Fabrication and squeeze casting infiltration of graphite/alumina preforms

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The manufacturing of a suitable rigid porous graphite/alumina preform has been investigated taking into account the influence of the binder type, the graphite/alumina content in the preform and the percentage of binder in water. The preforms showing an acceptable rigidity have been infiltrated with a CuSn12 bronze alloy by squeeze casting considering two different pouring temperatures. The composite quality is strongly influenced by both the graphite/alumina volume fractions and the binder type. The optimal quality has been obtained by infiltrating a Carsil 2000^{TM} binded preform containing 30 vol % graphite flakes and 70 vol % alumina fibres.

1. Introduction

Most studies in the field of sliding systems such as bearings are related to the development of new materials offering a high thermal conductivity, a low coefficient of friction [1] with respect to the counterpart material, and, the ability to withstand high surface temperatures [2]. Presently employed bearings are mainly either single metal systems or bimetal ones made of high thermal conductive materials such as copper or aluminium, being their main drawbacks related to poor mechanical and frictional properties [3].

The objective of the work is to produce a copper matrix composite for sliding bearing applications able to compete with actual systems in terms of performance and price. Copper is a high thermal conductive material able to withstand high temperatures, but showing poor frictional and mechanical properties [4]. The copper frictional properties should be significantly improved by introducing lubricant particles to copper. Graphite is the most widely employed solid lubricant due to its low coefficient of friction, good thermal properties, a rather low cost and an increasing efficiency under impure atmosphere such as hydrocarbons, moisture, or oil often encountered in bearing's area [5]. Mechanical properties of copper are likely to be increased by adding oxide, nitride or carbon reinforcements [6]. Although long fibres or whiskers would be able to impart high strength, low cost short oxide fibres have been chosen in the study. Squeeze casting has been employed to produce copper composite materials. Squeeze casting is particularly suited to the production of near-net-shape components with an excellent properties/price ratio [7, 8]. The production of a copper reinforced composite by squeeze casting involves the manufacturing of a rigid porous graphite/alumina preform able to withstand the extreme infiltrating conditions required by copper alloys. The strength and stiffness of the preform is achieved through the use of a binder able to support the pressure and temperature to be applied in the squeeze casting process. The first part of the work is dedicated to the investigation a suitable binder for producing a preform made with both graphite lubricating particles and short oxide fibres. Then, the processing parameters involved in the squeeze casting process are revised, including the suitable characteristics recommended for each of them.

2. Experimental procedure

2.1. Materials

The oxide fibres are "Saffil" short fibres, 3–5 μm in diameter and 200–400 μm long. The chemical composition is around 96–97% δ-alumina and 3–4% silica. The alumina/silica "shot" content, i.e. large spherical particles, resulting from the production of the fibres is very high and has to be eliminated before the fabrication of the preforms. Natural graphite flakes, 160 μm in size, were used for lubrication purposes. The particles have a carbon content of 97% with a density close to 0.7 g/cm^3. Different binders have been investigated for the preparation of a suitable rigid porous graphite/alumina preform. The sodium silicate binder (Carsil 2000^{TM}) is a typical SiO_2/Na_2O inorganic liquid binder widely used for the production of sand moulds [9]. Carsil 2000^{TM} is able to chemically react with alumina fibres by applying a CO_2 flow and heat treating the green preform at 1000 °C for 15 min under inert atmosphere. The colloidal silica binder (Syton X30^{TM}) is a dispersion of submicronic silica particles in aqueous solution. The strength of the graphite/alumina preform is achieved through a chemical reaction between the silica particles and the alumina fibres after a heat treatment at 1000 °C for 30 min under inert atmosphere. The binder Fabutit 705^{TM} is based on the ortophosphoric acid (H_3PO_4), which reacts with oxide products such as alumina to form aluminium phosphates. A typical thermal treatment at 900 °C for 1 h under inert atmosphere is recommended for optimum strength of the body. The binder Silubit FB10^{TM} is a liquid mixture of 81 wt % alumina particles and 15 wt % submicron silica particles [10]. The preform heat treatment is one hour at 900 °C under inert atmosphere. The binders used are non-expensive products of easy manipulation that do not require special storage conditions. The alloy used for preform infiltration is a Cu-12wt%Sn binary bronze (CuSn12), in accordance with the standards DIN 1705. This material is sometimes used to produce bearings able to withstand high loads and sliding speeds.

2.2. Fabrication of the preform

The industrial process usually used for the production of both commercial δ-alumina short fibre preforms and thermal insulating materials was applied at a laboratory scale to produce the preforms. A given amount of both short fibres and graphite particles is immersed in water together with the suitable content of binder and dispersing agent. The quantity of water is approximately two times the total weight of solids. The mixture is then mechanically stirred until a uniform slurry is obtained (Fig. 1). The mixture is poured into a cylindrical mould containing multisize filters and connected to a vacuum pump. The body formed is removed from the mould and dried at 80 °C. A heat treatment is applied before the infiltration process.

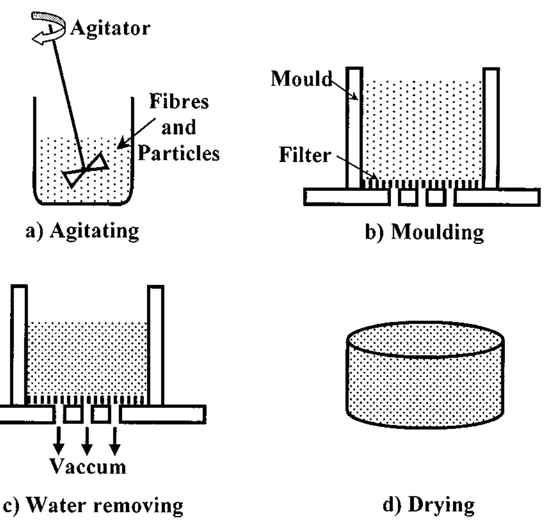


Figure 1 Experimental procedure for the graphite/alumina preform production.

Depending on the graphite/alumina content, the final volume fraction of reinforcements ranges from 12% to 15%, while the amount of binder in the preform runs from 5% to 10% of the total volume of solid (low binder contents in the preform are always obtained with high dilute solutions). For each binder, three different graphite/alumina volume fractions, i.e. 30/70, 50/50 and 70/30 and two percentages of binder dilution in water have been investigated. The quality of the preforms after heat-treating has been estimated by considering four standards corresponding to:

Standard 1: brittleness of the preform before heat treatment, damaged in green manipulation

Standard 2: brittleness of the preform after heat treatment, difficult to handle without breaking

Standard 3: rather brittle preform after heat-treating but quite easy to handle

Standard 4: strong preform after heat treating and easy to handle

The preforms with a standard of quality no. 1 and 2 have not been considered strong enough to be infiltrated, while those characterised by standards no. 3 or 4 have been chosen for a later infiltration by squeeze casting.

2.3. Squeeze casting infiltration

The preform is introduced within a furnace at least 30 min before its infiltration in order to ensure a homogeneous temperature around 1000 °C within the whole body. An inert atmosphere is maintained during heating by means of an argon flow of 8 l/min. The squeeze casting mould and ram are heated with electric resistances to a temperature in the range 300–350 °C during 30 min. The mould/ram system is coated with colloidal graphite for lubrication purpose. The alloy is melted under inert atmosphere in a refractory-coated silicon carbide crucible. The preheated preform is taken out of the electric furnace and located inside the heated mould (Fig. 2a). The molten alloy is poured inside the heated mould, over the porous preform, and a pressure around 110 MPa is applied onto the liquid to infiltrate

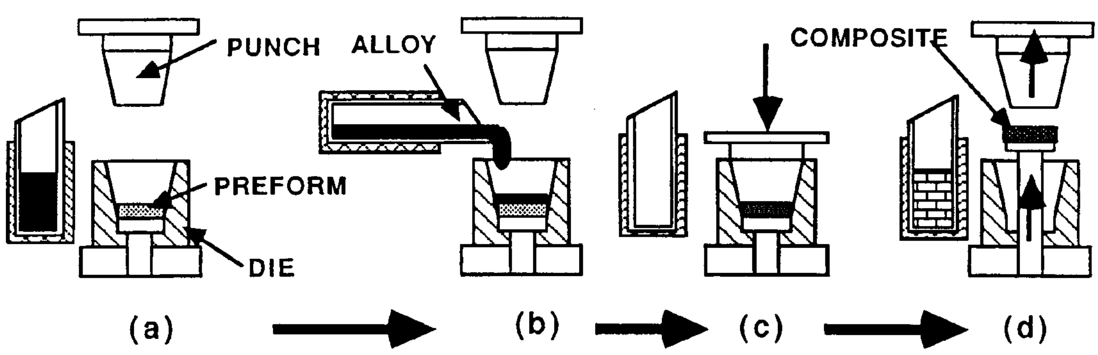


Figure 2 Squeeze casting infiltration process of a preheated preform.

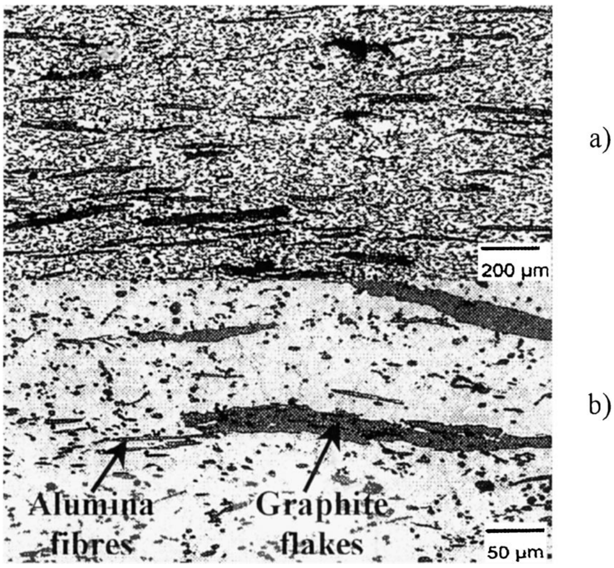


Figure 3 Microstructure of a CuSn12/Graphite-Alumina infiltrated composite at low (a) and high (b) magnification.

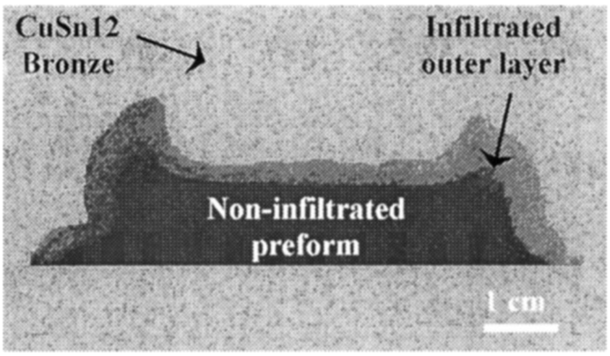


Figure 4 Partially infiltrated preform with a standard of quality no. 1.

the porous body (Fig. 2b–2c). After 15 sec, the mould is rapidly cooled still under pressure, leading to small grain microstructures. The component is finally extracted (Fig. 2d) and cut in different areas in order to estimate the infiltration quality. A typical microstructure of the CuSn12/Graphite-Alumina composite is shown in Fig. 3. The quality of the infiltrated preform has been estimated by considering five standards corresponding to:

Standard 1: partial infiltration of a thin layer at the outer surface of the preform—great deformation (Fig. 4)

Standard 2: quasi complete infiltration of the preform—great deformation (Fig. 5)

Standard 3: quasi complete infiltration of the preform—not much deformation (Fig. 6)

Standard 4: complete infiltration of the preform—some deformation (Fig. 7)

Standard 5: complete infiltration of the preform—no deformation (Fig. 8)

3. Results and discussion

3.1. Preform quality

Experimental results concerning the study of the preform quality with respect to the binder type, the

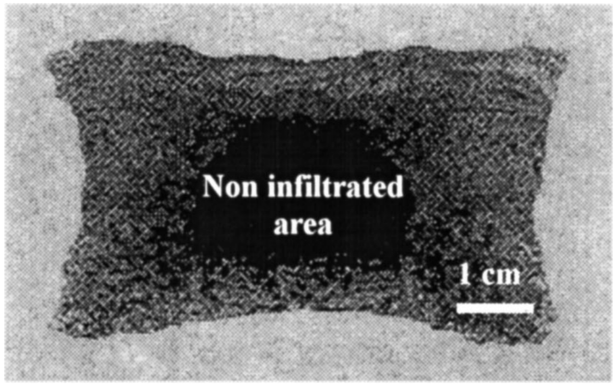


Figure 5 Nearly fully infiltrated preform with great deformation (standard of quality no. 2).

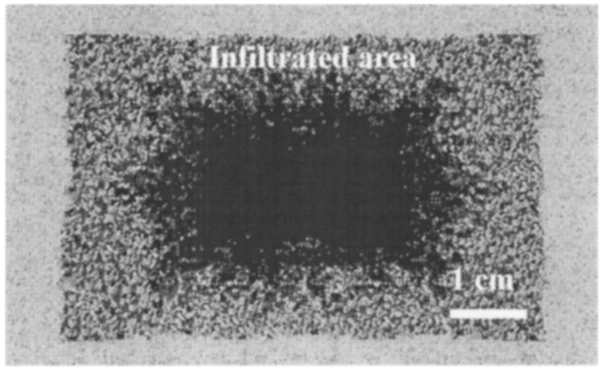


Figure 6 Nearly fully infiltrated preform with some deformation (standard of quality no. 3).

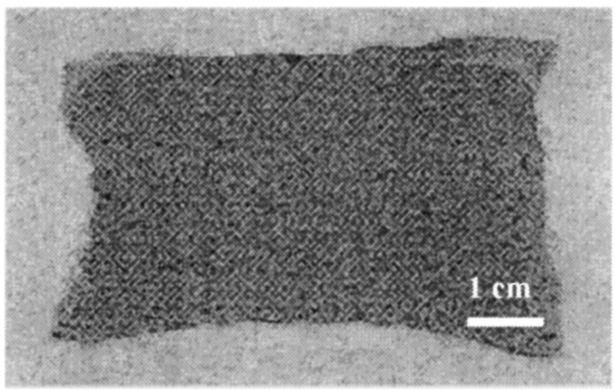


Figure 7 Fully infiltrated preform with some deformation (Standard of quality no. 4).

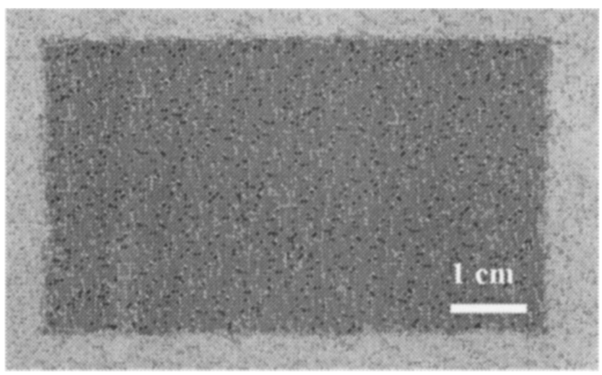
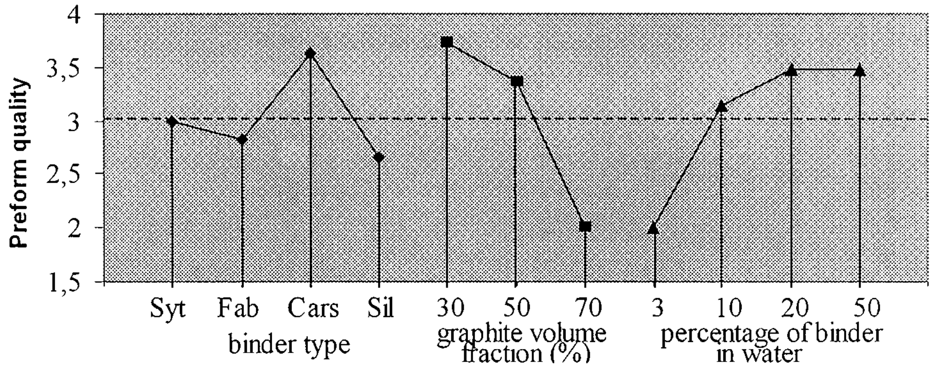


Figure 8 Fully infiltrated preform without deformation (level of quality no. 5).

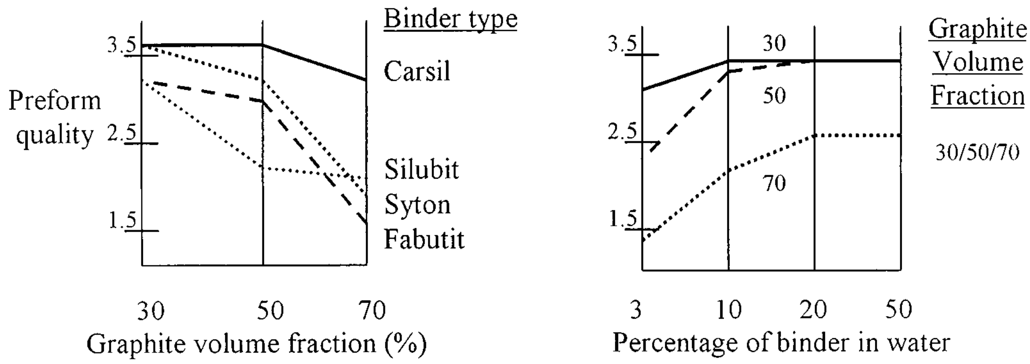
(a)

MAIN EFFECTS PLOT



(b)

INTERACTION PLOT



(c)

Parameters	Variance	Fisher coefficient	Threshold of confidence
Binder type	1.15	6.19	0.006
Graphite volume fraction	6.79	36.49	0.000
Percentage of binder in water	4.12	16.34	0.000

Figure 9 Results of the statistical design for the graphite/alumina preform fabrication.

graphite/alumina content $V_{F\text{-}Gr/Al}$ in the preform and the percentage B% of binder dilution in water are shown in Table I. A statistical experimental design has been applied to investigate the influence of each parameter on the preform quality. Fig. 9 summarises the results related to (i) the main effects plot (Fig. 9a), (ii) the interaction plot (Fig. 9b), and (iii) the analysis of variance (Fig. 9c). Taking into account the main effect plot, the three parameters are likely to be significant, which is confirmed by the analysis of variance. Each parameter has a high Fisher coefficient associated with a low threshold of confidence inferior to 1%. Considering the interaction plot, there is no evidence of interactions between the graphite/alumina volume fractions and the percentage of binder in water, while there may be one between the binder type and the graphite/alumina volume fractions. The binders selected in the study enable the fabrication of suitable preforms with a no. 3 minimum standard when the preform processing conditions are properly chosen. The graphite/alumina volume fraction is a very critical parameter as confirmed by its high Fisher coefficient. In any case, low graphite contents (30%) enable adequate preforms with standards no. 3 or 4 to be produced whatever the binder dilution and type. Increasing the graphite content gives rise to a decrease in the preform quality (Fig. 9a), but this

TABLE I Summary of the perform processing experiments with the four binders selected

Preform no.	Binder type	$V_{\text{f-Gr/Al}}$	B%	Preform quality
1	Syton X30™	30/70	10 vol %	4
2	Syton X30™	50/50	10 vol %	3
3	Syton X30™	70/30	10 vol %	1
4	Syton X30™	30/70	50 vol %	4
5	Syton X30™	50/50	50 vol %	4
6	Syton X30™	70/30	50 vol %	2
7	Fabutit 705™	30/70	3 wt %	3
8	Fabutit 705™	50/50	3 wt %	3
9	Fabutit 705™	70/30	3 wt %	1
10	Fabutit 705™	30/70	20 wt %	4
11	Fabutit 705™	50/50	20 wt %	4
12	Fabutit 705™	70/30	20 wt %	2
13	Carsil 2000™	30/70	10 wt %	4
14	Carsil 2000™	50/50	10 wt %	4
15	Carsil 2000™	70/30	10 wt %	3
16	Carsil 2000™	30/70	50 wt %	4
17	Carsil 2000™	50/50	50 wt %	4
18	Carsil 2000™	70/30	50 wt %	3
19	Silubit FB10™	30/70	3 wt %	3
20	Silubit FB10™	50/50	3 wt %	1
21	Silubit FB10™	70/30	3 wt %	1
22	Silubit FB10™	30/70	20 wt %	4
23	Silubit FB10™	50/50	20 wt %	4
24	Silubit FB10™	70/30	20 wt %	3

behaviour is more or less pronounced depending on the binder used. This observation can be explained considering that all the binders investigated are especially dedicated to bond oxide materials together. When the graphite content increases, the alumina fiber volume fraction decreases and so does the quality of the preform as a result of less bonds per unit volume. Whatever the graphite volume fraction, the lower the dilution of the binder, the better the quality of the preform (Fig. 9a). Silubit FB10™ can be considered as a suitable binder when low dilute solutions are used, but the mixture is difficult to filter by suction whatever the processing route chosen. For high dilute solutions, this binder must be rejected, except for low graphite contents where a grade no. 3 is obtained. Fabutit 705™ and Syton X30™ give rise to similar conclusions, but some differences can be noticed. For high concentrated solutions, these binders are able to generate adequate quality preforms as Silubit FB10™ does, except for high graphite contents around 70% where a standard no. 2 is obtained. For high diluted solutions, these binders are not so bad comparing with Silubit FB10™, since adequate preforms can be produced for the two lowest graphite percentages. Carsil 2000™ is the most effective binder for producing a suitable preform since a no. 3 minimum quality is always achieved whatever both the graphite content and the binder dilution. Carsil 2000™ is the only binder which allow the fabrication of a 70/30 graphite/alumina preform. It is especially important for high lubrication requirements.

3.2. Preform infiltration quality

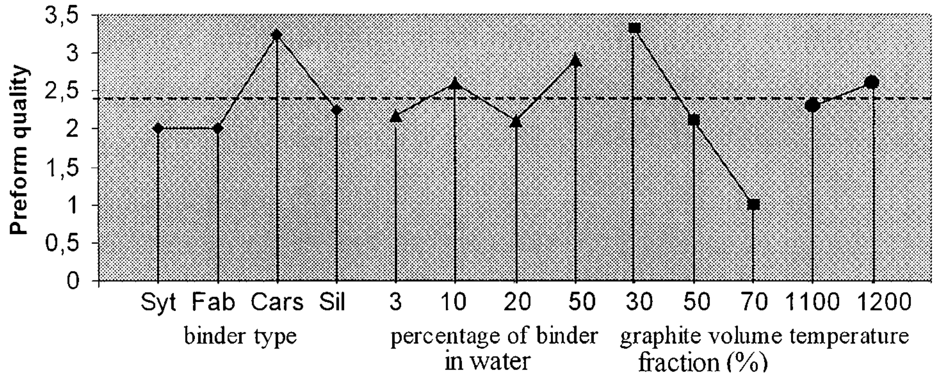
Experimental results concerning the study of the preform infiltration quality with respect to the binder type, the graphite/alumina content $V_{\text{F-Gr/Al}}$ in the preform, the percentage B% of binder in water and the metal pouring temperature T_M are shown in Table II. A statistical experimental design has been used to investigate the influence of those parameters on the preform infiltration quality. Fig. 10 summarises the results related to (i) the main effects plot (Fig. 10a), (ii) the interaction plot (Fig. 10b), and (iii) the analysis of variance (Fig. 10c). The binder type and the graphite/alumina volume fractions are the most influencing parameters as presumed by the main effects plot (Fig. 10a) and established by the analysis of variance. Both parameters have a high Fisher coefficient associated to a low threshold of confidence. The infiltration temperature is not a significant parameter in the range running from 1100 °C to 1200 °C. The interaction plot did not show any significant interaction between the squeeze casting temperature and the other parameters. The infiltration quality is strongly influenced by both the binder type and the graphite/alumina volume fractions. As shown in Table II, Carsil 2000™ is the most effective binder in producing adequately infiltrated preform. A standard 4 or 5 preform is always obtained, except for high graphite volume fraction around 70 vol %. The regression equation for the

TABLE II Summary of the perform infiltration experiments with the four binders selected

Composite no.	Preform no.	Binder type	B%	$V_{\text{f-Gr/Al}}$	T_M	Preform infiltration quality
1	1	Syton	10 vol %	30/70	1200	3
2	1	Syton	10 vol %	30/70	1100	2
3	2	Syton	10 vol %	50/50	1200	1
4	2	Syton	10 vol %	50/50	1100	1
5	4	Syton	50 vol %	30/70	1200	3
6	4	Syton	50 vol %	30/70	1100	3
7	5	Syton	50 vol %	50/50	1200	2
8	5	Syton	50 vol %	50/50	1100	1
9	7	Fabutit	3 wt %	30/70	1200	3
10	7	Fabutit	3 wt %	30/70	1100	2
11	8	Fabutit	3 wt %	50/50	1200	1
12	8	Fabutit	3 wt %	50/50	1100	1
13	10	Fabutit	20 wt %	30/70	1200	3
14	10	Fabutit	20 wt %	30/70	1100	3
15	11	Fabutit	20 wt %	50/50	1200	2
16	11	Fabutit	20 wt %	50/50	1100	1
17	13	Carsil	10 vol %	30/70	1200	5
18	13	Carsil	10 vol %	30/70	1100	4
19	14	Carsil	10 vol %	50/50	1200	4
20	14	Carsil	10 vol %	50/50	1100	4
21	15	Carsil	10 vol %	70/30	1200	1
22	15	Carsil	10 vol %	70/30	1100	1
23	16	Carsil	50 vol %	30/70	1200	5
24	16	Carsil	50 vol %	30/70	1100	5
25	17	Carsil	50 vol %	50/50	1200	4
26	17	Carsil	50 vol %	50/50	1100	4
27	18	Carsil	50 vol %	70/30	1200	1
28	18	Carsil	50 vol %	70/30	1100	1
29	19	Silubit	3 wt %	30/70	1200	3
30	19	Silubit	3 wt %	30/70	1100	3
31	22	Silubit	20 wt %	30/70	1200	3
32	22	Silubit	20 wt %	30/70	1100	3
33	23	Silubit	20 wt %	50/50	1200	2
34	23	Silubit	20 wt %	50/50	1100	2
35	24	Silubit	20 wt %	70/30	1200	1
36	24	Silubit	20 wt %	70/30	1100	1

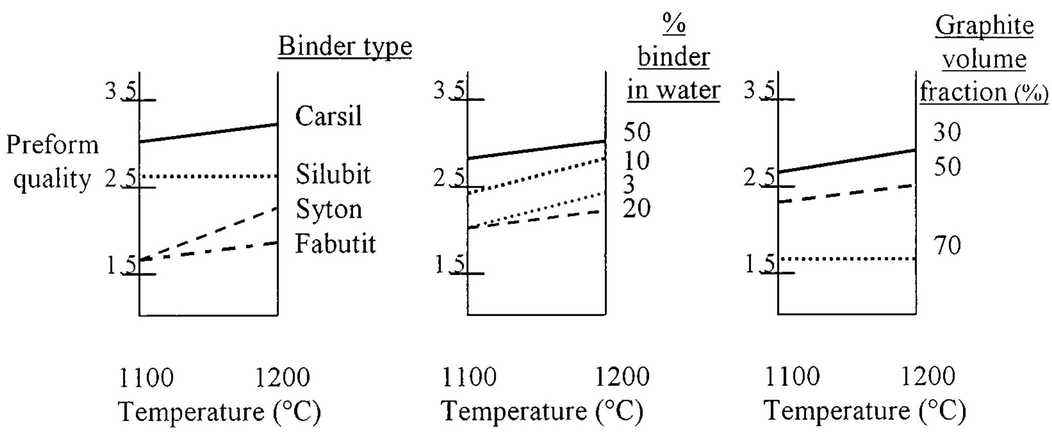
(a)

MAIN EFFECTS PLOT



(b)

INTERACTION PLOT



(c)

Parameters	Variance	Fisher coefficient	Threshold of confidence
Binder type	3.74	5.04	0.007
Graphite volume fraction	12.91	17.4	0.000
Percentage of binder in water	1.31	1.77	0.178
temperature	0.69	0.94	0.342

Figure 10 Results of the statistical design for the squeeze casting infiltration of preforms.

infiltration quality of Carsil binded preforms is given as follows:

$$\text{Infiltration quality} = 5.9 + 0.0042 \times \%\text{binder} - 0.094 \times \text{vol}\,\%\,\text{graphite} + 0.0017 \times \text{temperature} \quad (1)$$

The optimum infiltration quality is strongly related to the graphite volume fraction as shown by its high 0.094 proportional coefficient. Low graphite contents around 30 vol % are recommended for better infiltration quality. However, taking into account the objective of producing a self-lubricating composite, a 50/50 graphite/alumina preform binded with Carsil 2000^{TM} should be prefered.

4. Conclusions

The binder type and the graphite/alumina volume fractions are the main processing parameters which have to be carefully optimised in order to obtain a suitable preform before and after infiltration. Carsil 2000™ is the most effective binder of those considered in the study. It is also worth noting that the lower the graphite content, the better the quality of the preform whatever the binder type. The composite material with the highest level of quality is obtained by infiltrating a Carsil 2000™ bonded preform containing 30 V_F% of graphite. For lubrication purposes, a 50/50 graphite/alumina preform binded with Carsil 2000™ is recommended.

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