REACTIVE FLASH SINTERING OF HIGH ENTROPY (Mn_{0.2}Co_{0.2}Ni_{0.2}Cu_{0.2}X_{0.2})Fe₂O₄ (X=Fe, Mg) SPINEL OXIDES

Alejandro F. Manchón-Gordón, ICMSE CSIC-Universidad de Sevilla, Spain afmanchon@us.es Sandra Molina-Molina, ICMSE CSIC-Universidad de Sevilla, Spain Antonio Perejón, ICMSE CSIC-Universidad de Sevilla; Universidad de Sevilla, Spain Javier S. Blázquez, ICMSE-CSIC, Universidad de Sevilla, Spain Pedro E. Sánchez-Jiménez, ICMSE CSIC-Universidad de Sevilla; Universidad de Sevilla, Spain Luis A. Pérez-Maqueda, ICMSE CSIC-Universidad de Sevilla, Spain

Key Words: Reactive flash sintering, Strontium ferrite; Permanent magnets, Mössbauer spectroscopy, Magnetic properties.

The possibilities of finding new element combinations that cause a meaningful improvement of materials' properties was getting exhausted, which motivated the search for completely new routes for new materials development. This change of philosophy arrived with the concept of high-entropy alloys (HEAs) [1], which utilize multiple principal elements (five or more) in relatively high concentrations to form materials with a high entropy of mixing. Therefore, HEAs encompass a vast compositional space, which provides a large window of promising opportunities for discovery of new alloys with valuable properties. This strategy of enhancing configurational entropy to promote single phase systems from multiphase precursors has been extended to other type of materials and has also been successfully used even in oxides with five cations, extending the HEA idea beyond alloys to ceramics [2]. Since then, high entropy oxides (HEOs) have attracted great attention and several HEOs with different structures, microstructures, compositions and, most importantly, tailorable properties, have been developed [3].

In this work, (Mn_{0.2}Co_{0.2}Ni_{0.2}Cu_{0.2}X_{0.2})Fe₂O₄ (X=Fe, Mg) spinel high-entropy oxides were prepared by reactive flash sintering (RFS). Previous studies show that fabrication of such materials by conventional pressure-less sintering was heated at 1523 K for 10 h [4]. Here, pure single phase was successfully formed from mixed oxide powders by reactive flash sintering after only 30 min at 1173 K. The structural, magnetic and electrical properties of the produced compounds were evaluated and compared with those of the counterparts that were conventionally sintered, analyzing the interlink between the functional properties and crystal structure. Both compositions exhibit a clear soft magnetic behavior, with very low coercive fields and saturation magnetization reaches at low fields, resulting the nonmagnetic Mg²⁺ substitution magnetic Fe²⁺ ions in the decrease of magnetic parameters (Curie temperature, saturation magnetization, coercive fields) due to the weakness of the super-exchange interaction of the magnetic moments. Obtained Mössbauer spectra prove the existence of the different states of Fe for nonequivalent sites in the case of the Mg-free compounds, which also allows us to determine the inversion degree of the spinel structure. Impedance spectroscopy measurements revealed an electrical response characterized by the bulk and grain boundary combined contributions. The samples prepared by RFS exhibit enhanced conductivity when compared to the conventionally sintered material, whereas activation energies for the charge carriers were found to be similar in all cases. The results presented here show the potential of RFS technique for the synthesis and consolidation of HEOs in a greener way and enhancement properties, which should translate well to other high entropy materials.

References

[1] J.W. Yeh, S.K. Chen, S.J. Lin, J.Y. Gan, T.S. Chin, T.T. Shun, C.H. Tsau, S.Y. Chang, Nanostructured highentropy alloys with multiple principal elements: novel alloy design concepts and outcomes, Advanced engineering materials 6(5) (2004) 299-303.

[2] C.M. Rost, E. Sachet, T. Borman, A. Moballegh, E.C. Dickey, D. Hou, J.L. Jones, S. Curtarolo, J.-P. Maria, Entropy-stabilized oxides, Nature Communications 6(1) (2015) 8485.

[3] S. Akrami, P. Edalati, M. Fuji, K. Edalati, High-entropy ceramics: Review of principles, production and applications, Materials Science and Engineering: R: Reports 146 (2021) 100644.

[4] B. Musicó, Q. Wright, T.Z. Ward, A. Grutter, E. Arenholz, D. Gilbert, D. Mandrus, V. Keppens, Tunable magnetic ordering through cation selection in entropic spinel oxides, Physical Review Materials 3(10) (2019) 104416.