FORMATION OF INTERMETALLLICS DURING REACTIVE SINTERING PRODUCTION OF Fe-AL ALLOYS

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Abstract

Fe-Al alloys have been known for more than 100 years. Their exceptional oxidation and sulphidation resistance makes them promising for many industrial applications. However, these materials suffer from poor room-temperature ductility and problems during a production by conventional melting metallurgy. Therefore, reactive sintering of iron and aluminium compressed powder mixtures have been proposed as the alternative technology of their production. Unfortunately, this production technology leads to highly porous products. There are many theories why the porosity is so high and all of them deal with the intermetallic phases’ formation mechanisms. There are at least three different phases’ formation sequences during heating described in the literature. In this work, the in situ high energy X-ray diffraction analysis was carried out during the reactive sintering process and the mechanism of the formation of intermetallics in FeAl25 powder mixture was described during isothermal annealing at 800°C as well as during a slow continuous heating to this temperature. In both cases, no phases are formed prior the melting of aluminium particles. After that, Fe2Al5 and FeAl phases arise. The effect of the intermetallics’ formation on the porosity of Fe-Al alloys is discussed.

Klíčová slova:
iron aluminide, reactive sintering, XRD

1. INTRODUCTION

Materials based on FeAl and Fe3Al phases are promising materials for high-temperature applications. However, low room-temperature ductility and problematic production of these materials by conventional melting processes limit their applicability. There were many attempts to overcome these limitations [1-5], while one of them is the powder metallurgy using reactive sintering. In this process, intermetallics are produced by thermally activated in situ reactions during sintering of compressed elemental powder mixtures. The big advantage of this technology is the fact that the reactions leading to the formation of Fe-Al intermetallics initiate at significantly lower temperature than the melting temperature of iron and Fe-Al phases. However, in the case of iron-aluminium alloys extremely high porosity is achieved, especially when pressureless reactive sintering is applied (over 25 vol. %) [6]. There are many theories aiming to explain this behaviour. Most of them state that iron aluminides (Fe2Al5 or FeAl3) start to form below the melting temperature of aluminium [6-8] thus producing higher porosity than solid-liquid reaction. Porosity is also explained by volume changes in the structure resulting from lattice and density differences between the
temporary and final phases [6]. The other presented explanation of the enormous porosity is the Kirkendall phenomenon. It can be observed when the rate of interdiffusion of reacting metals strongly differs. In that case, unidirectional diffusion of one metal is compensated by vacancies diffusion. Coalescence of vacancies produces pores.

However, all of the above described results and theories are based on the ex-post study after the reactive sintering only. In some cases, the XRD study of the product is extended by the thermal analysis showing thermal effects of the reactions [8]. In this work, in situ XRD was applied to investigate the reactive sintering progress in Fe-Al powder mixtures.

2. EXPERIMENTAL

Green bodies of FeAl25 (in wt. %) were prepared by blending of commercial fine iron powder (<10 μm, p.a. purity) and coarse irregular aluminium powder (200-600 μm, 99.999 wt. % purity) and by pressing at the room temperature by a pressure of 320 MPa. In in-situ XRD analysis of the phases’ formation during reactive sintering of FeAl25 green body was carried out at Hasylab, DESY Hamburg using the X-ray beam with 100 keV energy on DORIS III ring, BW5 experimental stage. Analyses were conducted in two modes - slow continuous heating and isothermal reactive sintering.

In continuous heating regime, samples were heated from the laboratory temperature to 900°C with the heating rate of 10 K.min⁻¹ during DTA in our previous work [9]. The continuous heating regime consisted of placing the sample directly to the furnace preheated to 800°C and reactive sintering for 300 s. In both modes, XRD patterns were acquired each 3 s. Diffraction patterns obtained as 2D images were integrated to conventional intensity-angle dependence by Fit2D software and solved using an PANalytical HighScore Plus Software with PDF2 database. The microstructure of sintered samples was observed using light microscope (Olympus PME3) and TESCAN VEGA 3 LMU scanning electron microscope equipped with OXFORD Instruments INCA 350 EDS analyser.

3. RESULTS AND DISCUSSION

In our previous work [9], DTA analysis with the heating rate of 10 K.min⁻¹ revealed only one thermal effect – an exothermic peak at 660°C. In order to explain this behaviour, in situ XRD experiment was carried out under the same heating rate (10 K.min⁻¹). This experiment revealed that the first change is the aluminium melting, seen as gradual widening and disappearing of the aluminium diffraction lines. It indicates that any intermetallic phase is not formed below the melting temperature of aluminium. Immediately after the melting of aluminium, Fe₂Al₅, FeAl₂, FeAl and Fe₃Al phases arise, see XRD patterns in Fig.1. When the temperature increases, the intensities of FeAl diffraction lines increase, while the Fe₃Al phase signal reduces and it almost completely disappears until 900°C is reached. The Fe₂Al₅, FeAl₂ and FeAl phases retain in the reaction mixture up to 900°C.
The XRD experiment presented above aimed to model the conditions of previous DTA results. However, our recommended reactive sintering route for aluminides and silicides requires extremely high heating rates [10,11]. Such conditions can be easily achieved by placing the sample into a furnace preheated to the reactive sintering temperature. This experimental setup was used in isothermal in-situ experiment, aiming to simulate real reactive sintering as closely as possible. Temperature of 800°C was applied in this experiment. The results are similar to continuous heating experiments. At first, aluminium particles melt, directly followed by the formation of Fe$_5$Al$_6$, FeAl$_2$ and FeAl phases. The only difference from continuous heating lies in the fact that Fe$_3$Al phase was not identified.
To describe the effect of the particle size, the same experiment was carried out using coarser iron particles (< 250 µm, purity 99.5 wt. %), while the fraction of aluminium powder remained unchanged. In this case, similar behaviour was observed, i.e. aluminium melts and after that intermetallics (Fe$_2$Al$_5$, FeAl$_2$ and FeAl) form. In addition to these phases, γ-iron was determined when the formation of intermetallics started. After the formation of intermetallics was completed, the γ-iron slowly changed to α-iron.

All samples after reactive sintering are composed of Fe$_2$Al$_5$, FeAl and small fraction of FeAl$_2$ phase, see Fig.3. FeAl phase surrounds the Fe$_2$Al$_5$ areas, while FeAl$_2$ lies inside them. In addition to these intermetallics, unreacted iron was identified. EDS analyses showed that the residual iron contains only very small amount of aluminium.
CONCLUSION

In this work, the formation of intermetallics during reactive sintering of Fe–Al powder mixtures was investigated. It was found that formation of intermetallic compounds in the given system was initiated by melting of aluminium. The Fe2Al5 phase is formed preferentially, immediately followed by the formation of FeAl phase. FeAl2 phase forms in small amount, probably as a result of a limited diffusion. During slow heating, temporary formation of Fe3Al phase was also observed at temperatures of approx. 660-800°C. High porosity is probably connected with the formation of intermetallics immediately after melting of aluminium, which causes that the melt is not able to fill the pores, and with extreme Kirkendall effect, producing pores in spaces after aluminium particles.

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REFERENCES


