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Possibility of wettability improvement of Al_2O_3 preforms infiltrated by liquid aluminium alloy by deposition Ni-P coating

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ABSTRACT

Purpose: The purpose of this work is to present the method of wettability improvement of sintered Al_2O_3 preforms by deposition of Ni-P coating.

Design/methodology/approach: The ceramic preforms were manufactured by sintering of powder Al_2O_3 Alcoa CL 2500, with the addition of pores forming agent in the form of carbon fibres Sigrafil C10 M250 UNS of Company SGL Carbon Group. The internal surfaces of ceramic preforms were coated with Ni-P in order to improve the Al_2O_3 wettability by the liquid aluminium alloy. Coated by Ni-P ceramic preforms were pressure infiltrated with the liquid EN AC-AlSi12 alloy. Metallographic examinations were made in the scanning electron microscope (SEM) equipped with an energy dispersive spectrometer (EDS) of the structures and chemical composition of obtained materials.

Findings: Presented in this paper, deposition technology of Ni-P coating on the inner surfaces of ceramic preform can be used as a method of improving the wettability of porous Al_2O_3 ceramics by infiltrated liquid aluminium alloy.

Practical implications: The composite materials made by the developed method can find application in many industries as the elements of devices where beside the benefits from utilizable properties the small weight is required.

Originality/value: The obtained results show the possibility of manufacturing the composite materials by the pressure infiltration method of porous sintered preforms inner coated by Ni-P with liquid aluminium alloy being a cheaper alternative for conventional materials.

Keywords: Composites; Wettability; Infiltration

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MATERIALS

1. Introduction

Metal matrix composites reinforced with ceramic fibres or particles have become of great interest. The combined effects of metallic and ceramic materials relative to the corresponding monolithic alloys. Thus, various manufacturing methods have been developed, such as stir casting, powder metallurgy or infiltration of the porous, ceramic preforms with liquid metals alloys being a connection of both methods [1-11].

Infiltration of ceramic porous preforms by liquid alloys has attracted interest recently due to its relatively low cost and capability to produce net or near net shape fibre/particle reinforced or partially reinforced components. In the case of particle or short fibre reinforcement the predominant interest has been from engine manufacturers incorporating the material into the reciprocating parts of the internal combustion engine. Any reduction of weight of these components has a great effect on the efficiency of the engine. In addition the incorporation of ceramic phase within the material can greatly improve its wear and thermal fatigue resistance [12-18].

The connection of aluminium's properties as a matrix and particles or ceramic fibres as reinforcement in composite materials is dependent on the created during the technological process structure metal-ceramics. Unfortunately, the wettability of ceramic particles or fibres with liquid aluminium alloys is generally poor. Consequently, the introduction of reinforcement into a liquid matrix id difficult. Various procedures have been recommended to improve the wetting of ceramic particles or fibres by liquid metal, and include increasing metal liquid temperature, increasing infiltration pressure, and deposition of coating into the reinforcement material. One of the coating conception, mainly described in literature, is deposition of Ni-P coating on the Al_2O_3 [19-23].

The degree of wetting is usually estimated using the value of the contact angle θ (in the range 0° - total wettability to 180° no wettability). Conventionally systems metal-ceramic are divided into wettable system θ<90° and not wettable ones θ>90° (Fig. 1).

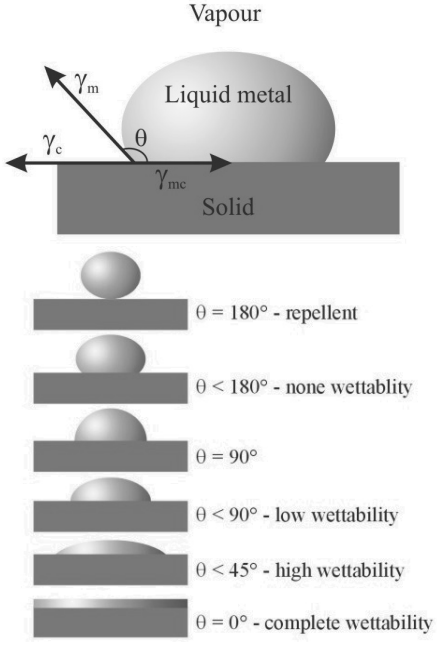


Fig. 1. Schema illustrates the definition of wettability angle θ according to the Young's equation of equilibrium and characteristic of metal ceramics systems depending on wettability angle θ value [24, 25]

On the value of θ angle, in case of influence of liquid aluminium into Al_2O_3, can impact the following factors: surface roughness, base porosity and chemical composition, contaminations, contact time, atmosphere, presence of oxide coating on the metal drop, what in the case of aluminium is the most essential factor deciding about the wettability. The reason of that is the fact that aluminium has high value to oxidation, creating continuous and compact oxide coating, which is the barrier inhibiting the creation of sufficiently good contact for metal with the base. The coating makes difficult the interaction between phases contribute to increase of θ angle [7, 15].

The goal of this work is present the possible method of wettability improvement of Al_2O_3 preforms infiltrated by liquid aluminium alloy by deposition Ni-P coating. Moreover influence of Ni-P layer deposited onto reinforcement on the structure of Al-Al_2O_3 composite materials was examined.

2. Experimental procedure

The material for investigation was manufactured by pressure infiltration method of porous ceramic performs with liquid aluminium alloy. The composites matrix consisted of eutectic aluminium-silicon alloy EN AC - AlSi12, as the reinforcement the porous ceramic performs with different porosity were used.

Ceramic preforms were manufactured by Al_2O_3 Aloca CL 2500 powder sintering method with addition of pore forming agent in form of carbon fibres Sigrafil C 10 M250 UNS from SGL Carbon Group company. Properties and chemical composition are shown in Table 1. The use of carbon fibres as the pores forming agent decides of high purity process, because during their degradation only CO_2 is the oxidation product, while using the cellulose or sawdust, the furnace's walls are covered with hard to clean tar stains. The addition of the carbon fibres was 30, 40 and 50% of weight. The chemical composition of the used ceramic powder are shown in Table 2.

Table 1.
Properties of Sigrafil C10 M250 UNS carbon fibers

Property	Value
Fibre diameter, μm	8
Mean fibre length, μm	135
Fibre density, g/cm^3	1.75
Tensile strength, GPa	2.5
Young's modulus, GPa	26
Carbon content, %	>95

Table 2.
Chemical composition of Alcoa CL 2500 powder

Mean mass concentration of elements, wt.%						
Al_2O_3	Na_2O	Fe_2O_3	SiO_2	CaO	B_2O_3	The others
99.80	0.05	0.02	0.01	0.01	0.01	0.10

Manufacturing of the ceramic preforms included preparation of powder and carbon fibres mixture, their pressing and sintering. The Al_2O_3 powder was wet grinded in ball mill to destroy particles agglomerations. Into the suspension the carbon fibres were added and polyvinyl alcohol Moviol 18-8 soluble in water (binding agent). Mixture of powder prepared in such way was dried by freezing and water sublimation in low pressure. Dry powder was sieve through a sieve No 250 μm, and then placed onto the flat surface and sprayed with distilled water to activate the polyvinyl alcohol. After 24h the powder was submitted to uniaxial pressing with laboratory Fontune TP 400 hydraulic press fitted with 45×65 mm. steel form. The pressure was 100 MPa and pressing time was 15 s. Compacts were sintered in "Gero" pipe furnace in air atmosphere (20 l/min). The temperature during the sintering process was ensuring the carbon fibres degradation (heating by 10h in temp. 800ºC) and Al_2O_3 powder sintering in temperature of 1500ºC by 2 h.

The porosity (ceramic phase content) of preforms was established on the basis of geometric measurement of their weight with the known Al_2O_3 particles density. The porosity of the received semi-finished products was 68.80% (31.20% ceramic phase content) for perform obtained from powder with 30% addition of carbon fibres, 75.60% (24.40% ceramic phase content) for perform obtained from powder with 40% addition of carbon fibres and 80.40% (19.60% ceramic phase content) for perform obtained from powder with 50% addition of carbon fibres. For the comparison of properties of the composite materials on the grounds of the produced frameworks with materials reinforced by fibrous preforms for further studies the commercial semi-finished products were used with 25% portion of Al_2O_3 fibres.

Table 3.
Process of Ni-P deposition on the Al_2O_3 substrate

Treatment	Surface activation		Deposition of Ni-P layer
Reagent	Futuron Activator	Noviganth Activator AK II	Noviganth PA Chemical Nickel
Temperature °C	room temperature	40-45	55
Time, min	1	3	1.5

To improve the Al_2O_3 wettability by the liquid aluminium alloy, the internal surfaces of both types of ceramic preforms were coated with Ni-P. To activate the ceramic surface the technology of Futuron Atotech company was used to cover the polymer material by metal coating. After completing the procedure (futuron activation) on the surface th layer of tin and palladium was formed. In order to eliminate a solution of tin the Noviganth Activator AK II was used. This process is designed to replace tin from the surface of ceramics on metallic palladium. On so prepared substrate a layer of Ni-P alloy by chemical plating was applied. The exact process is described in Table 3.

Firstly compacted Al_2O_3 plates covered by Ni-P were used to confirm proper process progress as shown in Fig. 1.

Fig. 1. Sample of Al_2O_3 plates with deposited Ni-P coating

Deposition of internal surfaces of porous ceramic preforms was possible by application of special device which allows to pump solutions over the preforms (scheme is shown in Fig. 2). The device was made from aluminium alloy, on internal parts (to avoid the reaction aluminium-reagents) the ceramic lacquer coating was deposited. To assure the require temperature of solution during coating deposition, on the device the cooper spiral coil was installed.

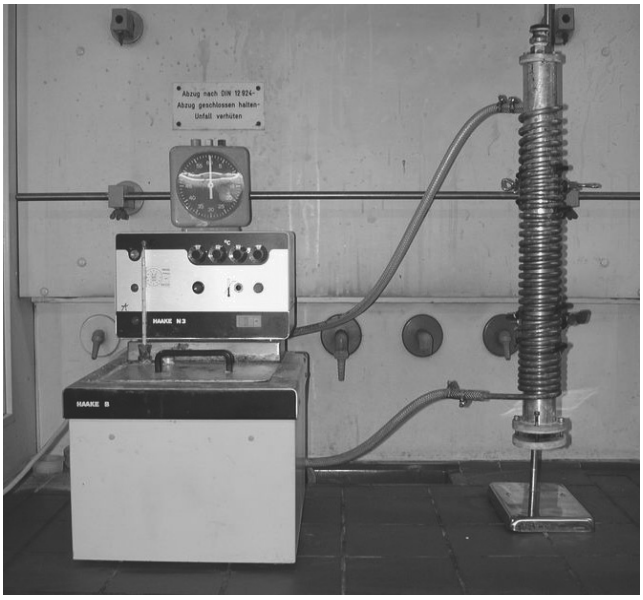
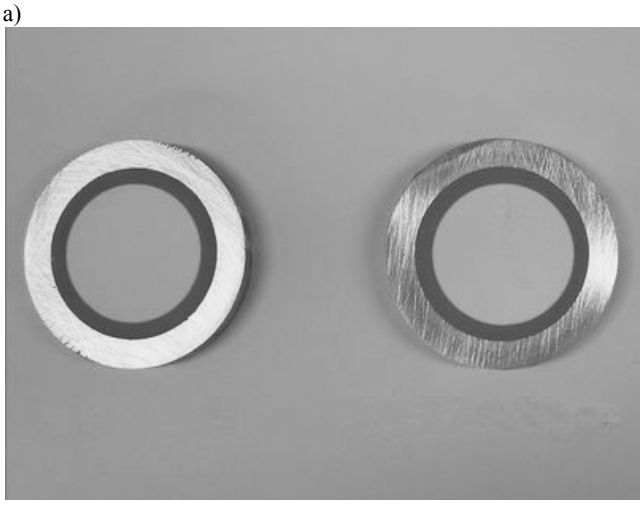


Fig. 2. Device for deposition of internal surface of porous ceramic preforms

To prevent damage during installation of skeletons in the device (the system must be leak-proof, which requires proper clamping of the sample) they were pasted using a two-component adhesive "UHU plus endfest 300" to aluminum rings. Pictures of skeletons before and after nickel deposition are shown in Fig. 3. Semi-products covered by nickel were cut from rings in the place of glue-ceramic boundary.

a)



b)

Fig. 3. Pictures of ceramic skeletons a) before nickel deposition, b) with Ni-P coating deposited onto internal surface

The permeability measurements were made on device for deposition of internal surface of porous ceramic preforms (Figs. 2 and 4) pumping over the water through the coated and uncoated porous materials. During the test, the flow time of 350 ml water was measured, adequately under the pressure of: 0.5; 1; 1.5 and 2 bars at room temperature. The permeability of preforms was calculated with formula:

$$D = \frac{\eta \cdot L \cdot \dot{V}}{A \cdot \Delta p} \tag{1}$$

where:
D – the permeability, m^2;
η – liquid viscosity, $N \cdot s/m^2$;
L –thickness of preform, m;
V – flow, m^3/s;
A - surface of preform, m^2;
Δp – pressure growth, Pa.

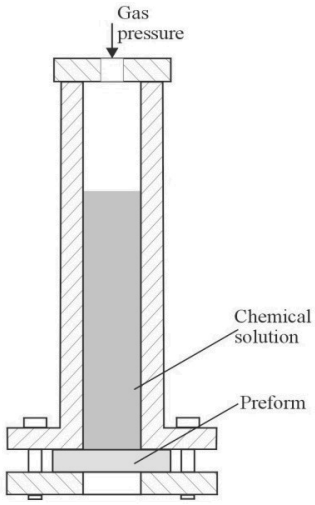


Fig. 4. The scheme of a permeability measure device of ceramic preforms

The observation of the structure of ceramic preforms fractures, manufactured by the sintering of Al_2O_3 Alcoa CL 2500 powder method after deposition of Ni-P layer was carried out using the Zeiss Supra 25 scanning electron microscope.

All types of uncoated and coated by Ni ceramic preforms were heated in furnace up to temperature 800ºC. Covered by graphite form was warmed up to 450ºC (maximal temperature of the press plates) and then fulfilled with preform and liquid alloy EN AC - AlSi12 with temperature of 800ºC, which chemical composition is shown in Table 4. The whole was covered by the stamp and placed in hydraulic plate press Fontune TP 400. The maximum infiltration pressure was 100 MPa and its influence was 120 s. After solidification obtained materials were removed from the form and cool down under pressured air stream.

Table 4.
Chemical composition of EN AC-AlSi12 aluminium alloy

Mean mass concentration of elements, wt.%							
Si	Fe	Cu	Mn	Zn	Ti	Others	Al
12	≤0.55	≤0.05	≤0.35	≤0.15	≤0.2	≤0.15	The others

Qualitative and quantitative X-ray microanalysis of obtained composite materials were made in scanning electron microscope (SEM) Zeiss Supra 25 equipped with a diffuse X-ray detector ROENTEC EDS at 15 kV accelerating voltage.

For the better identification of materials and a more transparent interpretation of their characteristics in tables and graphs, the following indications were used:

P30 – a composite material reinforced by a porous skeleton of Al_2O_3 powder prepared from a 30% mass portion of carbon fiber,

P40 – a composite material reinforced by a porous skeleton of Al_2O_3 powder prepared from a 40% mass portion of carbon fiber,

P50 – a composite material reinforced by a porous skeleton of Al_2O_3 powder prepared from a 50% mass portion of carbon fiber,

F – porous composite material reinforced with fiber backbone Al_2O_3 produced a 25% mass portion of ceramic phase.

3. Experimental results and their discusion

The observation of the structure of ceramic preforms fractures, manufactured by the sintering of Al_2O_3 Alcoa CL 2500 powder method after deposition of Ni-P layer carried out using the scanning electron microscope shown the uniform distribution of Al_2O_3 particles (Fig. 5). In addition, on ceramic's surface was observed the Ni-P coating deposited, but on the base of observation preforms it is difficult to examine its continuity and thickness which leads to similar studies of the final composite material.

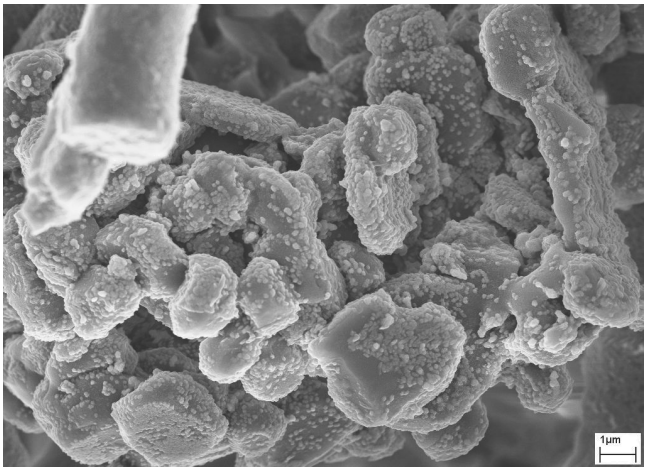


Fig. 5. The microstructure of ceramic preforms fracture based on Al_2O_3 powder with deposited Ni-P layer

The results of permeability measurements skeletons made of ceramic powders of Al_2O_3 with mass portion of carbon fibres 30, 40 and 50% pressed under 100 MPa load and commercial fibre semi-products are presented in Table 5 and in Figure 6. The greatest permeability show skeletons manufactured by sintering Al_2O_3 powder with 50% mass portion of carbon fibers forming the pore structure and channel rate of 11.12 $m^2 \cdot 10^{-13}$ while the lowest value - 2.56 $m^2 \cdot 10^{-13}$ have skeletons made of Al_2O_3 powder by the smallest 30% mass portion of carbon fibers. Commercial fiber semi products are characterized by a permeability of 10.07 $m^2 \cdot 10^{-13}$, more than twice the value for the skeletons produced by sintering of Al_2O_3 particles with 40% mass portion of carbon fiber with a similar portion of the ceramic phase, where the permeability is 4.56 $m^2 \cdot 10^{-13}$.

Table 5.
Results of permeability measurements of ceramic preforms

Material Description	Ni-P layer	Permeability $m^2 \cdot 10^{-13}$
P30	No	2.56
	Yes	2.48
P40	No	4.56
	Yes	4.22
P50	No	11.12
	Yes	10.56
F	No	10.07
	Yes	9.44

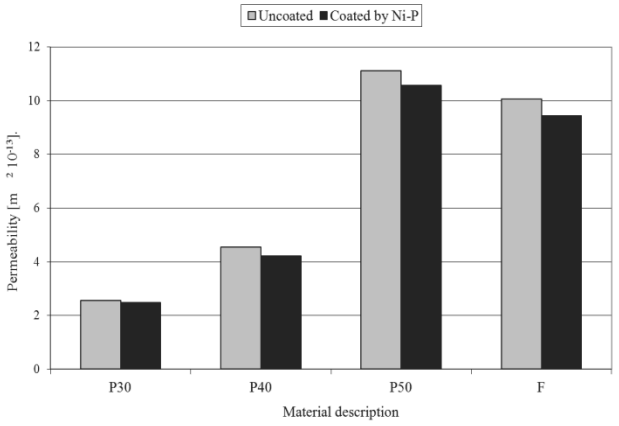


Fig. 6. Results of permeability measurements of ceramic preforms

This situation is caused by the fact that fibers of Al_2O_3 in skeletons are bigger than the particles and hence also the surrounding pores are larger than in the case of manufactured semi-products so they cause lower resistance for the flowing liquid. Figure 7 presents the impact of participation of the ceramic phase on the permeability of skeletons and can be observed its growth with increasing of porosity.

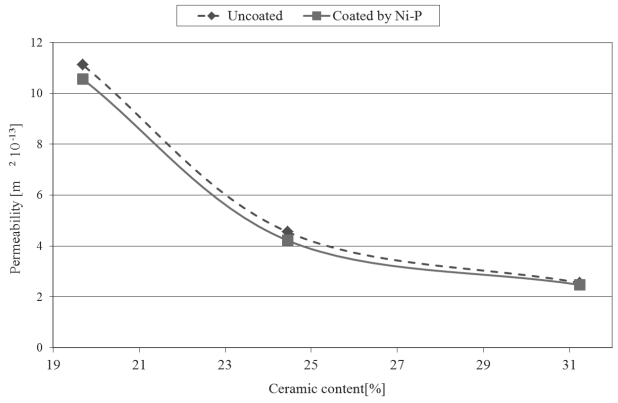


Fig. 7. The porosity influence on permeability of ceramic preforms

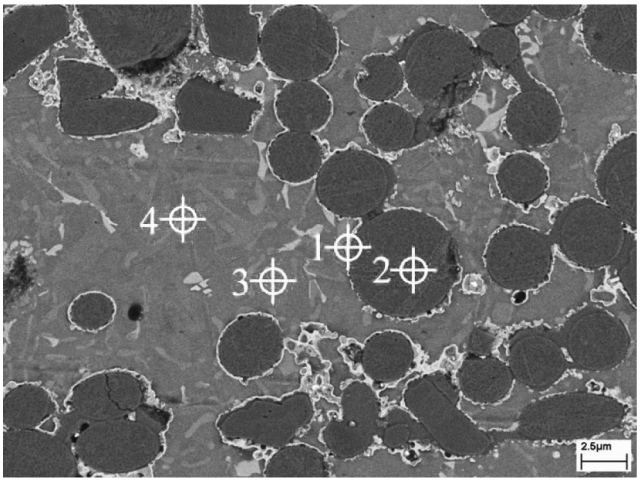


Fig. 8. Structure of composite material F with coated by Ni-P reinforcement

Table 6.
Chemical composition analysis of composite material F with coated by Ni-P reinforcement

Element	Mass concentration of elements, %			
	Point 1	Point 2	Point 3	Point 4
O	28.66	50.36	1.18	0.58
Al	38.18	47.80	81.15	75.70
Si	17.16	1.84	17.67	23.72
Ni	16.00	-	-	-
Sum	100.00	100.00	100.00	100.00

Measuring permeability of porous skeletons were repeated after applying on their inner surfaces of Ni-P alloy designed to improves the wettability of Al_2O_3 by molten aluminium, and improving the connectivity of the metal-ceramics. The results show a decrease in permeability compared to values obtained before that process, amounting to 3.13% respectively for P30, 7.46% for P40, 5.04% for P50 and 6.26% for F. A slight decrease in permeability proves that the coating deposited on the inner surfaces of the skeleton is not too thick, and thus does not cause clogging of pores and channels around the ceramic particles and fibers, which occlusion would lead to the occurrence of voids not saturated by the liquid metal in the final material composite Based on the results of permeability measurements, it was found that the pores and channels are present in the skeletons have open structure (they are connected) and they allow easy penetration of the molten alloy during infiltration, which in turn eliminates the occurrence of adverse mikrovoids (pores not filled with an alloy) in the final composite material.

Quantitative X-ray microanalysis performed using X-ray spectrometer EDS spread (Figs. 8 and 9, Tables 6 and 7) confirmed that the presence of Ni-P layer deposited to improve the wettability of the surface of both skeletons made of particles, and ceramic fibres. This layer is continuous, and its small thickness does not cause clogging of pores around the sintered ceramic particles and fibers. Strengthening both in the form of skeletons formed from particles of ceramic fibers and a phase of Al_2O_3, while the main components of the matrix are the Al and Si.

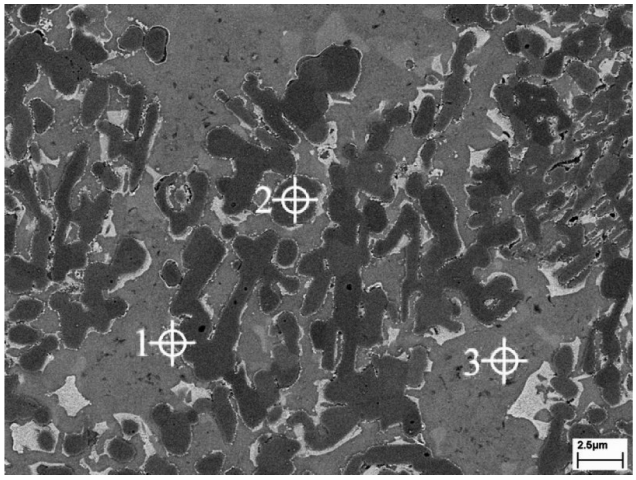


Fig. 9. Structure of composite material P30 with coated by Ni-P reinforcement

Table 7.
Chemical composition analysis of composite material P30 with coated by Ni-P reinforcement

Element	Mass concentration of elements, %		
	Point 1	Point 2	Point 3
O	36.21	47.49	15.02
Al	52.88	52.06	77.72
Si	0.87	0.46	7.26
Ni	10.04	-	-
Sum	100.00	100.00	100.00

4. Conclusions

Properly made porous ceramic skeleton should be characterized by structure of open interconnected pores and channels. Achieving of such structure in obtained sintered skeletons is confirmed by the high permeability values for both the uncoated preform and coated Ni-P coating, which allows easy penetration by liquid metal alloy during infiltration and consequently eliminates the occurrence of pores not filled by the metal that affects at a disadvantage the final properties of composite material.

The occurrence of Ni-P layer deposited to improve the wettability of the surface of the reinforcing phase, both in the form of particles and ceramic fibers was confirmed by quantitative X-ray microanalysis performed using diffuse X-ray spectrometer EDS.

This layer is characterized by a continuous structure, and its thickness does not cause clogging of the pores of sintered ceramic skeletons - which confirmed the permeability test. Furthermore, it was confirmed that a strengthening of the skeleton obtained both from particles and a ceramic fibres is Al_2O_3 phase while the main matrix component are aluminium and silicon.

The obtained results indicate the possibility of wettability improvement of Al_2O_3 preforms infiltrated by liquid aluminium alloy by deposition Ni-P coating and moreover ensures the required structure of final composite material.

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