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Predicting olivine composition using Raman spectroscopy through band shift and multivariate analyses

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Abstract

Olivine group minerals are ubiquitous in extrusive igneous rocks and play an important role in constraining equilibria for samples in the upper mantle and above. All Raman spectra of the olivine group minerals in the solid solution between forsterite (Fo, Mg_2SiO_4) and fayalite (Fa, Fe_2SiO_4) have a high-intensity doublet between 800 and 880 cm^{-1}. Previous studies used small sample suites with limited compositional ranges and varying spectrometers to relate energy shifts of these two bands to Mg/Fe contents. In this work, Raman spectra of 93 olivine samples were acquired on either Bruker's 532 nm (laser wavelength) Senterra or BRAVO (785/852.3 nm) spectrometer. This paper compares the two-peak band shift univariate method with two multivariate methods: partial least squares (PLS) and the least absolute shrinkage operator (Lasso). Data sets from several instruments are also examined to assess the most accurate method for predicting olivine composition from a Raman spectrum.

Our 181-spectra PLS model is recommended for use when determining olivine composition from a Raman spectrum. For Raman spectra of mixed phases where only the olivine doublet can be identified, composition can best be determined using the position of the peak ca. 838–857 cm^{-1} through use of the regression equation $\%Fo = -0.179625x^2 + 310.077x - 133\,717$ (where x = DB2 centroid in units of cm^{-1}).

In situ methods for predicting mineral composition on planetary surfaces are critically important to extraterrestrial exploration going forward; of these, Raman spectroscopy is likely the best, as shown by the impending deployment of several Raman instruments to Mars (ExoMars and Mars 2020). More broadly, application of machine learning methods to spectral data processing have implications to multiple fields that use spectroscopic data.

Keywords: Raman spectroscopy, olivine, forsterite, fayalite, PLS, Lasso