



Contribution ID: 44

Type: POSTER

## Magnetic-Electronic Pressure Response of Ilmenite (FeTiO<sub>3</sub>)

Polycrystalline powders of synthetic and natural ilmenite (FeTiO<sub>3</sub>) assemblages pressurized in a diamond anvil cell have been studied at room temperature using <sup>57</sup>Fe Mössbauer spectroscopy to elucidate the magnetic-electronic pressure response of different ilmenite samples. Natural ilmenite samples used included an untreated sample derived from the Hillindale mining area in KwaZulu-Natal, South Africa and a heat treated sample (to increase oxidation) from the same source. Asymmetry in the Mössbauer lineshape (doublet) profile is initiated at low pressure and persists to the highest pressure in all samples. This is especially evident in the synthetic sample which has a symmetric doublet (and only Fe<sup>2+</sup>) at ambient pressure. The asymmetry is perhaps attributed to Fe<sup>3+</sup> emerging (i.e., pressure-induced oxidation). Metal-metal charge transfer along the c-axis of the unit cell between face sharing octahedra of Fe<sup>2+</sup> and Ti<sup>4+</sup> cations may be one, albeit, contentious explanation for this [1,2]. If we consider the asymmetry in the relative intensities of the doublet to be a result of an increase in the ferric-ferrous ratio from charge transfer (and not from texture effects or the GKE), then the following quantitative behavior is obtained. The Fe<sup>3+</sup>/Fe<sup>2+</sup> ratio in the heat treated sample as deduced from the theoretical fits to the data shows a gradual increase of 0.28 to 0.38 in the pressure range 0 GPa to 14 GPa, the highest pressure reached for that sample. The phase abundance of the ferric component in the synthetic sample increases from 0 to 15% at 18.5 GPa, then seems to decrease at higher pressure as a structural transition ensues. A perovskite high pressure phase initiates at ~18 GPa corresponding to Fe<sup>2+</sup> in dodecahedral coordination. It coexists in ever increasing abundance, and over a wide pressure range with the corundum-type low-pressure phase (i.e., sluggish transition at room temperature). The relative content of Fe<sup>2+</sup> in the perovskite phase increases at the expense of Fe<sup>2+</sup> in the low-pressure ilmenite phase. The trend observed in the abundances of the ferric and ferrous components of the ilmenite-corundum structure of the untreated natural sample is similar to what is seen in both the heat treated and synthetic samples. In the low pressure region (up to 4 GPa), the Fe<sup>3+</sup>/Fe<sup>2+</sup> ratio in the untreated natural sample increases significantly and thereafter continues to increase into a “plateau” region. The ratio is ~0.10 at ambient conditions and ~0.30 at 15 GPa, the highest pressure attained in the study of this sample.

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yes

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**Track Classification:** Magnetism and Magnetic materials - Bulk and thin layers