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# A two-stage approach of manufacturing FeAl40 iron aluminides by self-propagating synthesis and pressureless sintering

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## Abstract:

A two-stage sintering process was successfully used to sinter FeAl to densification levels of just above 95% at a temperature of 1300 °C. In the first stage, mixed iron and aluminium powders were synthesised at 750°C via Self-Propagating High-temperature Synthesis (SHS) to form brittle and porous Fe<sub>2</sub>Al<sub>5</sub>. Then the pellets were crushed and milled to various sizes and mixed with iron powders in the nominal composition of FeAl40 and pressurelessly sintered at a higher temperature to obtain a higher densification by taking advantage of the less violent exothermic reaction of Fe<sub>2</sub>Al<sub>5</sub> and Fe. The intermediate and end products in SHS and sintering were characterised by SEM/EDX and XRD. The porosity level of the final FeAl40 product was controlled by the heating rate and powder size, which was also strongly influenced by the temperature, holding time and the ratio of the two powders.

**Key words:** sintering; pressureless; iron aluminide; self-propagating synthesis

## 1. Introduction

Of the iron aluminides, FeAl has a B2 structure and exists over a wide range of Al concentrations (36–50 at%) at room temperature. Iron aluminides based on FeAl exhibit better oxidation resistance than Fe<sub>3</sub>Al alloys and have lower densities compared to steels and commercial iron-based alloys, offering better strength-to-weight ratio [1]. These properties allow them to be considered for use as high-temperature structural materials, gas filters, heating elements, and fasteners etc..

Iron aluminides have been prepared by a variety of methods that include melting-casting, roll compaction, spark plasma sintering [2], mechanical alloying and **pressureless sintering** [3]. Consolidation of powder compacts for FeAl production has been achieved by using argon atmosphere hot isostatic pressing (HIP), hot pressing (HP) or hot extrusion in order to overcome the relatively poor sintering characteristics of elemental Fe–Al mixtures [4].

Reactive sintering methods, combustion synthesis and self-propagating high-temperature synthesis (SHS) based on the usage of elemental powders, have been extensively studied for various intermetallic systems like Ni–Al and Ni–Ti [5; 6]. These processes offer some advantages, including the use of less expensive and more readily available elemental powders, lower processing (sintering) temperatures, lower energy consumption and shorter processing times. Most of all, these processes can lead to stress-free products. However, pronounced porosity or extensive swelling during the heating stage is a major restriction to the development of these processes. Fast synthesis of FeAl via pressureless Spark Plasma Sintering at 900 °C for 3 min was reported to achieve porous compacts with open porosity of 41% for filter applications by making use of the rapid chemical reaction between iron and aluminium [2]. Charlot et.al. [7; 8] tried a low-temperature mechanically activated self-propagating synthesis process to synthesise nanocrystalline FeAl with a density close to 80%. In order to reduce the porosity, some researchers introduced pressure during the combustion or sintering stage which made the process complex and increased the cost; as a result the commercial uptake of such routes has been limited [4]. The swelling behaviour during reactive sintering of FeAl from the elemental powders was explained by an imbalance in solubility and a large diffusivity difference between the constituent elements, while some authors attribute this behaviour to the excess heat released during the formation of Fe<sub>2</sub>Al<sub>5</sub> [9]. Godlewska et. al. [10] prepared dense Fe<sub>40</sub>at%Al by pressureless sintering at 1200°C followed by hot-forging at 1100°C of intermetallic powders that had been obtained by SHS. In another pressureless sintering process involving SHS from elemental powders, a final FeAl density of 95 % was achieved by keeping the heating rate below 1 °C/min which minimised the volume expansion during the formation of the intermediate phase Fe<sub>2</sub>Al<sub>5</sub> [11].

A two-step process was reported as an alternative fabrication method of dense FeAl intermetallics [12]. Pre-sintering of compacted Fe and Al powders of a 60:40 ratio was conducted at 620°C/670°C under a static and a cyclic loading with a different frequency which consolidated and refined the grain of the intermetallic matrix to 10 µm. The second step involved pressureless sintering at a temperature of 1250°C. Kang et .al.[13] used Fe<sub>2</sub>Al<sub>5</sub> powder as a transient material to circumvent the poor sintering of FeAl specimen to fabricate a Fe<sub>3</sub>Al product with a density up to 96%. However, this process included cycles of cold rolling and annealing processes.

In the present study, a two-stage pressureless sintering method was investigated to produce FeAl<sub>40</sub> intermetallic; in the first stage, mixed iron and aluminium powders were compacted

and ignited at a temperature of 750°C to form Fe<sub>2</sub>Al<sub>5</sub> which was porous giving a swelling effect. Then the pellets were crushed and ball-milled and mixed with iron powder in the nominal composition of FeAl40 to sinter at a higher temperature. The second stage was a pressureless reactive liquid-phase sintering process utilising the low melting point of the Fe<sub>2</sub>Al<sub>5</sub> phase. The effect of temperature, heating rate, powder size and holding time on the porosity and densification of the sample were investigated to achieve a densified sintered product.

## 2. Experimental detail

During Stage I, the aluminium metal powders (-325 mesh, <44µm, 99.5%, oxygen <0.02%, Alfa Aesar) were mixed thoroughly with iron powders (<10µm, 99.5% and <1µm, 99.5%, 0.05% carbon and 0.27% oxygen, Alfa Aesar) and pressed at 70 N for 60 seconds to a pellet of diameter of 8 mm. The nominal weighed composition aimed to form Fe<sub>2</sub>Al<sub>5</sub>. The pellet was placed in a tube furnace in the presence of running argon and heated at a heating rate of 40°C/minute to 750°C for 5 minutes. The self-propagating high-temperature synthesis (SHS) was initiated by global ignition upon which the reactant powders were spontaneously converted to form a brittle and porous Fe<sub>2</sub>Al<sub>5</sub> product.

In Stage II, the pellets were crushed by a cleaned hammer and milled in a ceramic mortar to different sizes, and the powders were then screened and mixed with the aforementioned iron powders in the nominal composition of FeAl40 (Fe-40 at.% Al). The mixed milled powders were then weighed (0.5g) and cold pressed at 70 N for 60 seconds into cylindrical samples with a diameter of 8 mm by adding a few drops of isopropanol. After drying for 24 hours, the pellets were then placed in the tube furnace in running argon to undergo sintering at various temperatures for different durations as shown in Table 1.

**Table 1: FeAl40 sintering details in stage II.**

<b>Treatment parameters</b>	<b>Variation</b>
Heating rate (°C/minute)	1, 5, 20, 40
Temperature (°C)	600, 900, 1200, 1300, 1450
Duration (hour)	1, 2, 3, 4, 10
Powder size (Fe/Fe <sub>2</sub> Al <sub>5</sub> )	<10µm/<100µm, <10µm/<50µm, <1µm/<10µm, <1µm/<1µm
Composition (Fe/Fe <sub>2</sub> Al <sub>5</sub> )	Stoichiometric, 2%/5% excess Fe <sub>2</sub> Al <sub>5</sub> , 2% excess Fe

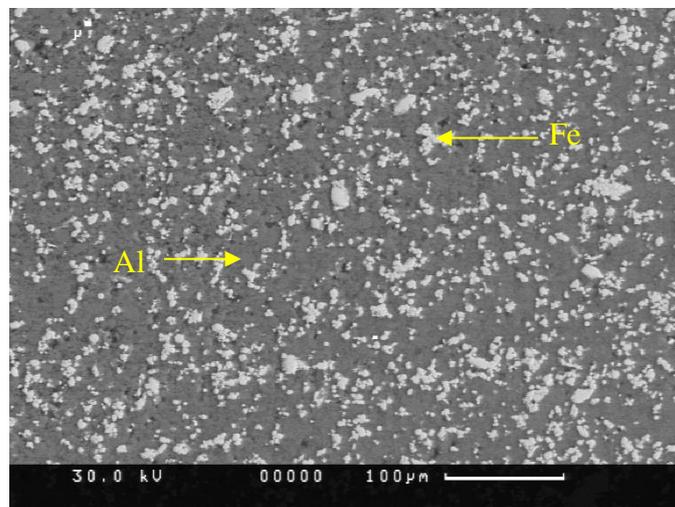
The porosity of the products was determined by examining the cross-section of sintered samples using optical microscopy with Struers Scantis software. Vickers Micro-hardness was

measured using a Leitz Wetzlar Micro-hardness Tester with a load of 50 g for 15 seconds. A JEOL 6300 scanning electron microscope (SEM) was used to examine the average powder size of the green compacts, the surface morphology and microstructure of the sintered samples. Elemental composition was analysed using the Energy dispersive X-ray spectroscopy (EDX) technique during SEM examination. The phase constituents were analysed by a X'pert Philips X-Ray Diffractometer using Cu-K $\alpha$  radiation ( $\lambda=0.154056\text{nm}$ ). The diffraction angle ( $2\theta$ ) was between  $20.01^\circ$ - $100^\circ$  and the scanning step was  $0.02^\circ$  for counting times of 1 second at each step. The experimental data were collected by a computer and analysed by PCAPD software for Automated powder diffraction.

### 3. Results

#### 3.1 Stage I: The formation of Fe<sub>2</sub>Al<sub>5</sub>

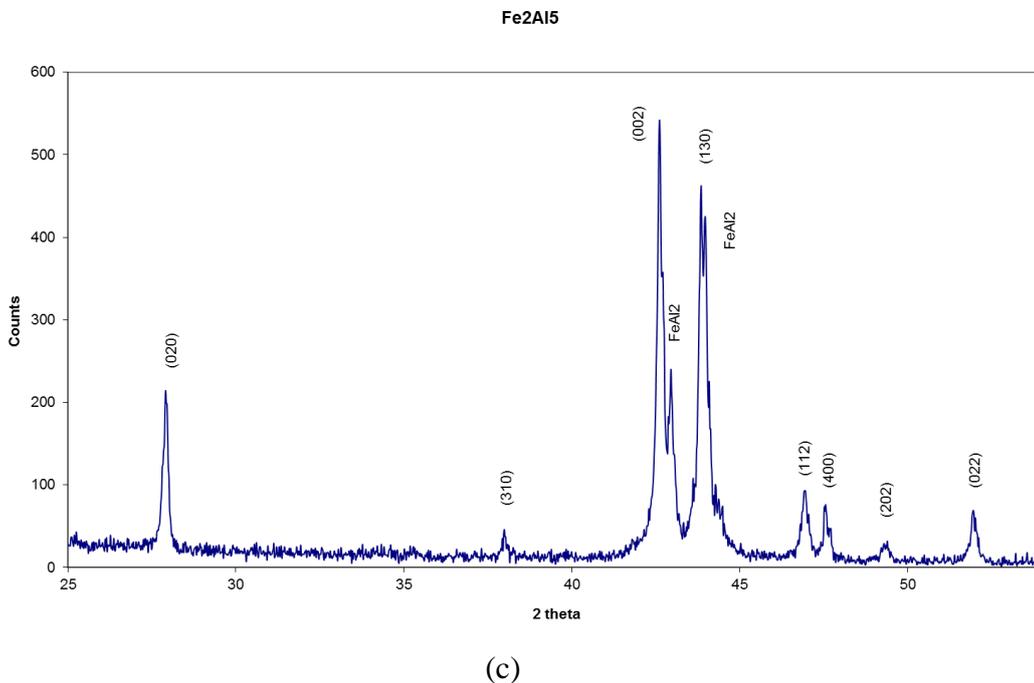
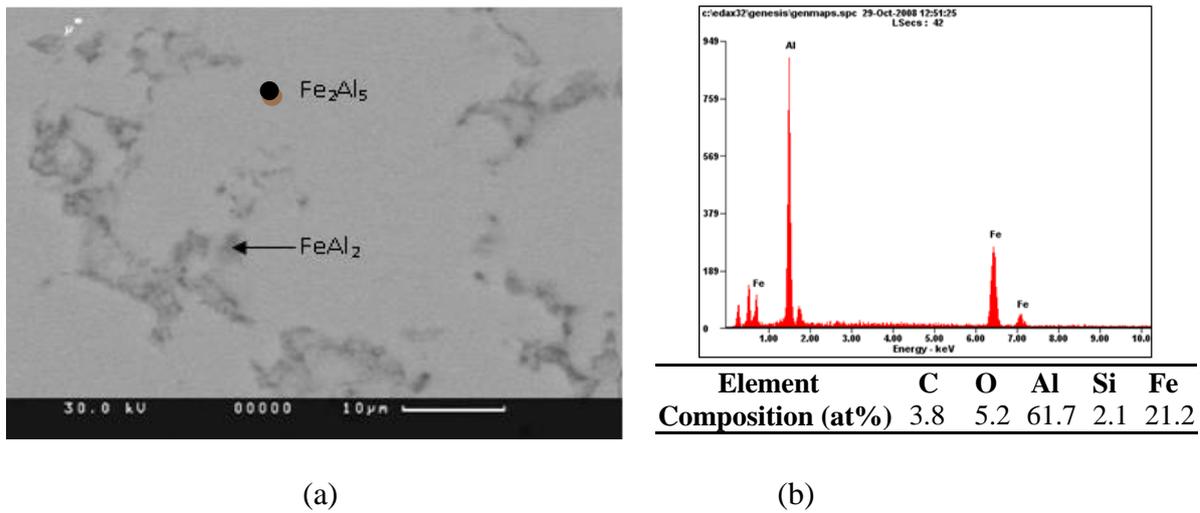
A pellet of mixed iron and aluminium powders in a molar ratio of 2:5 after pressing and drying is shown in Figure 1. The green compact was very dense due to the soft deformed aluminium powders accommodating the relatively hard iron powders. The bright iron powders were evenly distributed within the aluminium powders which were deformed due to high pressure.



**Figure 1: As-pressed iron and aluminium powder pellet mixture (BEI).**

During the rapid heating at  $40^\circ\text{C}/\text{min}$  to  $750^\circ\text{C}$ , an exothermal reaction occurred and a product with Fe<sub>2</sub>Al<sub>5</sub> as the dominant phase was formed as presented in Figure 2. The elemental composition of the main phase (dotted area) as collected by EDX analysis correlated the major product to Fe<sub>2</sub>Al<sub>5</sub>; the minor dark grey areas (Figure 2a) had a 1:2 ratio

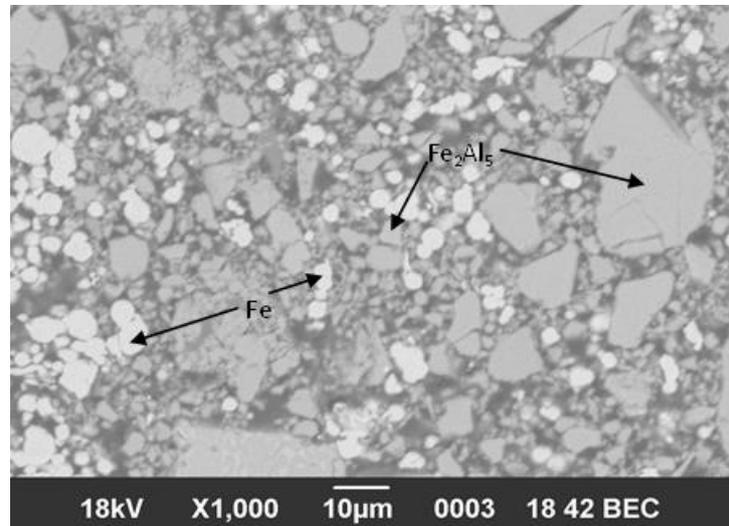
of Fe:Al which suggested the presence of FeAl<sub>2</sub> as a secondary phase. The phase constituents as identified by XRD demonstrated that Fe<sub>2</sub>Al<sub>5</sub> was the major product with a small amount of FeAl<sub>2</sub> (PDF34-0570) in agreement with the EDX analysis as shown in Figure 2c. Most of the Fe<sub>2</sub>Al<sub>5</sub> peaks can be correlated with the JCPDS Fe<sub>2</sub>Al<sub>5</sub> pattern (PDF 29-0043). The pellets were porous, brittle and easy to crush into small particles. The crushed particles were hand-milled and ball-milled to different sizes for use in the next stage of the process.



**Figure 2: Initial Fe<sub>2</sub>Al<sub>5</sub> product after combustion synthesis at 750°C for 5 minutes (a) microstructure (BEI), (b) EDX analysis on the dotted area and the elemental composition and (c) XRD of the phase constituents.**

### 3.2 Stage II: The formation and sintering of FeAl40

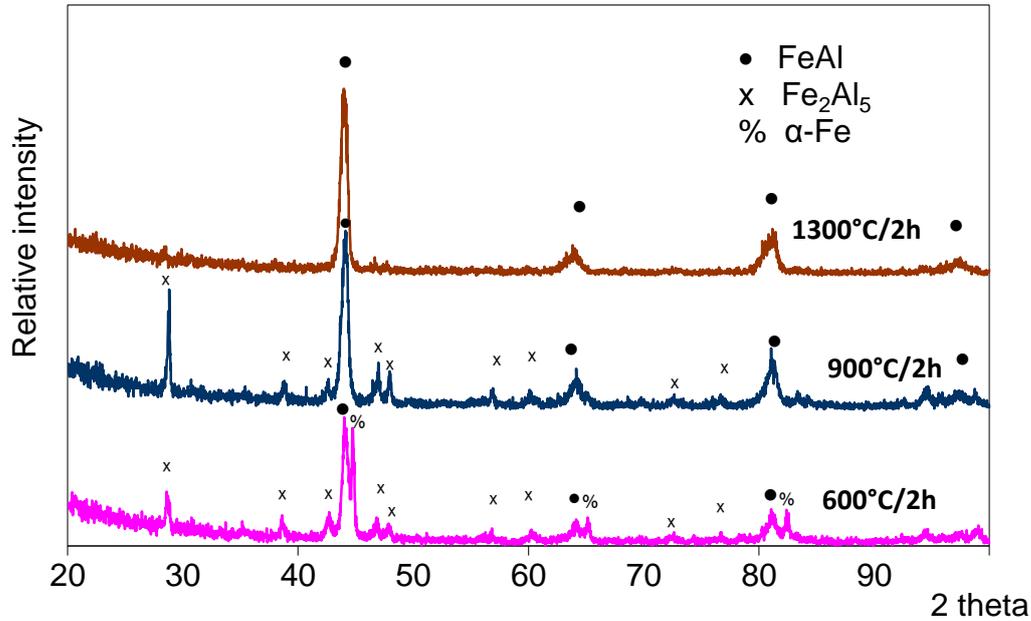
Milled  $\text{Fe}_2\text{Al}_5$  powders were mixed with Fe powders at such a ratio to produce a composition of FeAl40 and sintered under various conditions. A micrograph of the mixed green compacts is shown in Figure 3.



**Figure 3: BE image of the as-pressed mixtures of iron powders (<10μm) and milled  $\text{Fe}_2\text{Al}_5$  powders (<50μm).**

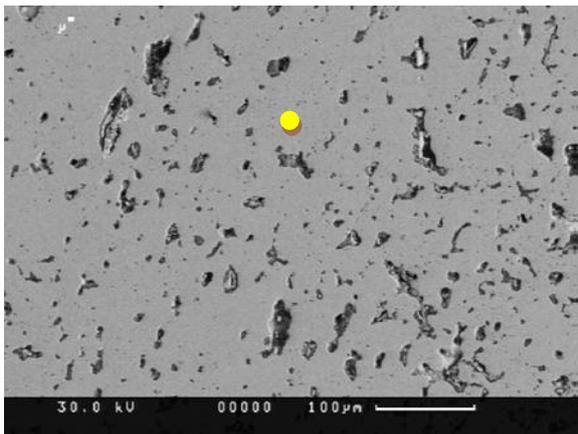
#### 3.2.1. Effect of sintering temperature

Different sintering temperatures (600°C/2h, 900°C/2h and 1300°C/2h) were used for the mixtures of powders of iron (<10μm) and  $\text{Fe}_2\text{Al}_5$  (<100μm), and the phase evolution is presented in Figure 4. Following sintering at 600 °C/2h, XRD peaks showing the presence of FeAl appeared but there were still substantial amounts of  $\alpha$ -Fe and  $\text{Fe}_2\text{Al}_5$ . After sintering at 900 °C/2h, the intensity of the FeAl peaks increased, while that of the  $\alpha$ -Fe peaks reduced significantly. After sintering at 1300°C/2h, only FeAl peaks could be detected with minor traces of  $\text{Fe}_2\text{Al}_5$ . Examination using SEM as presented in Figure 5a, showed the presence of large pores in the sintered sample; the porosity level was about 15% and the structure of the whole pellet was quite uniform with an elemental composition of iron of 59.5 at% and aluminium of 40.5 at% as indicated in Figure 5b.

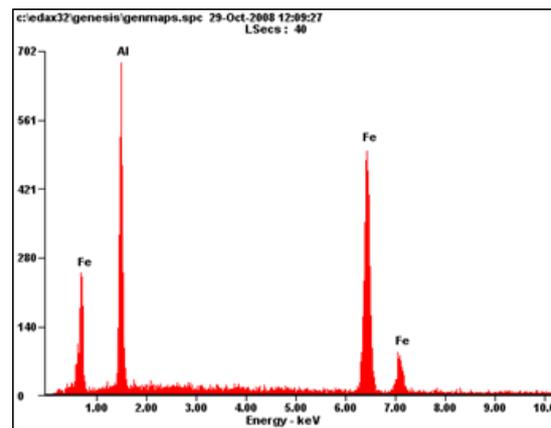


**Figure 4: Evolution of phase constituents after sintering at different temperatures.**

A lower sintering temperature generally led to a less dense structure. As shown in Table 2, the level of porosity of the pellet was about 15% after sintering at 1200°C for 2 hours with a heating rate of 40°C/min. The use of a higher temperature of 1450°C, which is above the melting temperature of FeAl<sub>40</sub>, led to a quite dense structure with low porosity especially in the centre of the pellet (6% or less), but it did not achieve full pellet densification (Table 2).



(a)



(b)

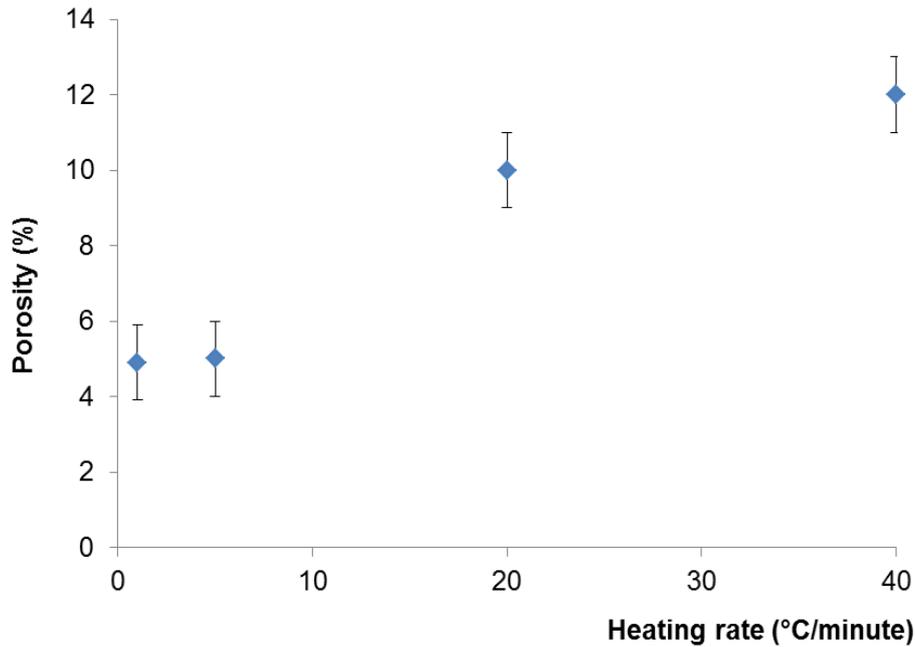
**Figure 5: FeAl sintered from a mixture of coarse powders of Fe<sub>2</sub>Al<sub>5</sub> (<100μm) and iron (<10μm) at 1300°C for 2 hours at 40°C/minute a) Microstructure (BEI) and b)EDX identification of the spot.**

**Table 2: Summary of the porosity of sintered FeAl40 pellets.**

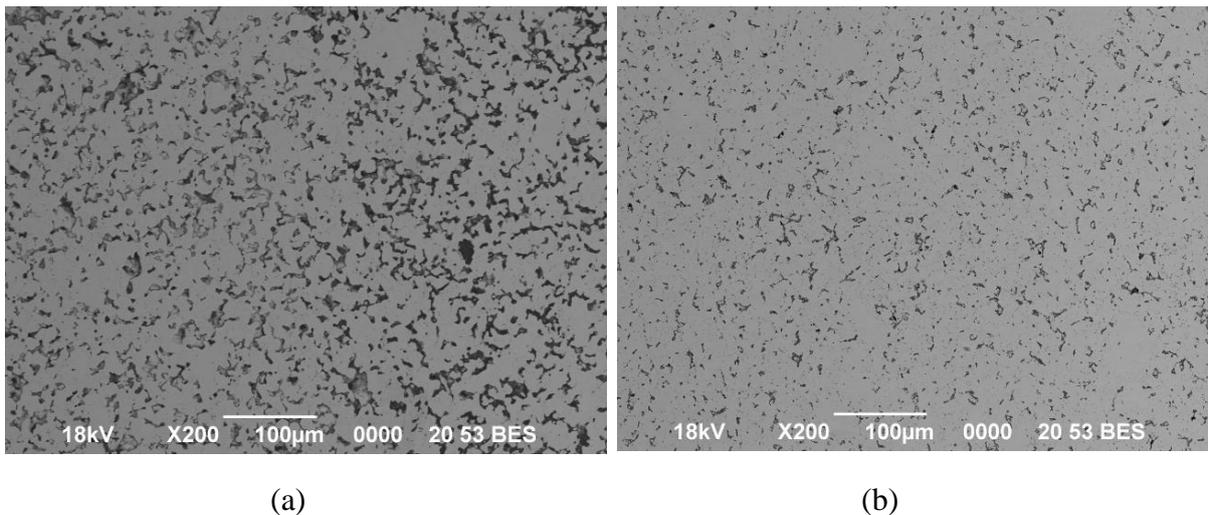
Powder		Treatment detail	Heating rate (°C/minute)	Porosity (%)
Fe	Fe <sub>2</sub> Al <sub>5</sub>			
<10µm	<100µm	600°C/2h	40	9
		900°C/2h	40	10
		1300°C/2h	40	15
<10µm	<50µm	1300°C/1h	40	12
			20	10
			5	5
			1	4.9
<10µm	<50µm	1300°C/10h	5	4.9
<1µm	<10µm	1200°C/2h	40	15
		1300°C/2h	40	7
		1450°C/2h	40	6
<1µm	<1µm	1300°C/1h	40	14.1
		1300°C/2h	40	14.0
		1300°C/3h	40	13.8
		1300°C/4h	40	13.5
<1µm	<10 µm, 2% excess	1300°C/1h	40	5
<1µm	<10 µm, 5% excess	1300°C/1h	40	10
<1µm, 2% excess	<10 µm	1300°C/1h	40	25

### 3.2.2. Effect of heating rate

The effect of the heating rate on the amount of sintering was investigated by varying the heating rate from 1, 5, 20 to 40 °C/minute as shown in Table 2. The level of porosity of the FeAl40 sample that was obtained from mixtures of Fe<sub>2</sub>Al<sub>5</sub> powders (<50µm) and iron powders (<10µm) sintered at 1300°C for 1 hour dropped gradually from 12% to 5% as the heating rate decreased (Figure 6). The compact sintered at 1300°C for 1h at the fastest heating rate of 40°C/minute had a porosity level of around 12% (Figure 7a) and the microhardness, HV<sub>0.05</sub>, of the sample was 203. Sintering at a heating rate of 5°C/min produced a quite dense structure with porosity of around 5% (Figure 7b) and the microhardness, HV<sub>0.05</sub>, was 344. This demonstrated that the sample microhardness increased as the level of sintering increased. The use of a heating rate of 1°C/min led to only marginally more densification than the 5°C/minute densification did.



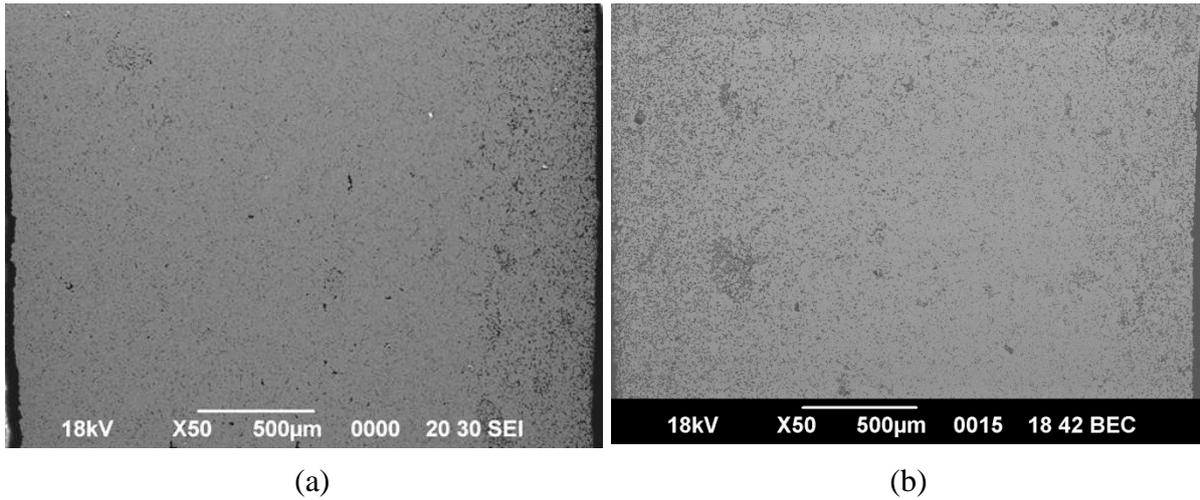
**Figure 6: The effect of heating rate on the porosity level of FeAl sintered from a mixture of  $\text{Fe}_2\text{Al}_5$  powders ( $<50\mu\text{m}$ ) and iron powders ( $<10\mu\text{m}$ ) at  $1300^\circ\text{C}$  for 1 hour.**



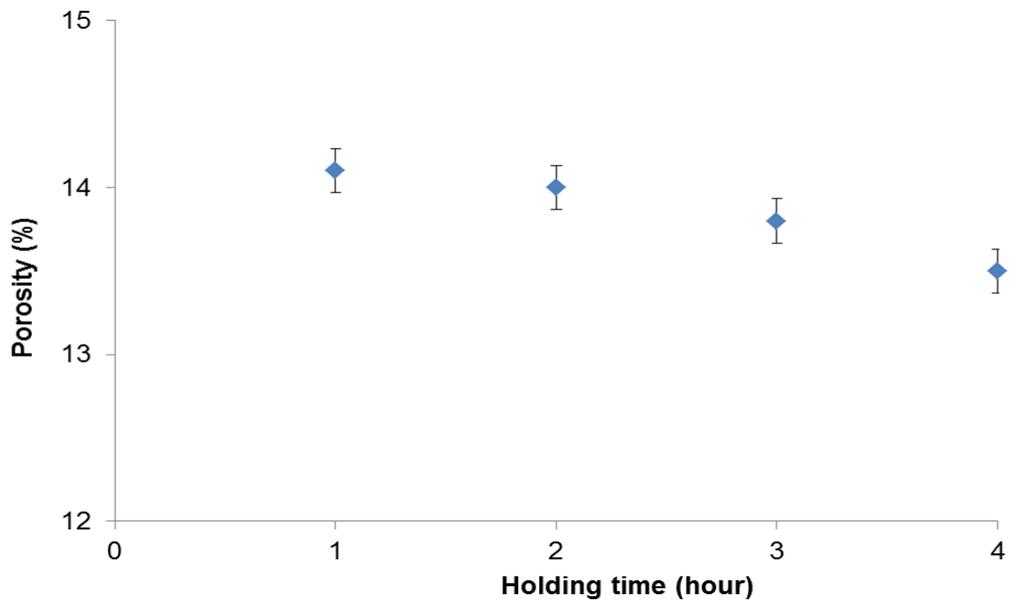
**Figure 7: BE images of FeAl sintered from mixtures of  $\text{Fe}_2\text{Al}_5$  powders ( $<50\mu\text{m}$ ) and iron powders ( $<10\mu\text{m}$ ) at  $1300^\circ\text{C}$  for 1 hour with different heating rates: a)  $40^\circ\text{C}/\text{min}$  d)  $5^\circ\text{C}/\text{min}$ .**

### 3.2.3. Effect of sintering time

The porosity of the sintered FeAl40 samples changed only marginally as the holding time extended from 1 hour to 10 hours for mixtures of  $\text{Fe}_2\text{Al}_5$  ( $<50\mu\text{m}$ ) and iron powders ( $<10\mu\text{m}$ ) sintered at  $1300^\circ\text{C}$  as displayed in Figure 8 and Table 2. A similar trend was also observed for the mixture of fined  $\text{Fe}_2\text{Al}_5$  ( $<1\mu\text{m}$ ) and iron powders ( $<1\mu\text{m}$ ) sintered at  $1300^\circ\text{C}$  as shown in Figure 9.



**Figure 8: FeAl sintered from mixtures of  $\text{Fe}_2\text{Al}_5$  ( $<50\mu\text{m}$ ) and iron powders ( $<10\mu\text{m}$ ) at  $1300^\circ\text{C}$  with a heating rate of  $5^\circ\text{C}/\text{minute}$  for (a) 1 hour (SEI) and (b) 10 hours (BEI).**

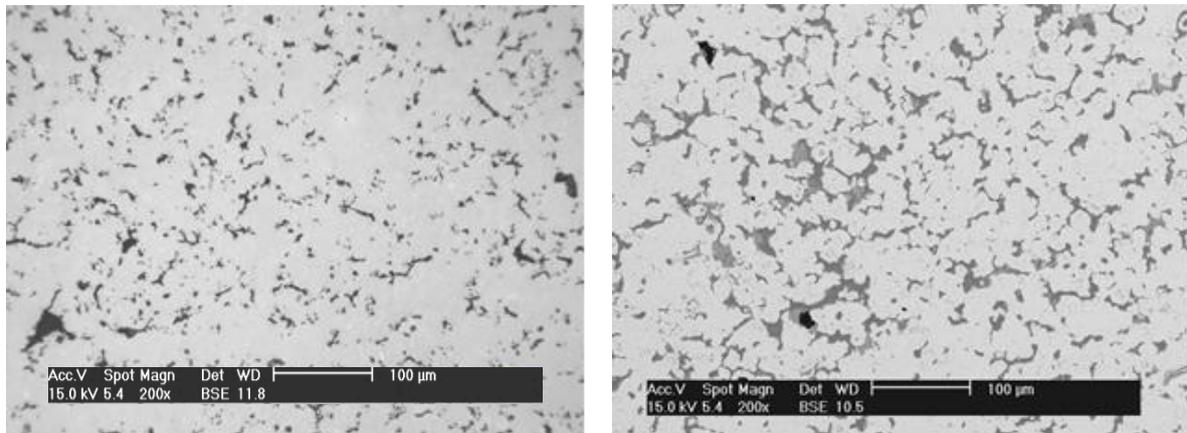


**Figure 9: Effect of holding time on the porosity level of FeAl sintered from a mixture of  $\text{Fe}_2\text{Al}_5$  powders ( $<1\mu\text{m}$ ) and iron powders ( $<1\mu\text{m}$ ) at  $1300^\circ\text{C}$  with a heating rate of  $40^\circ\text{C}/\text{minute}$**

### 3.3.4. Effect of powder size

It was observed that the reduction of the  $\text{Fe}_2\text{Al}_5$  powder size to below  $50\mu\text{m}$  from under  $100\mu\text{m}$  could obviously reduce the porosity of the sintering parts from 15% to 12% when sintered at  $1300^\circ\text{C}$  with a heating rate of  $40^\circ\text{C}/\text{minute}$  as shown in Table 2. The porosity level of the sintered parts was further reduced to 7% when a mixture of finer iron powders of less than  $1\mu\text{m}$  and  $\text{Fe}_2\text{Al}_5$  powder size below  $10\mu\text{m}$  was used (Figure 10a). When the size of both the powders was reduced to below  $1\mu\text{m}$ , the green compacts did not bind well and tended to break. Following careful handling, it was possible to sinter some pellets; sintering

at 1300°C from 1 to 4 hours led to porosity levels between 13.5-14.1% as shown in Figure 9 and Figure 10b.



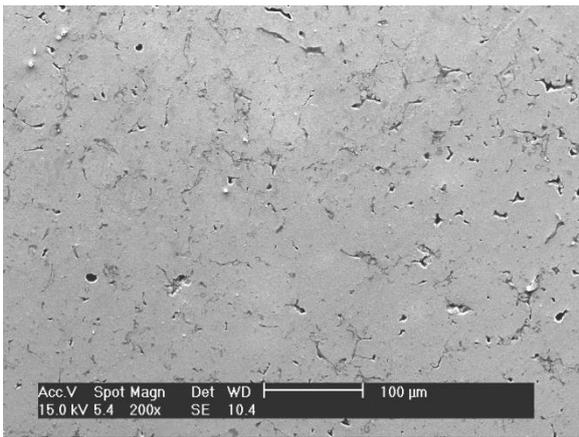
(a)

(b)

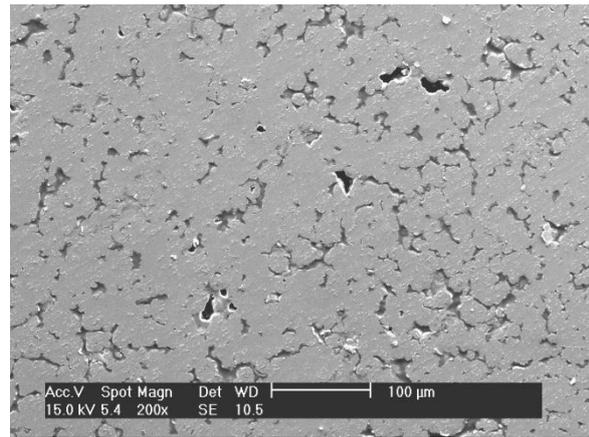
**Figure 10: BE images of the FeAl sintered at 1300°C for 2 hour with a heating rate of 40°C/minute from the mixture powders of (a) Fe <1μm and Fe<sub>2</sub>Al<sub>5</sub> < 10μm, and (b) Fe <1μm and Fe<sub>2</sub>Al<sub>5</sub> <1μm.**

### 3.2.5. Effect of composition and variation of the amounts of Fe<sub>2</sub>Al<sub>5</sub> and iron powder

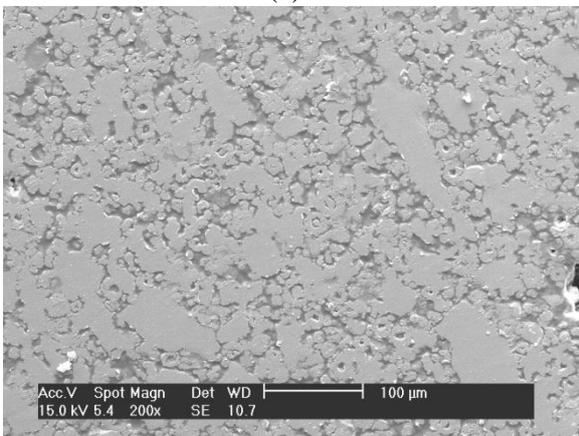
Variation of the relative amounts of Fe<sub>2</sub>Al<sub>5</sub> and iron powders was used in order to understand its effect on sintering. The use of 2 % excess of Fe<sub>2</sub>Al<sub>5</sub> powders in the composition resulted in a better result yielding a product of lower porosity <5% (Figure 11a) and a dense and nearly uniform single phase structure was obtained (Figure 12). However, the use of a 5% excess of Fe<sub>2</sub>Al<sub>5</sub> powders resulted in a more porous structure (13.7%) along with a two-phase microstructure containing FeAl and Fe<sub>2</sub>Al<sub>5</sub> (Figure 11b). The use of an excess of iron powders normally led to higher level of porosity (25%) and poor sintering (Figure 11c).



(a)

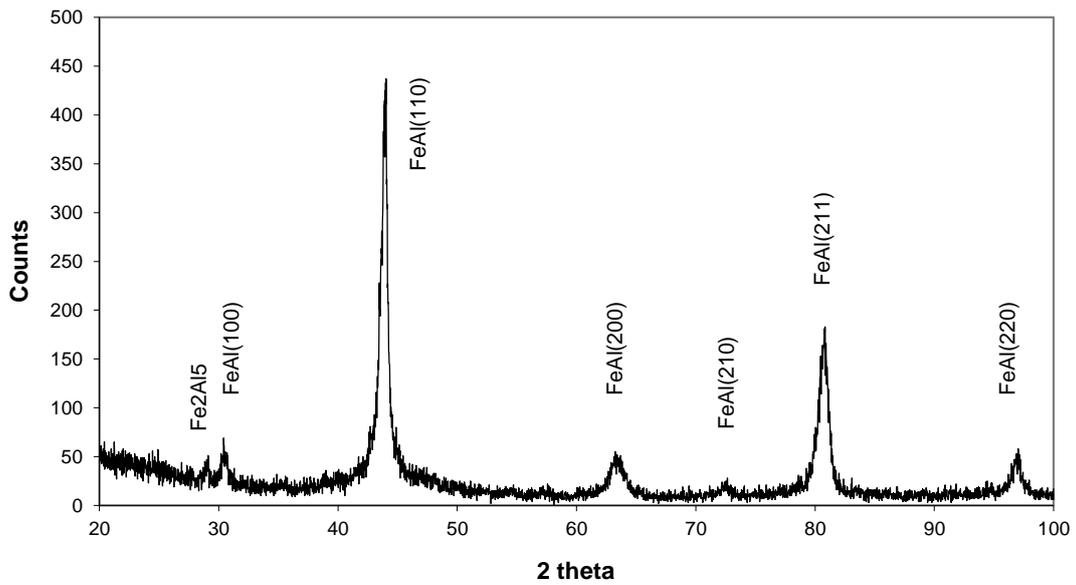


(b)



(c)

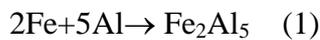
**Figure 11: SE images of FeAl sintered (1300°C/1 h) from a mixture of iron (<1μm) and Fe<sub>2</sub>Al<sub>5</sub> (<10μm) powders with different composition a) 2% excess Fe<sub>2</sub>Al<sub>5</sub> powder; b) 5% excess Fe<sub>2</sub>Al<sub>5</sub> powder and c) 5% excess iron powder.**



**Figure 12: FeAl sintered (1300°C/1 h) from a mixture of Fe<sub>2</sub>Al<sub>5</sub> and iron powders (<1µm/<10µm) with 2% excess Fe<sub>2</sub>Al<sub>5</sub> powders (FeAl, PDF45-0983).**

#### 4. Discussion

Developed by Merzhanov and Borovinskaya [14] in the late 1960s, the self-propagating high-temperature synthesis (SHS) process has been extensively employed for the production of ceramic, ceramic matrix composites, metal matrix composites and intermetallics. The technique is based on the use of exothermic reactions whereby the exothermic heat that is released converts the reactants to products at a fast rate. In the first stage of the present investigation, stoichiometrically mixed iron and aluminium powders bearing an intended composition of Fe<sub>2</sub>Al<sub>5</sub> were heated up to 750 °C for 5 minutes in order to initiate the SHS reaction (1):

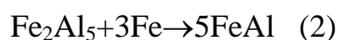


This led to global ignition of the reactants to spontaneously convert to Fe<sub>2</sub>Al<sub>5</sub>. As shown in Table 3, this reaction is exothermic with  $\Delta H^\circ = -30\sim 34.3\text{kJ mol}^{-1}$  [3; 9]. The reaction between Fe and Al was more violent and faster with a lower starting temperature than that between Ni and Al particles which led to a temperature hike and expansion or swelling of the reactant pellet [5]. In previous research [3], the maximum temperature during SHS in vacuum was reported to be 725°C for Fe29at%Al and 974°C for Fe71.5at%Al. Shearsby [15] observed a rapid expansion of the compacted powders of about 15% at a temperature around the melting point of Al. This violent and rapid release of exothermic heat was the driving force for the formation of Fe<sub>2</sub>Al<sub>5</sub> phase (Figure 2) as well as the cause of the expansion behaviour in many local contacted regions between iron and aluminium powders and resulted in the pronounced porosity in the products, therefore a brittle product.

**Table 3: Information of the materials used in this research**

	<i>Crystal structure</i>	<i>Density (g/cm<sup>3</sup>)</i>	<i>Melting point (°C)</i>	<i>Enthalpy change (ΔH° in kJ.mol<sup>-1</sup>)</i>
Al	FCC	2.7	660	
αFe	BCC	7.86	1538	
Fe <sub>2</sub> Al <sub>5</sub>	orthorhombic	3.96	1171	-30 to-34.3[3; 9; 11]
FeAl40	BCC	6.06	1370	-12 [3]

The formation of FeAl using Fe<sub>2</sub>Al<sub>5</sub> as an intermediate product can be described by reaction (2):



This is also an exothermic reaction with  $\Delta H^\circ = -6 \text{ kJ mol}^{-1}$  of FeAl as calculated from the data in Table 3 (using the less negative value for  $\text{Fe}_2\text{Al}_5$ ) and is less violent than reaction 1. The present investigation showed that although some FeAl was formed when sintering below 900 °C, near full densification of FeAl required a temperature above the melting temperature of  $\text{Fe}_2\text{Al}_5$  phase (1170 °C) as shown in Figure 4. Kang et al [13] reported that the  $\text{Fe}_2\text{Al}_5$  phase in Fe29%Al compacts disappeared after heating to above 850°C, while Gedevanishvili and Deevi [3] observed that Fe40at%Al powders began to sinter at a temperature of 1120 °C. Godlewska et al [10] reacted Fe and Al elemental powders in the proportion of Fe-40%atAl by SHS and subsequently milled the SHS product which contained various intermetallic phases. The milled SHS product was then pressed into compacts and underwent pressureless sintering at 1200°C. This second stage needed subsequent hot-pressing to yield products of low porosity. The use of a higher temperature of 1450°C led to higher levels of sintering with low porosity, but severe oxidation along grain boundaries even in an argon protected environment was observed. Oxidation had progressed inwards along the grain boundaries probably due to the fact that the sintering temperature had led to melting of the sample. In order to minimise the risk of oxidation, most of the sintering work in the current investigation was conducted below 1450°C, i.e. 1300°C.

The present work showed that a higher heating rate generally resulted in higher levels of porosity and in larger pores in the product from reaction (2) as displayed in Figure 5a, 7a and 7b. They also show residual  $\text{Fe}_2\text{Al}_5$  to be present within the pores and in particular within the larger pores. In comparative terms, the highest levels of densification were achieved when using a low heating rate of 1°C/minute. In order to understand this observation, it is important to consider the fact that the presence of a relatively low-melting phase like  $\text{Fe}_2\text{Al}_5$  is essential in the reactive liquid-phase sintering process which has been developed by the current work. The rate of conversion to the product has been described by Budnikov and Ginstling [16] by the equation,

$$\frac{dG}{dt} = K F C_1 C_2 \quad (3)$$

where G is the amount of conversion, t is time,  $C_1$  and  $C_2$  are the concentrations of the reactants, K is reaction rate constant and F represents the surface contact area. The value of K for both solid-solid and solid-liquid reactions will be dependent on the rate of diffusion of the reactants through the product.

Assuming that the thermodynamic conditions remain the same, the value of  $K$  can be expected to be the same for solid-solid and solid-liquid interactions. The reaction rate will thus depend on the value of  $F$  which is likely to be higher for solid-liquid interactions. Therefore, as in the case of the present research, the rate of conversion to the product is likely to be faster at temperatures that promote melting of  $\text{Fe}_2\text{Al}_5$ . However, by heating the reactants above the  $\text{Fe}_2\text{Al}_5$  melting point at a very fast rate of  $40^\circ\text{C}/\text{minute}$ , tends to promote the conversion reaction to  $\text{FeAl}$  due to the fast increasing temperature. Due to the formation of solid  $\text{FeAl}$ , the capillary action of molten  $\text{Fe}_2\text{Al}_5$  to surround the solid iron is restricted. The surface contact area,  $F$ , between the two reactants is therefore likely to be lower leading to a lower densification rate. Consequently, large irregular pores were left during the formation of  $\text{FeAl}$  (Figure 5a). The figure also shows the presence of small amounts of unreacted residual  $\text{Fe}_2\text{Al}_5$  which remained trapped within the larger pores giving further evidence that for this sample there was less surface contact between the two reactants.

The  $\text{Fe}_2\text{Al}_5$  powder that was prepared during the first part of the two-stage process with an average size between  $50\text{-}100\mu\text{m}$  also led to lower surface area contact resulting in lower levels of sintering. Reducing the size of the  $\text{Fe}_2\text{Al}_5$  powders obviously reduced the size of the pores when sintering at a high heating rate ( $40^\circ\text{C}/\text{min}$ ) although the sample was still relatively porous with a porosity about 12% as shown in Figure 7a. However, when the heating rate was reduced, i.e.  $5^\circ\text{C}/\text{min}$ , more liquid  $\text{Fe}_2\text{Al}_5$  phase was able to form and to flow by capillary action to surround the iron powder resulting in lower levels of porosity of 5% and to smaller pores (Figure 7b). As the temperature increased at a lower rate, the conversion to  $\text{FeAl}$  during the early stages was lower (because the temperature was lower). This was beneficial as there was a lower amount of  $\text{FeAl}$  in the early stages to impede the flow of the molten  $\text{Fe}_2\text{Al}_5$  to surround the iron powder. With time, this led to greater surface contact between the reactants and a greater level of porosity. At the same time, the pellet retained good cylindrical form with very little distortion. When the heating rate was further reduced to  $1^\circ\text{C}/\text{min}$ , a porosity level of 4.9% was measured which was very similar to the level that was obtained at a heating rate of  $5^\circ\text{C}/\text{min}$ . Sintering at a rate of  $5^\circ\text{C}/\text{min}$  for 10 hours also resulted in a porosity level of 4.9%. These observations suggest that the maximum densification that can be achieved by the process at  $1300^\circ\text{C}$  is about 95% and that there is hardly any benefit in using a heating rate below  $5^\circ\text{C}/\text{minute}$ . The figure of 95% as the limit of densification during pressureless heating is in agreement with earlier work by Gedevisishvili and Deevi [3].

The effect of the molten  $\text{Fe}_2\text{Al}_5$  during the densification process was further demonstrated by running some sintering experiments with 2%  $\text{Fe}_2\text{Al}_5$  in excess. This led to an increase in the liquid phase in the pellet and in the surface contact area after melting; the effect of this was to achieve porosity levels of 5% even with a sintering rate of  $40^\circ\text{C}/\text{minute}$  as shown in Figure 11a. However, more  $\text{Fe}_2\text{Al}_5$  (5% excess) didn't lead to higher densification which might be due to the oversupplied transient liquid phase helped to form a rigid solid FeAl skeleton faster thus left the formation of secondary pores rich in  $\text{Fe}_2\text{Al}_5$  residues as seen in Figure 11b. Similar phenomena was also reported on sintering  $\text{FeAl}_3$  with surplus aluminium powder by Savitskij [6]. On the other hand, excess iron powders normally led to poor sintering because there was comparatively a lower amount of the liquid phase and this resulted in less favourable sintering conditions as displayed in Figure 11c.

The effect of  $\text{Fe}_2\text{Al}_5$  and Fe powder size was more complex. Generally it is expected that the smaller the powder size, the more the material contact and thus the better the sintering. In this research, it was observed that when the size of  $\text{Fe}_2\text{Al}_5$  powder mixed with iron powders ( $<10\ \mu\text{m}$ ) was reduced from  $<100\ \mu\text{m}$  to  $<50\ \mu\text{m}$  and  $<10\ \mu\text{m}$ , the sintering result improved. However, when the size of both the iron and the  $\text{Fe}_2\text{Al}_5$  powders was below  $1\ \mu\text{m}$ , it became very difficult to get the green powders to bind together by pressing. Even when using isopropanol as a binder, once it dried up, the pellets were difficult to handle as they broke up easily. When sintering was attempted with such fine powders, sometimes a crack could develop in the sintered pellet.

High-purity argon was used for the pressureless reactive liquid-phase sintering of FeAl, but mild oxidation could still be a problem particularly when using finer powders. This is because high-purity argon can contain a small amount of oxygen (1 ppm) that is sufficient for some oxidation to occur as demonstrated in the analysis shown in Figure 2. However, according to the XRD result and EDX analysis, oxides were not clearly found in the sintering.

## 5. Conclusions

The following conclusions can be drawn from this research:

1. A two-stage sintering process was successfully used to sinter FeAl to densification levels of just above 95% at a temperature of  $1300\ ^\circ\text{C}$ . The initial stage involved the production of  $\text{Fe}_2\text{Al}_5$  pellets obtained using SHS by heating mixed iron and aluminum powders up to  $750^\circ\text{C}$ ; the  $\text{Fe}_2\text{Al}_5$  product which was porous due to the violent

exothermic self-propagating reaction was easy to crush into fine powder. FeAl intermetallic compound was subsequently synthesised and sintered during the second stage by means of a reactive liquid-phase sintering process by reacting mixed powders of milled Fe<sub>2</sub>Al<sub>5</sub> and iron at a higher temperature.

2. Dense structures (porosity less than 5 %) were obtained by sintering at 1300 °C using heating rates below 5°C/min, while higher porosity was observed when using high heating rates (20 °C/min and 40 °C/min). The effectiveness of the low heating rates in achieving high densification was due to the ability of Fe<sub>2</sub>Al<sub>5</sub> to melt and surround the iron to react. At the higher heating rates, the higher amount of the initially-formed FeAl product (due to the higher temperatures attained while heating) impeded the flow of the molten Fe<sub>2</sub>Al<sub>5</sub> and this resulted in a lower amount of surface contact with the iron.
3. Extra fine Fe<sub>2</sub>Al<sub>5</sub> and iron powders (<1µm) increased the surface area and the activity of the powders with a sacrifice of the adherence among the powders, therefore compromised the sintering product.

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