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Exploring the high-temperature thermoelectric performance of Al-doped ZnO ceramics prepared by in-situ aluminothermic reactions

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Zinc oxide (ZnO) is a highly versatile and well-known material, widely used for its useful optoelectronic, catalytic, and photochemical properties.[1] In the search for alternative energy sources, ZnO-based ceramics emerge as promising materials for high-temperature thermoelectric (TE) applications,[2] capable of direct conversion of (waste) heat to electricity, thanks to the Seebeck effect. Owing to the specific wurtzite crystal structure of this large band-gap semiconductor, donor substitutions are often performed/employed, for tailoring the electrical properties and, in this respect, aluminium is probably the most well-known and often used cation[3] for improving the charge carrier concentration and/or mobility. Additionally, nanostructuring approaches are also considered and implemented for the independent control of the different TE coefficients involved in the charge and heat transport,[4] provided by their ability to promote the formation of specific microstructural features, capable of simultaneously enhancing the electrical conductivity and decreasing the lattice thermal conductivity. This work reports on the preliminary results of a combined charge carrier transport improvement / grain boundary tailoring scheme, involving the simultaneous increase in electron concentration/mobility (by doping with Al) and the engineering of defects between the highly packed grains characteristic for this material (by controlled aluminothermic reactions, providing in-situ redox conditions). The solubility of Al and the exothermic effects of aluminothermy are controlled by the careful choice of 3 different aluminium sources (for the targeted nominal composition $Zn_{0.995}Al_{0.005}O$) and a one stage sintering cycle, performed in air. The samples prepared from metallic micrometric and nanometric Al particles show the highest density values (around 96% of the theoretical value), compared with the conventional, reference $Zn_{0.995}Al_{0.005}O$ samples prepared from Al_2O_3 (around 92% of the theoretical value). The Wurtzite phase has been found in all cases as the single phase, and the relevant microstructural changes/features, including defects formation at the grain boundaries, have only been observed for the samples prepared from the 2 different metallic aluminium sources, provided by the aluminothermic reactions. The electrical performance results show a dramatic increase in electrical conductivity, for the samples prepared from micro- and nano-sized aluminium particles, leading to a maximum power factor value of around 700 $\mu W/K^2m$, at 900°C for the samples prepared from micrometric particles, being among the best values found in literature.

Keywords: Thermoelectric Al-doped ZnO ceramics; Controlled interactions; Grain boundary defects engineering; In-situ redox conditions; Aluminothermic reactions; Electrical performance

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