has a relatively simple major element composition in comparison to other silicates.

In principle, many techniques can be applied to process an initial substrate in order to obtain clean surfaces (Rabinzohn et al. 1984): mechanical polishing, thermal annealing under fO2-controlled conditions, chemical etching, chemo-mechanical polishing (e.g., Siton is commonly used in commercial sur-