**Synthesis, Structural and EIS studies of La2-xAlxMo2O9 (x=0.2) oxygen ionic conductor**

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**Abstract**

La2Mo2O9 based ionic conductor with partial substitution of Al at La site, La2-xAlxMo2O9 (x=0.2), was synthesized by using conventional solid state reaction technique and characterised by XRD, DSC, Impedance analyser and FTIR. The RT XRD analysis reveals formation of pseudo monoclinic phase with impurity peaks pertaining to Al2O3 and La2Mo2O7. It has been observed that the conductivity is enhanced as compared to the parent compound by one order at 6000C which is a remarkable result. However, no suppression of phase transition was observed with Al doping.

**Keywords** solid electrolyte, phase transition, La2Mo2O9, doping, ionic conductivity.

1. **Introduction**

Recently, solid electrolyte has garnered huge attention due to its promising pertinence in solid state ionic devices [1-2]. La2Mo2O9 being the parent compound of LAMOX family is one of the most potential solid electrolytes originally introduced by Lacorre et.al. This compound has been under extensive research since its inception as ionic conductor in the year 2000. It has high anionic conductivity of ~10-2 S/cm at comparatively lower temperature (800oC) which is analogous to widely used solid electrolyte YSZ at 1000oC. It undergoes reversible β-α phase transition from high temperature cubic structure to low temperature monoclinic structure at around 580oC which deteriorates its conductivity and delimits its application at low temperature [3-5]. To enhance the conductivity, suppress the phase transition and to improve the mechanical stability, rigorous research is underway. Partial substitution being one of the most effective ways to overcome lacuna found in La2Mo2O9 [6]. Here we report, Al doping at La Site of La2Mo2O9 based compound, La2-xAlxMo2O9 (x=0.2), in search of desired result.

1. **Experimental**
   1. Synthesis

The specimen was prepared using conventional solid state reaction method.

Firstly, the ingredients were taken maintaining proper stoichiometric ratio. It was grinded under acetone environment in agate mortar for 12hrs initially, then, it was calcined at 5000C for 12hrs in muffle furnace. Subsequent, grinding and calcinations were performed at different temperatures. Lastly, it was pelletized and sintered at 9500C for 12hrs. The size of the pellet is ~11-13.

* 1. Characterisation

The powder sample was characterised using a RIGAKU ULTIMA IV X-ray diffractometer employing Cu-Ka radiation (λ=1.54059 Å) at room temperature in the 2θ range from 10o- 80o. The results thus obtained were further analysed by using “Fullprof” software. Average crystallite sizes were estimated using Scherrer’s formula

(1)

Where, L is the crystallite size in nm, k is the so-called “Scherrer constant” (k = 0.94), β is full width at half maxima (FWHM) of the diffraction peaks, Ө is obtained from corrected peak position 2Ө and λ represents the wavelength of Cu-Ka radiation ( λ = 1.154059Å).

Differential scanning calorimetric (DSC) measurement was performed by STA 449 F3–NETZSCH at the cooling rate of 50C/min in an argon atmosphere in the temperature range RT-6000C.

The Fourier transform infrared (FT-IR) spectrum of the prepared specimen was recorded in transmittance mode on Impact 410 (NICOLET, USA) at room temperature in the wave number region 400-4000 cm-1.

The gold paste was coated on both sides of the pellet for electrical measurements. HIOKI-LCR TESTER IM3536 was used for this study in temperature range RT -750oC. The results were analysed using EISSA software.

1. **Results and discussion**
   1. X-ray diffraction studies



**Fig.1** *XRD plot of La2Mo2O9 and La1.8AL0.2Mo2O9 compound.*

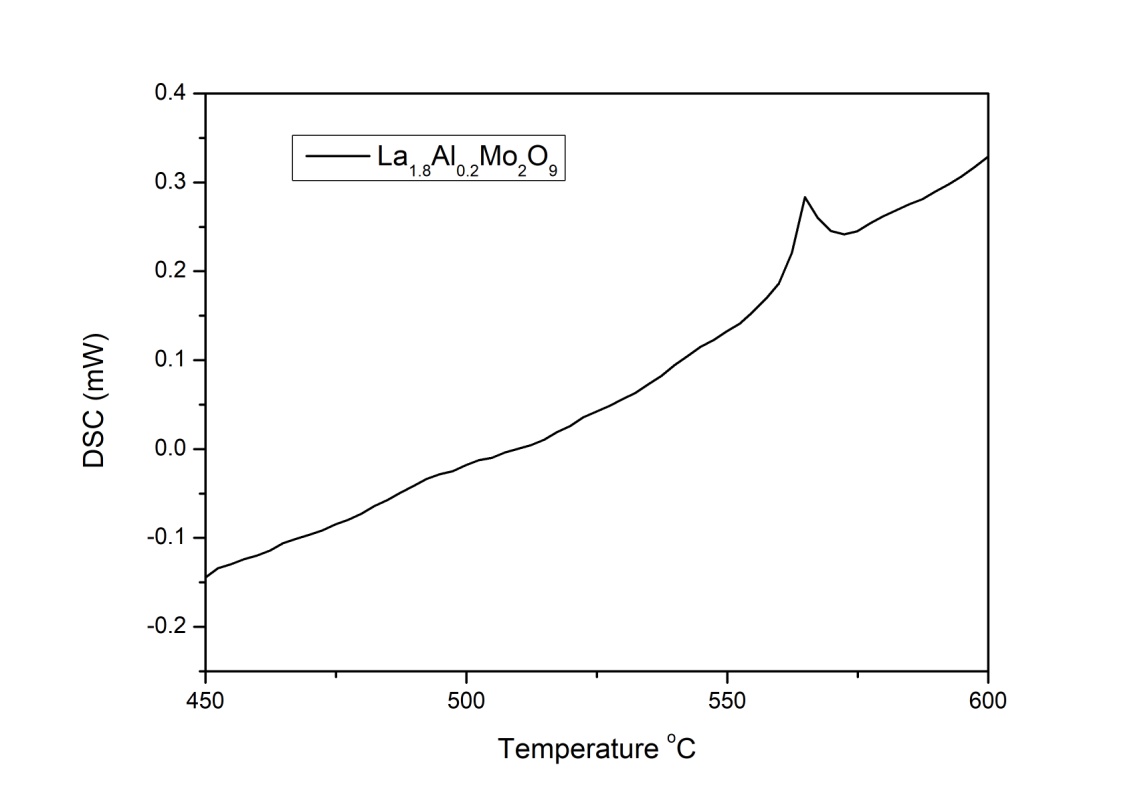
The XRD plots of La2Mo2O9 and La1.8AL0.2Mo2O9 compound are shown in Fig.1. The Fig. reveals pseudo monoclinic phase formation along with some impurity peaks in the doped compound when compared to the parent compound. Here impurity peaks corresponding to Al2O3 and La2MO2O7 (JCPDS card no. 861410 and 841234) are obtained at 2θ=13.450, 29.950 and 33.440. After analysing the parent and doped compound on Fullprof software and by using Scherrer’s formula the cell parameters found are shown in Table 1.

**Table 1**

*The cell parameter, cell volume and crystallite size of the pure and doped La2Mo2O9.*

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Sl.**  **no.** | **Specimen** | **a (Å)** | **b (Å)** | **c (Å)** | **Cell Volume (Å3)** | **Crystallite size (nm)** |
| 1. | La2Mo2O9 | 7.14404 | - | - | 364.61256 | 41.36 |
| 2. | La1.8AL0.2Mo2O9 | 7.14045 | - | - | 364.06317 | 39.81 |

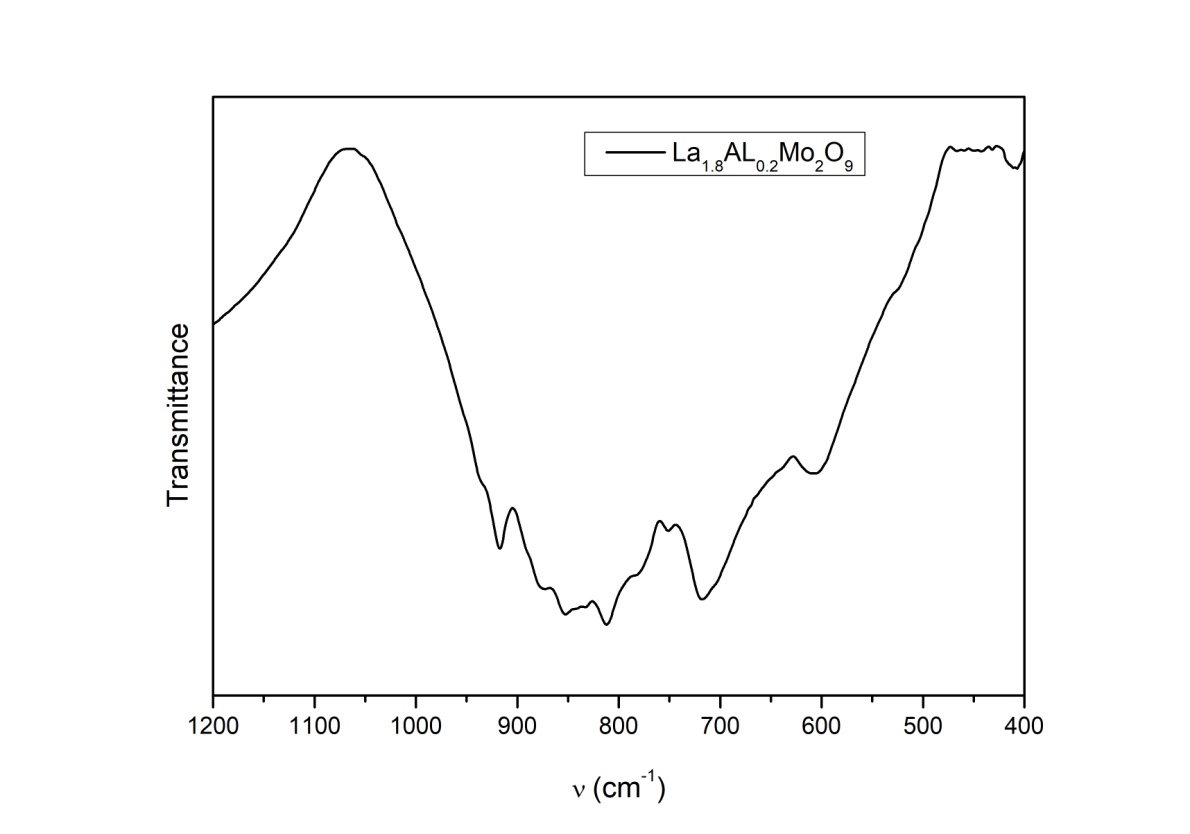
* 1. DSC study



**Fig. 2** *DSC curve of La1.8AL0.2Mo2O9 compound.*

The DSC curve of La1.8AL0.2Mo2O9 compound is shown in Fig.2. There exists exothermic peak in DSC curve at 564oC indicating that the doped compound doesn’t suppress the phase transition.

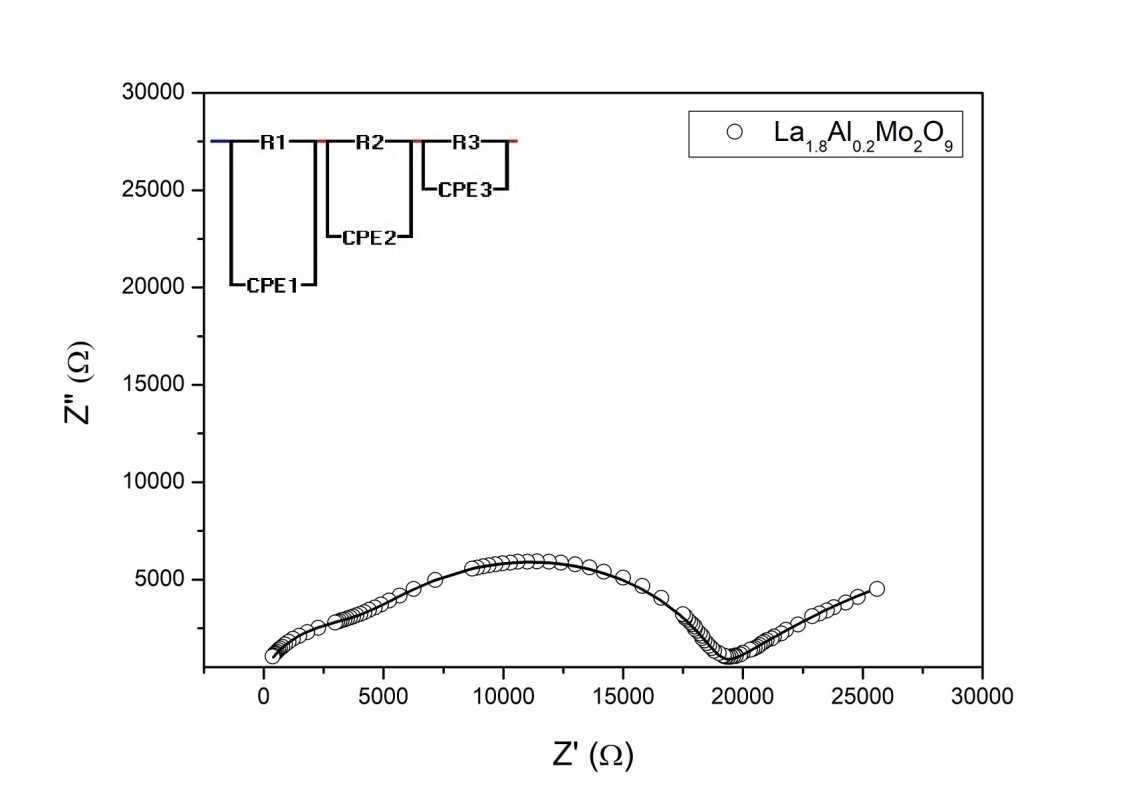
* 1. FT-IR



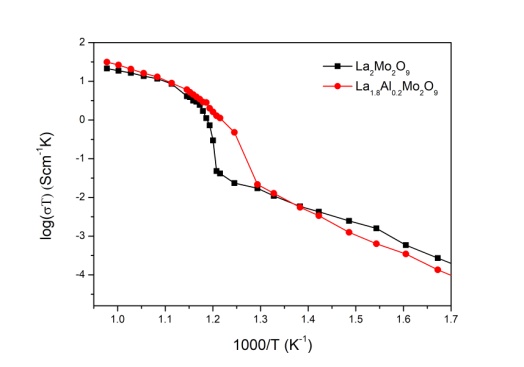
**Fig. 3** *FT-IR spectrum of La1.8Al0.2Mo2O9 compound.*

FT-IR spectrum for La1.8Al0.2Mo2O9 compound is shown in fig.3. The transmission band in the range ~950 cm-1 – ~600 cm-1 corresponds to Al-O and Mo-O stretching vibration which is in conformity with the result obtained in XRD [7-9]. Bands above 950 cm-1 corresponds to presence of ambient water [7-8].

* 1. EIS studies



**Fig.4** *The AC impedance spectrum of La1.8Al0.2Mo2O9 compounds at 430oC. The solid line is the fitted results obtained with equivalent circuit.*



**Fig.5** *1000/T versus log(σT) of La2Mo2O9 and La1.8Al0.2Mo2O9 compounds.*

* AC impedance Spectrum of La1.8Al0.2Mo2O9 at 430oC and Arrhenius plot of La2Mo2O9 and La1.8Al0.2Mo2O9 compounds are shown in Fig.4 and Fig.5, respectively.
* It is found that the conductivity of doped compound has been enhanced significantly from 500oC to 750oC whereas below 500oC the conductivity is low when compared with parent compound.
* At 600oC the conductivity of La1.8Al0.2Mo2O9 is found to be ~10-2 Scm-1 which is about one order higher at this intermediate temperature when compared with parent compound which is ~10-3 Scm-1.

1. **Conclusion**

The compound La2Mo2O9 and La1.8Al0.2Mo2O9 has been synthesized by conventional solid state reaction technique. Partial substitution of Al at La site of La2Mo2O9 doesn’t suppress the β-α phase transition. However, conductivity of La1.8Al0.2Mo2O9 is found to be higher than parent material in the temperature region (500oC < T < 750oC). At 600oC the conductivity is about one order higher as compare to the parent compound.

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