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IT-9-P-2894 Analysis of the ordering state of pyroxenes using precession electron diffraction.

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The precession electron diffraction (PED) technique [1] has been originally developed for structure determination at a submicrometer scale in a transmission electron microscope (TEM). Since, many structures have been solved using PED, recently combined with the tomographic acquisition of 3D electron diffraction data [2]. Using PED, integrated intensities of the diffracted beams as a function of the rocking beam orientation are collected. The resulting intensities keep dynamical in nature, due to residual multiple scattering, but are more closely related to the strength of the scattering events and ranking of reflections as a function of their intensities is generally correlated to the structure factor values, which is crucial for structure solution.

Recently, it has been shown that PED could also be used for structure refinement [3]. In this case, experimental intensities have to be compared with dynamical simulations of diffracted intensities, taking into account the multiple scattering occurring when the electron beam is passing through the crystal. Applied to structures with mixed occupancies, the analysis can be used to refine atomic occupancies of specific sites of the structure, giving access to the ordering parameter. In the field of mineralogy, the PED refinement has thus been used to analyze the ordering state of orthopyroxene (OPX) samples. Results have enabled the distinction between an equilibrated sample (natural OPX ($Mg_{0.60}Fe_{1.40}Si_2O_6$) and a non equilibrated one (heat-treated (1000°C, 48h) and quenched sample from the same origin), giving ordering parameter values in good agreement with those obtained at the grain scale using XRD [4].

To go further and use PED data to decipher the thermal history of the sample with sufficient precision, the sensitivity of the PED refinement method still appeal for a detailed quantitative evaluation. In this work we discuss the influence of experimental parameters such as the irradiation dose and/or heating of the sample under the electron beam. Analyses are performed on the previously studied equilibrated OPX sample. Our results show a noticeable evolution of the ordering parameter with the electron beam irradiation duration (Fig. 1), which assesses for the high sensitivity of the technique. Possible evolution of the ordering state associated with the in-situ heating of the sample will also be explored, opening the road to the study of intra-crystalline diffusion kinetics at a very local scale in a TEM using PED.

[1] R. Vincent and P.A. Midgley, Ultramicroscopy 53 (1994) p. 271. [2] U. Kolb et al., Crystal Research and Technology 46 (2011), p. 542. [3] L. Palatinus et al. Acta Crystallographica A (2013), 69(2), P. 171. [4] D. Jacob et al., American Mineralogist 98 (2013) p.152.

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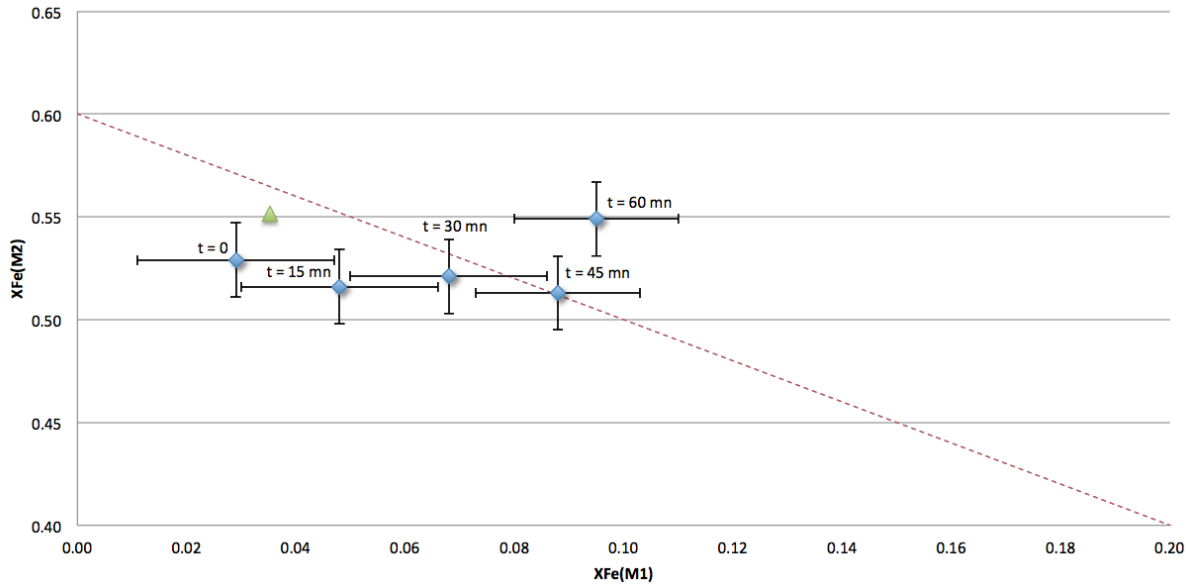

Fig. 1: Plot of XFe(M2) vs. XFe(M1) in a natural OPX sample as obtained from PED dynamical refinement as a function of the duration of the electron beam illumination (200kV, LaB6 Tecnai 20 microscope). Dashed line corresponds to the constant composition line. Green triangle corresponds to XRD results obtained at the grain scale