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New route to double perovskite oxides using the mixture of oxalate precursors

Marijana Jurić¹, Jasminka Popović¹, Lidija Androš Dubraja¹, Filip Torić², Damir Pajić²

1. Ruđer Bošković Institute, Zagreb, Croatia
2. Department of Physics, Faculty of Science, University of Zagreb, Zagreb, Croatia

email: Marijana.Juric@irb.hr

Properties of the mixed-metal oxides could be highly affected by the effect of crystallinity, particle size, phase composition and morphology. These can be tuned in part by changing the synthesis methods.

The possibility of using metal–organic coordination systems through the thermal decomposition process as molecular precursors in the synthesis of nanomaterials with high surface and specific morphology has been considered only recently. It has been observed that the use of a well-defined heterometallic precursor can produce crystalline oxide materials under conditions that are significantly milder than those applied in traditional solid-state synthesis. Also, the single-source precursors provide better control over the stoichiometry of the metal ions in the final products as well as the homogeneity of the materials due to the mixing of the metals at the molecular level. The existence of bridging or chelating ligands in the precursors prevents metal separation during oxide formation. For example, the $C_2O_4^{2-}$ anion easily decomposes to the vapour phases CO_2 and CO , by the low-temperature routes, and hence, heterometallic oxalate complexes are very convenient for the preparation of mixed metal oxides.^[1]

Most of the perovskite compounds that have potential technological interests are not simple systems, but rather ternary oxides such as $A(B'B'')O_3$. Heterometallic oxalate complexes do not always contain the appropriate stoichiometry for the formation of the desired single phase oxide. So, we have tested whether the multimetallic oxides containing two or more metals could be prepared by mixing two or more different oxalate precursor in various ratios prior to thermal decomposition.^[2]

A highly crystalline materials $Ba(M_{1/3-x}M'_xNb_{2/3}^V)O_3$ ($M = Ni^{II}$, $M' = Co^{II}$; $x = 0–1/3$) were obtained after thermal decomposition of the mixture of the well-defined and structurally characterized heterometallic oxalate-based compounds $Ba_2(H_2O)_5[NbO(C_2O_4)_3]HC_2O_4 \cdot H_2O$,^[1] $[Ni(bpy)_3]_2[NbO(C_2O_4)_3]Cl \cdot 12H_2O$ ^[3] and $[Co(bpy)_3]_2[NbO(C_2O_4)_3]Cl \cdot 12H_2O$,^[3] grinded in an agate mortar in different ratios.

The phase formation and structural ordering of the tri- or tetrametallic perovskite oxides obtained by this modified molecular precursor route have been characterized by powder X-ray diffraction, scanning electron microscopy and energy-dispersive X-ray spectroscopy. The magnetic properties of newly prepared materials which adopt the disordered cubic structure (with random distribution of B' and B'' ions) have been also investigated.

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