Synthesis and Characterization of the Oxynitride $\text{Y}_2\text{Mo}_2\text{O}_{4.5}\text{N}_{2.5}$ Pyrochlore: A Neutron Diffraction and Magnetic Study

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A new molybdenum oxynitride $\text{Y}_2\text{Mo}_2\text{O}_{4.5}\text{N}_{2.5}$ with cubic pyrochlore structure ($a = 10.3350(2) \ \text{Å}$, space group $Fd\bar{3}m$, $Z = 8$) has been synthesized by heating the parent $\text{Y}_2\text{Mo}_2\text{O}_7$ oxide in flowing ammonia at 898 K. The polycrystalline sample has been characterized by thermal analysis, X-ray and neutron diffraction (NPD), and magnetic susceptibility measurements. The analysis of high resolution NPD data, based on the contrast existing between the scattering lengths of O and N, shows that both atoms are distributed at random at the anion substructure; the refined crystallographic formula implies an oxidation state of +5.25 for Mo cations. The thermogravimetric curve shows a weight gain of 7.5% at 1000 K in air, corresponding to the complete elimination of $\text{N}_2$ and oxidation to Mo(VI) oxide, in good agreement with the proposed composition. The magnetic susceptibility exhibits a Pauli-like, temperature-independent term which derives from the partial delocalization of Mo electrons on Mo-(O,N) bands with a broader bandwidth, as a consequence of the significant opening of the Mo-(O,N)-Mo angle and strengthening of the Mo-(O,N) interactions with respect to the parent $\text{Y}_2\text{Mo}_2\text{O}_7$ oxide. As in this oxide, a reminiscent spin-glass behaviour is observed at low temperature.

Key words: Oxynitride, Pyrochlore Structure, Neutron Diffraction, Ammonolysis, Pauli Susceptibility