STRUCTURAL AND ELECTRICAL CHARACTERIZATION OF Sr- AND Al- DOPED APATITE TYPE LANTHANUM SILICATES PREPARED BY PECHINI METHOD.

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Abstract

Oxide ion conductors are important parts of solid oxide fuel cells (SOFCs) technology. A major challenge in SOFC development is to produce a solid electrolyte material with sufficient conductivity which could maintain acceptable low ohmic losses at intermediate temperatures. Apatite rare-earth silicates with a general formula $RE_{10-x}(TO_4)_6O_{3-\delta}$ (RE: rare earth, T: Si or Ge) have been attracting considerable attention as alternative solid electrolyte materials [1] because of their low activation energy and especially their excellent stability over a wide oxygen partial pressure range. Among many apatite rare-earth silicates, the lanthanum silicates, exhibit the highest values of oxide ion conductivity at intermediate temperatures ($La_{10}Si_6O_{27}$: $\sigma = 4x10^{-3}$ Scm^{-1} at 500^oC). It has been found that the observed conductivity is very sensitive to the doping regime and the cation/anion nonstoichiometry [2,3].

In this paper we report the preparation of $La_{9.83-x}Sr_xSi_{6-y}Al_yO_{26+\delta}$ (x=0, 0.45 and $0 \leq y \leq 0.50$) via Pechini method. To characterize the precursors, intermediate and final products, a number of analytical techniques (such as TG, XRD, FTIR and SEM) were applied. The total conductivity of the sintered samples was measured under air in the temperature range 175–700 °C using AC impedance spectroscopy. In order to reveal the correlation between stoichiometry, structure and properties of the compounds, extensive structural analysis was determined by means of Retvield structure refinement.

As it is concluded pure $La_{9.83}Si_6O_{26+\delta}$, $La_{9.38}Sr_{0.45}Si_6O_{26+\delta}$ and $La_{9.38}Sr_{0.45}Si_{5.70}Al_{0.30}O_{26+\delta}$ can be prepared after sintering at 1400 °C for 20h while $La_{9.38}Sr_{0.45}Si_{5.55}Al_{0.45}O_{26+\delta}$ and $La_{9.38}Sr_{0.45}Si_{5.50}Al_{0.50}O_{26+\delta}$ compounds contain traces (<8%) of La_2SiO_5 secondary phase. It seems that an increase in Al content leads in the production of compounds with altered extend of secondary phase. $La_{9.83-x}Sr_xSi_{6-y}Al_yO_{26+\delta}$ (x=0, 0.45 and $0 \leq y \leq 0.50$) compounds own hexagonal structure with $P6_3/m$ space group while the existence of interstitial oxygen was reported in all samples. The highest conductivity values (11 and $14x10^{-3}$ Scm^{-1}) were observed for $La_{9.83}Si_6O_{26+\delta}$ and $La_{9.38}Sr_{0.45}Si_{5.70}Al_{0.30}O_{26+\delta}$, respectively. The electrical conductivity of these compounds is inextricably connected to oxygen hexagonal channels surface of the apatite structure.

1. S. Nakayama, T. Kageyama, H. Aono, Y. Sadaoka, J Mater Chem, 5 (1995) 1801
2. A. Najib, J.E.H. Sansom, J.R. Tolchard, P.R. Slater, M.S. Islam, Dalton Trans, 19 (2004) 3106
3. J.E.H. Sansom, E. Kendrick, J.R. Tolchard, M.S. Islam, P.R. Slater, J. Solid State Electrochem, 10 (2006) 562