The study of oxygen vacancy ordering in the LaSr1-xMnOy system has shown a strong correlation between Mn formal valence and coordination to oxygen. The Mn2+ was found forming octahedra (Oc) and tetrahedra (Te), Mn3+ octahedra and pyramids (Py) and Mn4+ only octahedra. This tendency was derived from the presence of three kinds of vacancy orderings in the system. For y>2.5 and the average Mn charge of 2<m<3 (x>0.5), brownmillerite-type structures are found with Mn2+/3+ Oc and Mn2+ Te. [1] For 3<m<4 (x<0.5) structures of the homologous Sr4+nMn4+O10+3n –type series are found with Mn3+ Py and Mn3+/4+ Oc. [2] For y<2.5 and 2<m<3 (x<0.5) a complex layered structure with OcTeOcOcTe'Oc ordering and step defects of Mn3+ Py and Mn2+ Te [3] is observed.

Brownmillerite-type structure is absent in the Sr-rich region since mostly Mn3+ is present, which does not show tendency to form Te. Compounds of the Sr4+nMn4+O10+3n series have been described as arrangements of groups of four Py and n Oc in symmetrical patterns. [2] This description did not elucidate the crystal chemistry reasons for specific pattern (n=0, 1 and 3) since it neglected the coordination stabilization associated with oxygen vacancy, charge and orbital ordering observed in the structures. [2] Using high-resolution transmission electron microscopy (HR-TEM) for SrMnOy with y values located between corresponding n=0, 1 and 3 compositions, we have determined that the oxygen vacancy ordering directs the formation of these patterns. The structural patterns can be described as perovskites with lines of oxygen vacancies along [001] with nearest lines of vacancies in the cubic (310) plane. Successive (310) planes are n perovskite blocks (Oc) apart in the [010] direction. This ordering pattern allows the coherent growth of phases with different n at the sides of (310) plane as observed in grains of a sample of SrMnO2.668 where n=1 and n=3 phases grown coherently one at each side of the vacancy plane.


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