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# Kinetics of hydrogen-induced direct phase transformation in $Y_2Fe_{17}$ hard magnetic alloy

E.V. Dodonova, S.B. Rybalka†

† sbrybalka@yandex.ru

Donetsk National Technical University, Artema Street 58, 283000 Donetsk

# Кинетика индуцированного водородом прямого фазового превращения в магнитотвердом сплаве $Y_2Fe_{17}$

Додонова Е.В., Рыбалка С.Б.

Донецкий национальный технический университет, ул. Артема 58, 283000 Донецк

The kinetics of hydrogen-induced direct phase transformation, i.e. decomposition process of $Y_2Fe_{17}$ initial alloy at interaction with hydrogen on $YH_2$ hydride phase and α-Fe phase of iron, in $Y_2Fe_{17}$ hard magnetic alloy has been studied. It has been established that, as the temperature increase from 610 to 750˚C, a direct phase transformation significantly accelerates. The activation energy of phase transformation process has been determined from kinetic data (163÷242 kJ/mol) that it is correspond to the values of activation energy of the iron atoms diffusion in R–T type alloys. It has been shown that the kinetics of hydrogen-induced direct phase transformation in $Y_2Fe_{17}$ hard magnetic alloy is controlled by the Fe atoms diffusion to growing new α-Fe phase centers.

**Keywords:** intermetallics; magnetic alloys; gas–solid reactions

Исследована кинетика индуцированного водородом прямого фазового превращения, т.е. процесс распада исходного сплава $Y_2Fe_{17}$ при взаимодействия с водородом на гидридную фазу $YH_2$ и α-Fe фазу железа, в магнитотвердом сплаве $Y_2Fe_{17}$. Установлено, что при увеличении температуры от 610 до 750˚C, прямое фазовое превращение значительно ускоряется. Определена энергия активации процесса фазового превращения из кинетических данных (163÷242 кДж/моль) что соответствует значениям энергии активации диффузии атомов железа в сплавах типа R–T. Показано, что кинетика индуцированного водородом прямого фазового превращения в магнитотвердом сплаве $Y_2Fe_{17}$ контролируется диффузией атомов Fe к растущим центрам новой α-Fe фазы.

**Ключевые слова:** интерметаллиды; магнитные сплавы; газ–твердое тело реакции

## 1. Introduction

At present the intermetallic compounds of $R_2M_{17}$ (R=Sm, Y, Dy, Ho, Gd) type have attracted much attention because of their interesting magnetic properties [1,2]. In particularly, $R_2M_{17}$ compounds demonstrate very interesting magnetic phenomenon during their interaction with interstitial atoms (H, N, C, B) [3-5].

In particular, the new perspective technology well known as a HDDR-process (Hydrogenation-Decomposition-Desorption-Recombination) in $R_2M_{17}$ type alloys ($Sm_2Fe_{17}$, $Sm_2Co_{17}$ etc.) alloys for permanent magnets allows improve their structure and magnetic properties by hydrogen-induced reversible phase transformations [6].

The most significant aspect of the HDDR process is that there is a dramatic change in the microstructure alloy from an initial grain size of typically ~150μm to a very fine, uniform grain size of about 0.1-0.3μm [6]. On magnetisation, the HDDR nanocrystalline powder exhibits an appreciable coercivity and this means that, in its simplest net shape form, the powder can be mixed with a thermosetting resin to produce an isotropic and anisotropic permanent magnet by compression moulding

In particular, at HDDR-treatment the $Y_2Fe_{17}$ alloy undergoes the direct hydrogen-induced phase transformation at temperatures above 500˚C with decomposition of initial alloy on hydride $YH_2$ phase and α-phase of Fe that can be described by the following phase scheme:

$$Y_2Fe_{17} + H_2 \rightarrow YH_2 + \alpha\text{-}Fe \quad (1)$$

Then, the reverse phase transformation takes place during hydrogen evacuation at higher temperatures with recombination decomposed phases into initial $Y_2Fe_{17}$ matrix phase and can be described by the following phase reaction:

$$YH_2 + \alpha\text{-}Fe \rightarrow Y_2Fe_{17} + 2H_2 \rightarrow Y_2Fe_{17} \quad (2)$$

And finally, after the completion of recombination stage the treated alloy as a rule consist of the nanocrystalline phase of $Y_2Fe_{17}$.

It is obviously that the clear outstanding of kinetic features of the above hydrogen-induced phase transformations will allow in follows to control microstructure and magnetic properties of this alloy. For instance, investigation of kinetics of such type direct and reverse hydrogen-induced transformations carried out earlier in $Sm_2Fe_{17}$ and $Nd_2Fe_{14}B$ type alloys [7-8], give us possibility to establish the basic features of these transformations: temperature intervals of development of transformations in $Nd_2Fe_{14}B$ type alloys; to establish diffusive-controlled character of transformations; to construct for the first time T-T-T diagrams (Temperature-Time-Transformation or isothermal kinetics diagrams) for transformations; to establish influence of kinetics parameters on microstructure features of alloys [9-10].

For $Y_2Fe_{17}$ alloy above-mentioned kinetic peculiarities have been not known yet until present and therefore, the main goal of present work was to investigate the features of kinetics of the hydrogen-induced direct phase transformation (1) in $Y_2Fe_{17}$ alloy at temperatures range of 610-750˚C in hydrogen pressure of 0.1 MPa.

## 2. Materials and methods

Samples of $Y_2Fe_{17}$ alloy were prepared by arc melting in an argon atmosphere of high purity. The raw materials of Y, Fe were at least 99.9% pure. The arcmelted ingots were wrapped in molybdenum foil, sealed in a steel tube, annealed at 1273 K for 24 h in a highly-purified argon atmosphere and then quenched to room temperature, resulting in a single-phase compound of the 2:17-type structure.

All kinetic experiments by investigations of kinetics of direct hydrogen-induced phase transformation has been carried out on special hydrogen-vacuum equipment using a special magnetometric Sadikov's method [7]. Samples of $Y_2Fe_{17}$ alloy as powders (~50 μm, ~1.2 g) were placed into a reaction chamber which was evacuated up to ~0.1 Pa vacuum. Then alloy was heated in vacuum to temperatures within 610-750˚C with continuous evacuation. After establishing isothermal conditions, the reaction chamber was filled with hydrogen up to 0.1 MPa pressure. The evolution of hydrogen-induced direct phase transformation in $Y_2Fe_{17}$ alloy was monitored continuously by meaning the increase of the amount of α-Fe ferromagnetic phase in the sample. Further, in accordance with obtained data the isothermal kinetic curves of phase transformation have been plotted. X-ray analysis was performed using of DRON-3 difractometer in Fe-Kα radiation.

## 3. Results and discussion

Thus, we shall remind, that the heating of an alloy $Y_2Fe_{17}$ in hydrogen atmosphere results in development of direct phase transformation leading to alloy decomposition with formation of a hydride $YH_2$ phase and phase α-Fe (see the equation (1)) which was established by X-ray difraction experiment.

The results of research of kinetics of hydrogen-induced direct phase transformation in $Y_2Fe_{17}$ alloy are generalized in Fig. 1 in form of kinetic curves.

The results of studying a direct hydrogen-induced phase transformation in $Y_2Fe_{17}$ alloy are generalised in Figure 1. As follows from Fig. 1, at temperatures 610, 630, 640, 650, 670 and 710˚C hydrogen-induced direct phase transformation not has been completed for transformation time of $1.44\times10^4$ s. At lower temperatures the direct phase transformation is not finished completely for experimental time with reaching at 610˚C only 20% of completeness, then at 630˚C – 27%, 640˚C – 43%, 650˚C – 46%, 670˚C – 66%, 690˚C – 75% and at 710˚C – 82%. Thus, at increase of temperature up to 730 and 750˚C transformation has been completed for $1.08\times10^4$ s and $0.912\times10^4$ s, respectively.

As can be seen from Fig. 1, with increasing of transformation temperature in narrow interval (140˚C) from 610˚C up to 750˚C the direct phase transformation lead to very strong acceleration of transformation in some order of magnitude.

Because of this, it is necessary also to note, that at all temperatures there is a noticeable incubation period of a phase transformation (from $0.33\times10^4$ s at 640°C to $0.015\times10^4$ s at 760°C).

As can be seen in Figures 1, the shape of the kinetic curves with a gradual slowdown of the transformation rate with decrease temperature and also dependence of the incubation period on the temperature let us suggest [11] that phase transformations develop by the mechanism of nucleation and growth. It is necessary to note that the results obtained in this work are similar to the our earlier data [7,8], where the kinetics of hydrogen-induced phase transformations in $Nd_2Fe_{14}B$ and $Sm_2Fe_{17}$ alloys was studied; direct and reverse phase transformations in these alloys greatly accelerates as the temperature increases within 610°C–750°C, too.

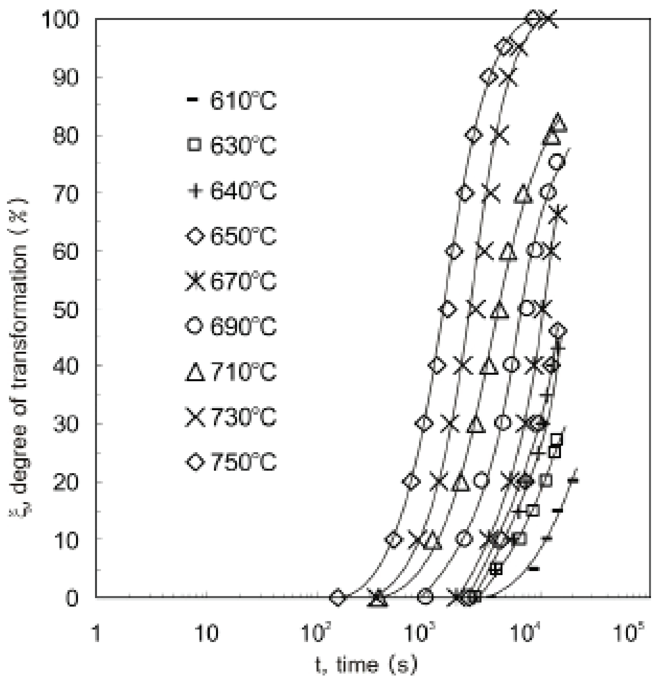


**Fig. 1.** Kinetic curves of direct hydrogen-induced phase transformation in $Y_2Fe_{17}$ alloy at temperatures from 610 to 750˚C and under hydrogen pressure of 0.1 MPa.

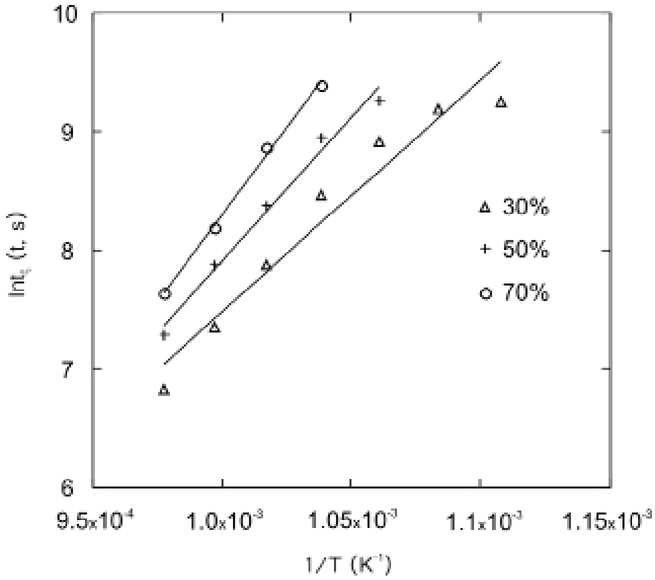


**Fig.2.** Dependence lntξ on 1/T for direct phase transformations in Y2Fe17 alloy for 30, 50 and 70 % of degrees of phase transformation.

Further, as well known from classic kinetic theory of phase transformations in condensed state [11], in particular in accordance with Becker-Döering model of nucleation kinetics [12-13] if plots dependence ln$t_ξ$ on $1/T$, where $t_ξ$ is the transformation time, which it is necessary for reaching of some degree of transformation ξ and $T$ is the transformation temperature, we can determine an effective energy of phase transformation process. For this goal experimental data from Fig. 1 were re-plotted in co-ordinates ln$t_ξ$ versus $1/T$ which are shown in Fig. 2. The slopes of the straight lines give us the values of the effective activation energies for hydrogen induced direct phase transformation in $Y_2Fe_{17}$ alloy.

The obtained values of an activation energy determined for some degrees of transformation varying from 163 up to 242 kJ/mol. Thus, above obtained values of an activation energy correspond to the values of an activation energy of the iron atoms diffusion in R–T alloys (where R is a rare-earth metal, T – a transition metal) ~250 kJ/mol [14] and self-diffusion of Fe atoms in α-Fe phase of iron [15,16], whereas activation energy for hydrogen atoms diffusion in R–T alloys is ~45 kJ/mol [14].

Thus, can be believed that the above investigated direct hydrogen-induced phase transformations in $Y_2Fe_{17}$ alloy are controllable by diffusion of iron atoms to growing α-Fe phase new centers.

## 4. Conclusions

It is established experimentally that with increase of temperature from 610°C up to 750˚C the evolution process of direct hydrogen-induced phase transformations in $Y_2Fe_{17}$ alloy accelerates too. In addition, it is established that at all transformation temperatures there is noticeable incubation period of transformation (from 0.33×10$^4$ s at 640°C to 0.015×10$^4$ s at 760°C).

On the base of the analysis carried out on the base of classic kinetic theory of phase transformations in condensed state has been established that hydrogen-induced phase transformations in $Y_2Fe_{17}$ alloy occur by the mechanism of nucleation and growth.

Activation energy of transformation process determined from kinetic data varying from 163 up to 242 kJ/mol that it is correspond to the values of activation energy of the iron atoms diffusion in R–T type alloys.

It is shown that phase transformations kinetics in the investigated interval of temperatures is controllable by diffusion of iron atoms to growing new α-Fe phase centers.

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