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Crystal growth of off-stoichiometric $Co_2Cr_{1-x}Al_{1+x}$ Heusler compounds: Avoiding the solid state miscibility gap

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Abstract

Co_2CrAl is predicted as a promising candidate for half-metallic ferromagnetism. Unfortunately, synthesis of the stoichiometric composition is a challenge as the samples prepared so far using various techniques suffer from formation of secondary phases and consequently anomalous physical properties. Off-stoichiometric compounds have been explored as a possible approach to avoid the secondary phase precipitation. The $Co_2Cr_{0.8}Al_{1.2}$, $Co_2Cr_{0.6}Al_{1.4}$ and $Co_2Cr_{0.4}Al_{1.6}$ compositions were synthesized using Floating Zone (FZ) technique as they melt incongruently. The secondary phase fraction was reduced as Cr was partially substituted by Al. FZ-grown $Co_2Cr_{0.4}Al_{1.6}$ was obtained as a single phase, signifying that the composition is out of the two-phase region. Furthermore, combining the data with an earlier work on annealing of spinodally decomposed samples, valuable insight into the extent of immiscibility was obtained.

Keywords: B2. Heusler compounds, A2. Floating Zone technique, A1. phase diagram, A1. off-stoichiometry, A1. spinodal decomposition

1. Introduction

Co-based Heusler compounds, in addition to exhibiting a wide range of properties, are also predicted to show half-metallic ferromagnetism in certain compositions, which is highly sought after in the field of spintronics [1]. One of more promising compounds as a potential half-metallic ferromagnet is Co_2CrAl [2–4]. However, the prepared samples suffer from element segregation [5–9] and, therefore, discrepancies in physical properties have been observed [5, 10–12]. The bulk samples prepared by conventional techniques such as arc-melting show dendritic microstructure as Co_2CrAl melts incongruently [5, 13]. Furthermore, samples grown by Floating Zone technique, which is one of the techniques of choice to grow incongruent melting compounds, suffer from a low temperature phase transformation via spinodal decomposition, which further led to secondary phase precipitates [13]. In a recent work by the authors, annealing experiments were done on the Floating Zone grown samples, but it was not possible to obtain a single phase, homogeneous sample [14].

Since the aforementioned results show that it is rather difficult to dissolve the secondary phase once it is formed, alternate approaches must be explored in order to avoid its precipitation from the beginning. A possible scenario is to access a single phase field in the phase diagram by varying the stoichiometry or through additional element substitution, such that the new composition is outside the miscibility gap. Therefore, the present work focuses on exploring off-stoichiometry with respect to Co_2CrAl Heusler composition in the

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