

STUDY OF $(\text{La}_{1-x}\text{Ce}_x)_3\text{Al}$ UNIT CELL VARIATION BY X-RAY DIFFRACTION

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ABSTRACT

The intermetallic compounds $(\text{La}_{1-x}\text{Ce}_x)_3\text{Al}$ have been studied using the X-ray diffraction technique. The data for La_3Al showed that the structure of this compound is hexagonal with $a = 7.25(1) \text{ \AA}$ and $c = 5.50(1) \text{ \AA}$. X-ray data for the entire range of Ce concentration (x) have also shown that there is a linear decrement of the unit cell volume as x increases.

I. INTRODUCTION

The compound $\text{Ce}_3\text{Al}^{1,2,3}$ can be obtained in two crystallographic structures which depend on the thermal treatment conditions: the $\alpha\text{-Ce}_3\text{Al}$ which is hexagonal with the Ni_3Sn type structure, space group $P6/mmc$ and the cubic phase $\beta\text{-Ce}_3\text{Al}$, space group $Pm3m$. Since the hexagonal phase has the most interesting properties only this phase is studied in this paper. In case of the compound La_3Al there is some controversy in the literature. Iandelli⁴ has reported a cubic phase with $a = 5.093 \text{ \AA}$ while Buchow⁵ has reported an hexagonal phase with $a = 7.195 \text{ \AA}$ and $c = 5.503 \text{ \AA}$ which has also been obtained by Garde et al⁶. However, Suzuki et al³ were able to obtain the $(\text{La}_{1-x}\text{Ce}_x)_3\text{Al}$ hexagonal phase only for the concentration range $1 > x > 0.33$.

The interesting competition between the RKKY interaction, that is characterized by $T_{\text{RKKY}} \approx \rho J^2$, and the Kondo effect where $T_K \approx \exp(-1/\rho J)$ in the system $(\text{La}_{1-x}\text{Ce}_x)_3\text{Al}$ is the main idea behind this work. This competition affects the physical properties of the compound as specific heat and electrical resistivity in a such way that a minimum in the resistivity at 20 K ($T_K \approx 3\text{K}$) and an anti-ferromagnetic ordering were observed at $T_N \approx 2.8 \text{ K}$ for $x = 1$ whereas a superconductor behavior with $T_C \approx 6\text{K}$ was observed for $x = 0$. The ρJ product where ρ is the density of states and J is the exchange parameter is a very important parameter in this competition since, it depends on the unit cell volume⁷. Therefore one can change it by applying an external mechanical pressure or equivalently by chemical substitution which is experimentally easier to make.

In this paper, the X-ray diffraction technique is used to determine and to monitor changes in the $(\text{La}_{1-x}\text{Ce}_x)_3\text{Al}$ unit cell volume which allow for the investigation of the ρJ product variation.

II. EXPERIMENTAL

The polycrystalline samples of Ce_3Al can be easily prepared by arc melting the stoichiometric quantities of 99.999% purity Al and 99.99% purity Ce, in argon atmosphere. The hexagonal phase is obtained after a 30 days thermal treatment at 500°C followed by 7 days at 200°C to avoid the cubic phase formation. It is usual to substitute Al for other elements (In, Sn, etc.) but in this case, the compounds are not isostructural (for instance, Ce_3In is cubic). For this reason, in the preparation of the series of compounds $(\text{La}_{1-x}\text{Ce}_x)_3\text{Al}$ an alternative procedure that substitutes Ce for a non magnetic similar element such as La or Y (99.99%) was used here.

The same extended (30 days) thermal treatment for the La rich samples has been adopted in order to reduce eutectic phases formation. The solid solutions $(\text{La}_{1-x}\text{Ce}_x)_3\text{Al}$ of polycrystalline samples were prepared by arc melting in argon atmosphere.

X-ray powder diffraction patterns ($\theta:2\theta$ scans) of the samples were carried out in a Philips PW1730 diffractometer using $\text{CuK}\alpha$ radiation and a graphite secondary monochromator. The samples were cut from the button and the larger face was polished to allow for the X-ray measurements. Metallography was also used to check the sample phases.

III. RESULTS AND DISCUSSION

The X-ray powder diffraction patterns obtained for the solid solutions $(\text{La}_{1-x}\text{Ce}_x)_3\text{Al}$ with $x = 0, 0.05, 0.10, 0.40, 0.50, 0.70, 0.85$ and 1.0 are shown in fig. 1. The analysis of these X-ray results indicates that the starting (La_3Al) as well as

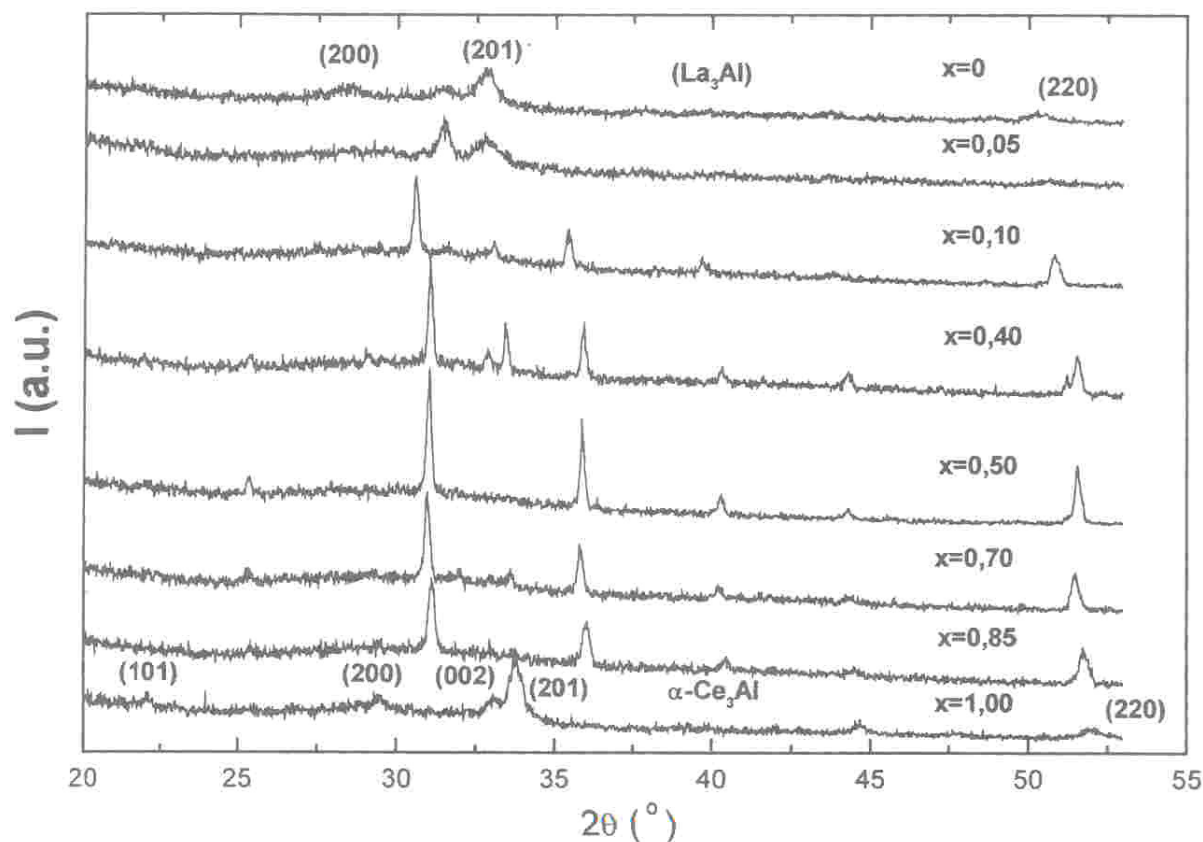


Figure 1- The X-ray powder diffraction patterns at 300 K ($\lambda = 1.5418 \text{ \AA}$) obtained for several x values in the solid solution $(\text{La}_{1-x}\text{Ce}_x)_3\text{Al}$.

the final ($\alpha\text{-Ce}_3\text{Al}$) materials are not presenting a crystallization as good as one could expect once, the peaks exhibit a broad width. Nevertheless, the intermediate phases show stronger and narrower peaks indicating better sample crystallization. These peaks change angular positions from one pattern to another due structural changes within the chemical substitution process. The $\alpha\text{-Ce}_3\text{Al}$ unit cell lattice parameters $a = 7.019 \text{ \AA}$ and $c = 5.426 \text{ \AA}$ obtained from the $x = 1$ pattern are in agreement with the literature reported values^{1,8}.

Regarding the X-ray results for La_3Al compound, an hexagonal structure with $a = 7.25(1) \text{ \AA}$ and $c = 5.50(1) \text{ \AA}$ was determined that agrees with the results found by Buschow (see table I). The metallographic analysis has shown that the sample presents the La_3Al as the major phase. However, a small amount of eutectics is also observed in the sample, as expected from the extended thermal treatment.

The $(\text{La}_{1-x}\text{Ce}_x)_3\text{Al}$ lattice parameters were determined from the sequence of X-ray powder diffraction patterns depicted in fig. 1. They are plotted as a function of the Ce

concentration (x) in fig. 2. It is clearly seen in this figure that both lattice parameters (a and c) reduce linearly on

Table I. Lattice parameters obtained for La_3Al .

ref. 5	$a = 7.195(5) \text{ \AA}; c = 5.503(5) \text{ \AA}$
ref. 6	$a = 7.192(20) \text{ \AA}; c = 5.528(13) \text{ \AA}$
this work	$a = 7.25(1) \text{ \AA}; c = 5.50(1) \text{ \AA}$

going from the La side to the Ce side. Therefore, since the unit cell volume has decreased from $V = 250 \text{ \AA}^3$ for La_3Al down to $V = 233 \text{ \AA}^3$ for Ce_3Al the pJ product increases with the Ce concentration and consequently T_K also increases¹⁰, as expected.

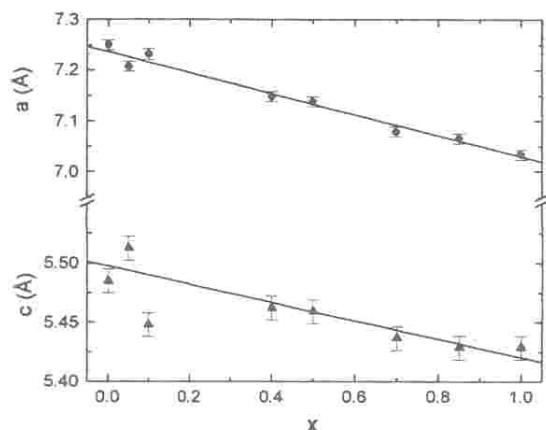


Figure 2-Lattice parameters a and c for the solid solution $(\text{La}_{1-x}\text{Ce}_x)_3\text{Al}$ and the linear fit.

IV. CONCLUSIONS

Using X-ray diffraction we were able to characterize the hexagonal structure for La_3Al samples. Lattice parameters obtained for the whole sequence of $(\text{La}_{1-x}\text{Ce}_x)_3\text{Al}$ solid solution have shown a linear contraction of the unit cell volume for higher Ce concentration values. This result leads to a linear behavior of the Kondo temperature as was observed in specific heat measurements⁹.

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