PREPARATION OF A SINGLE PHASE $R_3(Fe, Mo)_{29}$ (R= Ce, Nd, Sm, Gd, Tb, Dy AND Y) ①

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ABSTRACT The new type of rare earth iron intermetallic compounds $R_3(Fe, Mo)_{29}$ in the field of permanent magnetic materials have been paid more attention to, but it is very difficult to prepare a single phase 3∶29. The preparation of a single phase $R_3(Fe, Mo)_{29}$(R= Ce, Nd, Sm, Gd, Tb, Dy and Y) including the effects of the content of stabilizing element Mo, rare earth Y and annealing conditions on the formation of $R_3(Fe, Mo)_{29}$ compounds, has been discussed in detail. The results showed that the content of stabilizing element Mo in single phase 3∶29 compounds is equal to the corresponding Mo content x at maximum Curie temperatures of 2∶17 phase in nominal composition $R_3(Fe_{1-x}Mo_x)_{29}$ under the unsuitable annealing conditions. The single phase 3∶29 compounds can be prepared at this x value by verifying the annealing temperatures.

Key words 3∶29 compounds preparation a single phase

1 INTRODUCTION

In recent years, researchers in the field of permanent magnetic materials have paid more attention to the new type of rare earth iron intermetallic compounds $R_3(Fe, M)_{29}$(R= Ce, Nd, Sm, Gd, Tb, Dy and Y; M = Ti, V, Mn, Mo)[1-13]. Unfortunately, it is very difficult to prepare a single phase 3∶29 material, because of coexistence of 1∶12, 2∶17 or both of them[1-7]. However, it is very important to prepare a single phase materials for studying the intrinsic magnetic properties of 3∶29 compounds. In previous papers[8-10], we have reported the intrinsic magnetic properties of the single phase $R_3(Fe, Mo)_{29}$. The reports also indicated that both of the annealing temperature range and the content of stabilizing element Mo are very critical for preparing a single phase $R_3(Fe, Mo)_{29}$. If the content of stabilizing element Mo is right but the annealing temperature is not perfect, the single phase 3∶29 can not be obtained. For example, when an annealing temperature is much higher or lower than the right temperature for preparing a single phase 3∶29, the material consist of 2∶17 or 1∶12 phases; when a temperature is close to right temperature, they may consist of 3∶29 and 1∶12, or 3∶29 and 2∶17, or 3∶29, 2∶17 and 1∶12 phases[8-10]. In this case, we could get a single phase 3∶29 in materials by verifying annealing temperature carefully. The key problems are what the suitable content of stabilizing element Mo and annealing conditions are, and how to determine the suitable content of Mo and annealing conditions. In this paper, we present how to prepare a single phase $R_3(Fe, Mo)_{29}$ compounds (R= Ce, Nd, Sm, Gd, Tb, Dy and Y).

2 EXPERIMENTAL

The nominal compositions $R_3(Fe_{1-x}Mo_x)_{29}$ with R= Ce, Nd, Sm, Gd, Tb, Dy and Y were prepared by argon-arc melting at least with 99.9% pure constituent elements. The ingots were remelted at least 5 times for homogenization. The ingots were annealed at 1 200~ 1 473 K for 24 ~ 72 h under argon atmosphere, and then quenched in water.

X-ray diffraction with Cu-K_α radiation was used to identify the phases of the compounds and

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to determine the lattice parameters. The thermomagnetic analysis (TMA) was performed in a low field of about 0.04 T in the temperature range from 273 K to above the Curie temperature. The Curie temperatures T_C were determined from $\sigma^2 - T$ plots by extrapolating σ^2 to zero.

3 RESULTS AND DISCUSSION

The effect of content of stabilizing element Mo (nominal composition $R_3(Fe_{1-x}Mo_x)_{29}$) on the formation of phases under annealing condition of 1 203 K for 72 h was investigated in detail. X-ray diffraction patterns and thermomagnetic curves show that all compounds are composed of 1∶12 and 2∶17 phases, instead of 3∶29 phase. Further observation of the dependence of Mo content on Curie temperatures of 2∶17 phase in the nominal composition $R_{0.103}Fe_{1-x}Mo_x$ (R = Y, Tb and Dy) is shown in Fig. 1. It can be seen that the Curie temperatures increase with an increasing x (the content of Mo) initially. After a pass through maximum at certain x, the T_C decreases with a continuously increasing Mo concentration. The corresponding Mo content x at maximum Curie temperatures of 2∶17 phase are 0.039 for nominal composition $Ce_3(Fe_{1-x}Mo_x)_{29}$, 0.044 for $Nd_3(Fe_{1-x}Mo_x)_{29}$, 0.034 for $Sm_3(Fe_{1-x}Mo_x)_{29}$, 0.026 for $Gd_3(Fe_{1-x}Mo_x)_{29}$, 0.024 for $Tb_3(Fe_{1-x}Mo_x)_{29}$, 0.016 for $Dy_3(Fe_{1-x}Mo_x)_{29}$, and 0.031 for $Y_3(Fe_{1-x}Mo_x)_{29}$, respectively, which is very similar to those of the content of stabilizing element Ti in single phase $R_3(Fe_{1-x}Ti_x)_{29}$[11, 12]. Therefore, we deduce a conclusion, from above statements, that the corresponding Mo content x at maximum Curie temperatures of 2∶17 phase in nominal composition $R_3(Fe_{1-x}Mo_x)_{29}$ is equal to that of Mo content x for preparing the single phase $R_3(Fe_{1-x}Mo_x)_{29}$ compounds, and if the annealing conditions are suitable, the single phase $R_3(Fe, Mo)_{29}$ compounds can be synthesized under the nominal compositions mentioned above, otherwise it can not be synthesized.

In order to confirm this conclusion, the effect of different annealing temperatures on the formation of phase(s) at nominal composition $Y_3(Fe_{0.969}Mo_{0.031})_{29}$ has been studied in detail. The corresponding thermomagnetic curves at different annealing temperatures are shown in Fig. 2. Fig. 2(a) is as cast, Fig. 2(e) is annealed at 1 353 K for 72 h and then cooled in the air, and the others are annealed at certain temperature for 72 h, and then water quenched. Fortunately, it can be seen that the single phase yttrium 3∶29 compound has been successfully prepared at 1 353 K for 72 h and then water quenching (Fig. 2(d)). The corresponding X-ray diffraction pattern is shown in Fig. 3(g). If the samples annealed at 1 353 K for 72 h and then cooled in air, the single phase 3∶29 can not be synthesized and it contained impurity phase 2∶17 (Fig. 2(e)). It also can be found that if the annealing temperatures higher than 1 353 K, the impurity phase 1∶12 appeared, and the phase 3∶29 disappeared with the annealing temperature up to 1 403 K and the sample consisted of phases 2∶17 and 1∶12. If the annealing temperature was lower than 1 353 K, 1 333 K, for example, the impurity phase 2∶17 appeared. The phase 3∶29 also disappeared when the annealing temperature was lower than 1 293 K and the sample consisted of phases 2∶17 and 1∶12.

The annealing temperatures of preparing single phase 3∶29 using the nominal compositions discovered above by this means have been found. In summary, the corresponding X-ray diffraction patterns of $R_3(Fe, Mo)_{29}$ compounds are shown in Fig. 3, the best compositions and annealing conditions for preparing single phase

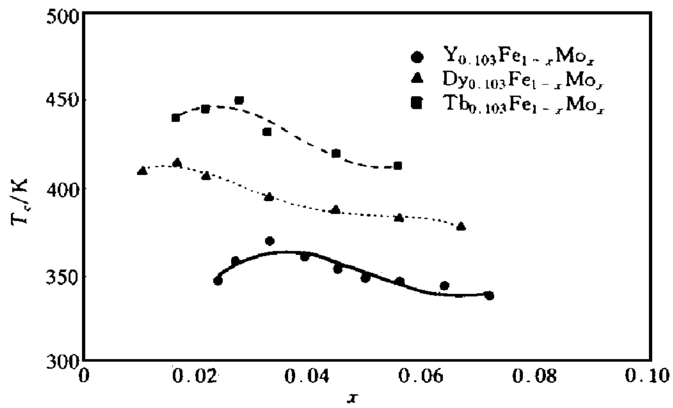

Fig. 1 The dependence of Mo content on the Curie temperatures of the phase 2∶17

$R_3(Fe, Mo)_{29}$(R= Ce, Nd, Sm, Gd, Tb, Dy, and Y) compounds are listed in Table 1.

Until now, a new method on how to determine the Mo content in a single phase 3∶29 has been found, but the content of the rare earth Y is also very important in preparing a single phase 3∶29 material. Fig. 4 illustrates the effects of the contents of rare earth Y and stabilizing element Mo on the formation of $Y_3(Fe, Mo)_{29}$ compound and also gives the details about the relationship between 3∶29 and 2∶17 or 1∶12 under the optimum annealing condition (1 353 K for 72 h). It can be found the zone of single phase 3∶29 is very narrow, and the conditions of obtaining a single phase 3∶29 is that if the compound consists of 3∶29 and α-Fe, the contents of rare earth should be increased; if the compound consists of 3∶29, 2∶17 and α-Fe, the content of stabilizing element Mo should be increased; if the compound consists of 3∶29, 1∶12 phases, the content of stabilizing element Mo should be decreased; and if the rare earth appeared in the compound, the content of rare earth should be decreased. The zone of single phase 3∶29 is very

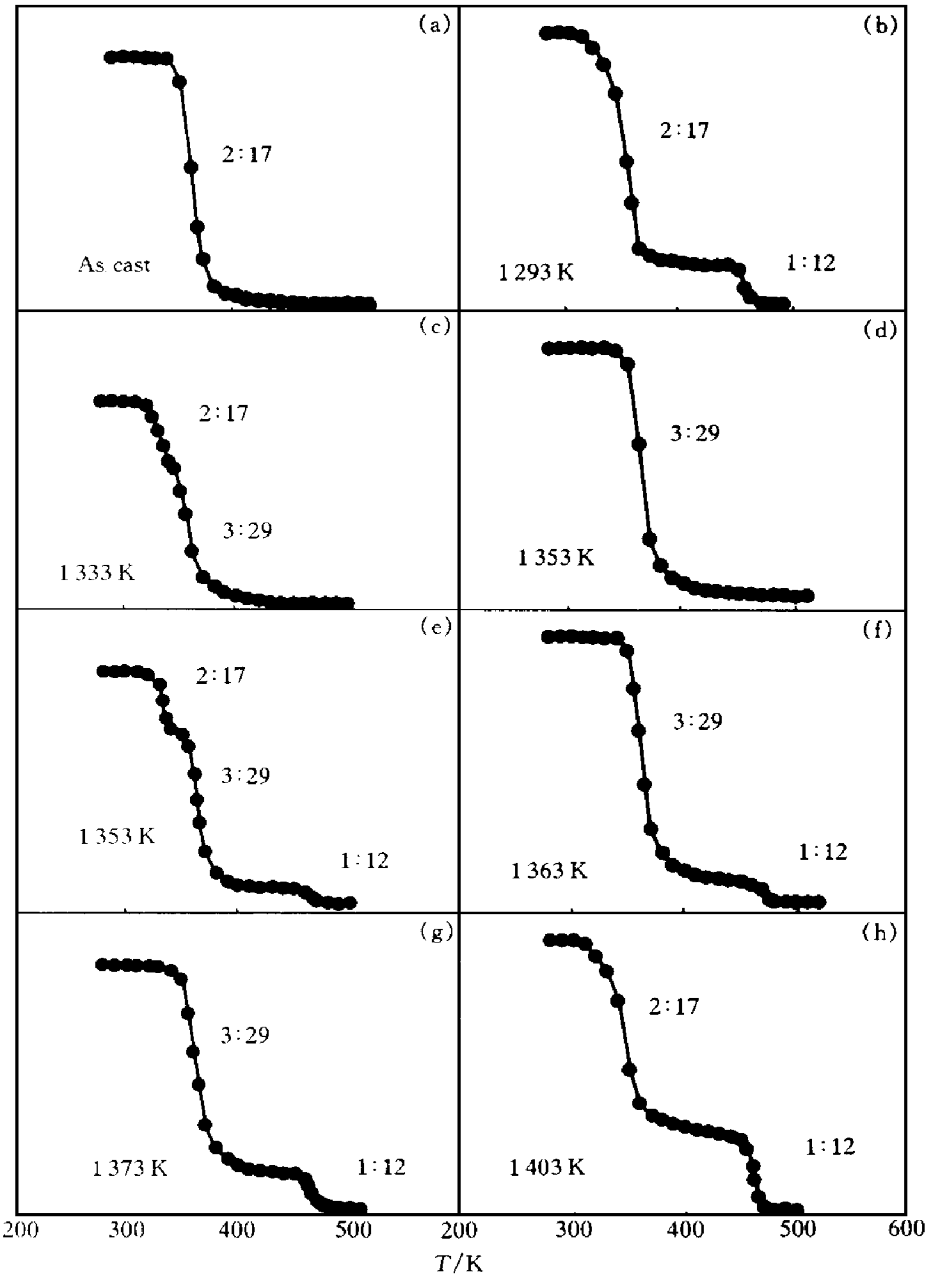

Fig. 2 The σ-T plots of $Y_3(Fe_{0.969}Mo_{0.031})_{29}$ compound annealed at different temperatures

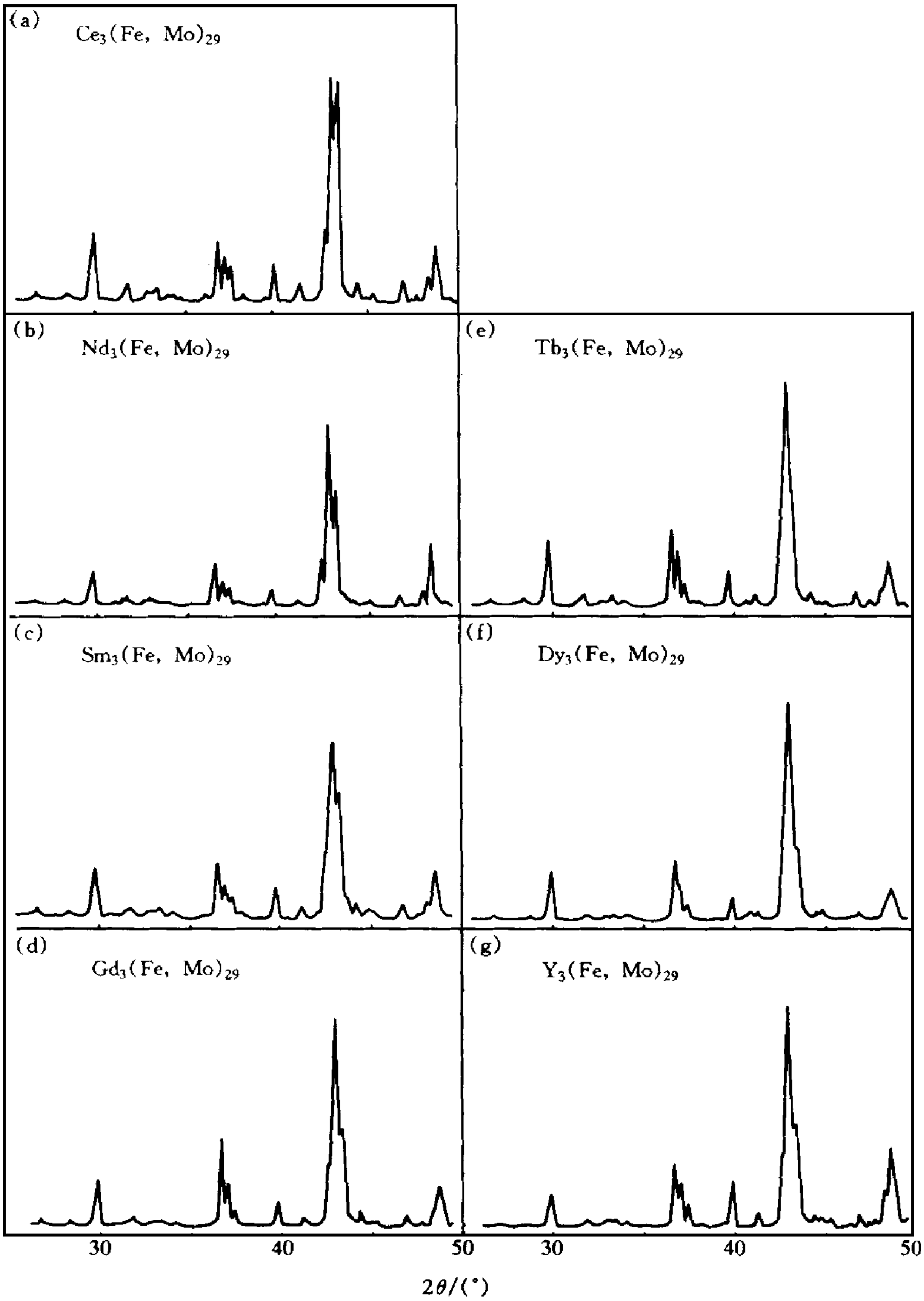

Fig. 3 The X-ray diffraction patterns of $R_3(Fe, Mo)_{29}$ compounds (R= Ce, Nd, Sm, Gd, Tb, Dy and Y)

narrow, and the conditions of obtaining a single phase 3∶29 is that if the compound consists of 3∶29 and α-Fe, the contents of rare earth should be increased; if the compound consists of 3∶29, 2∶17 and α-Fe, the content of stabilizing element Mo should be increased; if the compound consists of 3∶29, 1∶12 phases, the content of stabilizing element Mo should be decreased; and if the rare earth appeared in the compound, the content of rare earth should be decreased.

4 CONCLUSIONS

(1) The content of stabilizing element Mo in single phase 3∶29 compounds is equal to the corresponding Mo content x at maximum Curie temperatures of 2∶17 phase in nominal compositions $R_3(Fe_{1-x}Mo_x)_{29}$ under the unsuitable annealing conditions. The single phase 3∶29 com-

Table 1 The best compositions and annealing conditions of preparing single phase $R_3(Fe, Mo)_{29}$ (R= Ce, Nd, Sm, Gd, Tb, Dy and Y)

Compositions		Annealing Conditions	
Compounds	x	T_a^* / K	t_a^{**} / h
$Ce_3(Fe_{1-x}Mo_x)_{29}$	0.039	1313	24
$Nd_3(Fe_{1-x}Mo_x)_{29}$	0.044	1453	24
$Sm_3(Fe_{1-x}Mo_x)_{29}$	0.034	1453	24
$Gd_3(Fe_{1-x}Mo_x)_{29}$	0.026	1453	24
$Tb_3(Fe_{1-x}Mo_x)_{29}$	0.024	1323	72
$Dy_3(Fe_{1-x}Fe_x)_{29}$	0.016	1253	72
$Y_3(Fe_{1-x}Mo_x)_{29}$	0.031	1373	72

* : Annealing temperatures; * * : Annealing time

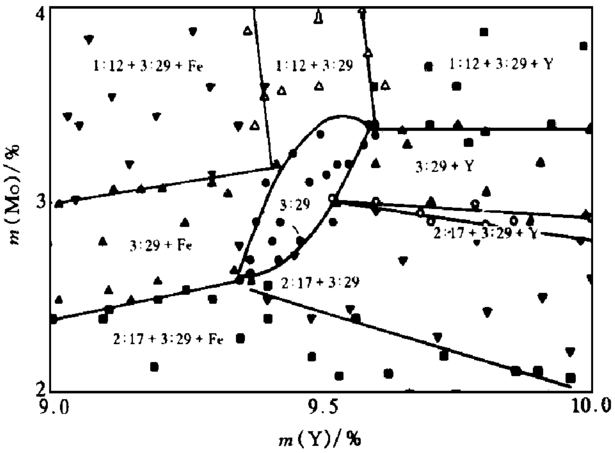

Fig. 4 Effects of contents of the element Y and Mo on the formation of 3∶29 phase

pounds can be prepared at this x value by verifying the annealing temperatures.

(2) The zone of single phase 3∶29 is very narrow, and the conditions of obtaining a single phase 3∶29 is that if the compound consists of 3∶29 and α-Fe, the contents of rare earth should be increased; if the compound consists of 3∶29, 2∶17 and α-Fe, the content of stabilizing element Mo should be increased; if the compound consists of 3∶29, 1∶12 phases, the content of stabilizing element Mo should be decreased; and if the rare earth appeared in the compound, the content of rare earth should be decreased.

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