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Manufacturing of ceramic porous preforms by sintering of Al_2O_3 powder

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ABSTRACT

Purpose: The aim of the study is to develop a method of manufacturing porous preforms based on ceramic powder Al_2O_3 used as the strengthening for the production of modern metal composite materials.

Design/methodology/approach: Semi-products were produced by sintering of ceramic powders with addition of the pores forming agent. The material of the preform was Al_2O_3 powder while as a pores and canals forming agent inside the sintered ceramic skeleton coal and charcoal were used. Particle size measurements of Al_2O_3 powder, charcoal, and coal using laser particle size measurer were made. Preforms were also observed in the scanning electron microscopy (SEM).

Findings: The obtained preforms have a volume fraction of ceramic phase in the range of 20-44% due to the differences of sintering temperature and various portion and coal origin used as pores forming agent.

Research limitations/implications: The main limitation of presented method is the possibility of obtaining preforms where a porosity are not exceeding 80%. Where, in the case of using ceramic fibers, the pores may be more than 90% volume fraction of the material.

Practical implications: Manufactured ceramic preforms are widely used as a reinforcement for production of composite materials by infiltration methods. This method enables the production of metal and locally reinforced composite products with an exact mapping shape.

Originality/value: Results indicate the possibility of obtaining new preforms which are a cheaper alternative to semi-finished products based on ceramic fibers. On the other hand, the use of coal and charcoal as a pores forming agent is an economically justified alternative to previously used materials such as fibers carbon and graphite.

Keywords: Materials; Composites; Ceramic preforms; Alumina

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MATERIALS

1. Introduction

Composite materials are now one of the fastest growing groups of engineering materials. They are more and more widely used practically in industry, primarily in the production of sports equipment, military industry, aerospace, and automotive where special attention should be paid on composite materials with a metal matrix. Metal composites are used where achieved component material border properties, are inadequate. Their biggest advantage is the ability to design properties for specific requirements by varying the volume ratio of the matrix material to the reinforcement. The increasing application of this prospective group of materials is due to the growing availability of technology and consequently, decrease of its costs, so composite materials keep up in economic terms to conventional materials [1-5].

Many modern composite materials did not yet beyond the experimental stage, and to the most interesting trends in their development should be included [6-11]:

- application as a matrix shape memory alloys and ferromagnetic materials, these materials could be used in different phenomena detection systems and giving an impulse to initiate specific functions;
- application of waste materials on the matrix (eg. production of aluminum powders from chips obtained in the machining) or reinforcement of composite materials (eg. fly ashes and slag from power plants).

Today, a development of three main technology of metal composite materials manufacturing is observed. The plastic processing, powder metallurgy and casting method, which is a specific type of pressure infiltration of porous ceramic preforms by liquid metal alloys. This method allows not only to produce homogenous composite parts, but also to locally reinforce products made from conventional materials which further justifies the use of this method in economic terms [5,12,13].

An important element in the manufacturing of composite materials by pressure infiltration are so called "preforms", a porous structures which are a strengthening element.

There are many methods of preparing such structures, such as: sintering of rough, not compacted ceramic powders with or without binders addition; selective laser sintering; production of foam from a ceramic slurry, in which gases are emitted as a result of chemical reaction or decomposition reaction taking place at elevated temperature, method of freezing and sublimation of the solvent from the ceramic suspensions - porous structure is designed by the volume fraction of the solvent in the slurry, rate of freezing and ice sublimation under reduced pressure; sintering of ceramic powders with the addition of pore forming agent [5,12-16].

In the case of the last method, skeletons are formed by uniaxial pressing of the previously prepared mixture of the ceramic powder with pore forming agent or injection molded, where the binder (polymer material) is also a pore forming agent. Moreover, this method is the most flexible, because of the degree of porosity and its character can be regulated by the addition of various pore forming agents (PFA) which decompose at high temperature and form pores.

To the most commonly used pores forming agent can be included materials with thermal decomposition temperature well below the temperature of sintering, such as polyethylene, wax, starch, cellulose, carbon fibres, graphite, etc. In this method, a low is also a risk of undesirable closed or blind pores, because gas products of thermal degradation of pore forming agent emitted from the material form channels through which flow, in the opposite direction, liquid metal during pressure infiltration process [5,12,14,16].

In manufacturing of porous ceramic skeletons being a reinforcement in composite materials, as metal pores forming agent various materials may be used, eg. fine carbon granules, polymer granulate, cellulose, or even sawdust which are waste from the processing of wood. They differ in ability to pores formation and, in particular, in price. In this study it was decided to use widely available and economical materials such as fuel coal and charcoal. It was assumed that the wide-operated carbon materials can be an alternative to costly laboratory-produced preparations [5,13-15].

The aim of this study is to develop technologies for manufacturing porous ceramic skeletons used as strengthening composite materials produced by pressure infiltration. The porous skeletons were produced by sintering a Al_2O_3 powder containing pores forming agent as charcoal and coal.

2. Experimental procedure

Ceramic preforms were manufactured by sintering the Al_2O_3 powder containing agent which forms the structure of pores and channels, in the form of fuel coal and commercial charcoal.

Manufacturing of ceramic preform included: powders preparation, pressing and sintering. Charcoal was

preliminarily pulverized in a mortar and then milled in a ball mill under milling speed of 400 rpm, for 5 minutes, on dry in the zirconia ZrO_2 vessel. The mass to powder ratio of grinding media was 10:1. Coal was pre-screened through a sieve having a mesh size of 0.5 mm and milled in a ball mill. The same milling conditions were used for charcoal but it a satisfactory grinding of the material (below 20 μm) was not obtained, hence the process was repeated.

Laser particle size analysis of pores forming agents and Al_2O_3 powder using device Fritsch Analysette 22 MircoTec plus with adapter Wet Dispersion Unit for measuring the size of particles that are insoluble in aqueous suspension was made.

In order to determine the portion of ashes in pore forming agents their thermal degradation at a temperature of 800°C for 5 hours in air was made

Four variants of mixtures of ceramic powder wit pore forming agent were used:

- 70% Al_2O_3 + 30% coal,
- 70% Al_2O_3 + 30% charcoal,
- 60% Al_2O_3 + 40% coal,
- 60% Al_2O_3 + 40% charcoal.

Then prepared mixtures were wet milled in ball mill Fritsch Pulverisette 6 using the vessel and ZrO_2 grinding media for 10 min. The mass to powder ratio of grinding media was 10:1. To facilitate the subsequent pressing to the mixture 1% polyvinyl alcohol Moviol 18-8 dissolving in water was added. So prepared powder was dried by freezing and sublimation of water under reduced pressure. The mixture was sieved through a sieve No. 0.25. To activate the polyvinyl alcohol powders were wetted by distilled water, packed into plastic bags and left to stand for 24 hours. The powders were subjected to uniaxial pressing on a hydraulic press Fontijne LabEcon 600 in a steel mold with an inner diameter of 30 mm (Fig. 1), under the pressure of 100 MPa for 20 s.

Fig. 1. Steel mold with an inner diameter of 30 mm

The compacts were sintered in the furnace chamber FCF 4/160M /PG Czylok Company in air atmosphere including the following steps:

- slow heating up to the temperature, 800°C at a rate of 50°C/h,
- holding at this temperature for 5 h for total thermal degradation of the coal or charcoal,
- heating to the temperature of 1500°C at a rate of 300°C/h,
- sintering for 2 h,
- cooling of the furnace.

Temperature course of sintering process is shown in Fig. 2

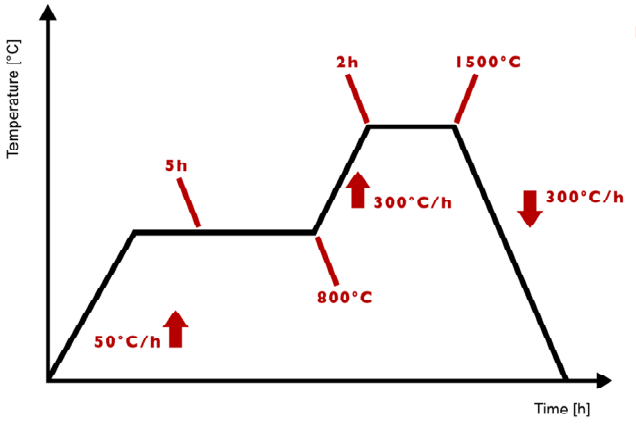

Fig. 2. Temperature course of sintering process

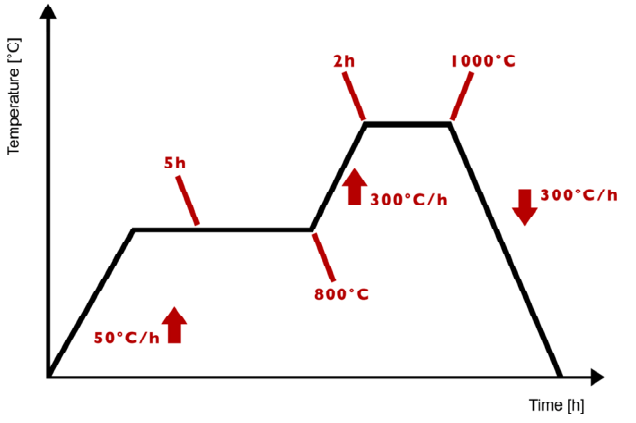

Fig. 3. Temperature course of sintering process in which a coal pore forming agent was used

In the fact that during the thermal degradation of coal, ashes are produced containing approx. 60% of the glassy slag phase consisting mainly of silicon compounds which according literature sources plasticized at a temperature of

approx. 1000°C [17,18]. Also attempted to sinter samples in which coal is used as a pore forming agent at this temperature with the assumption that the glassy phase will be a binder and will be able to provide sufficient strength ceramic skeletons. Temperature course of sintering is shown in Figure 3.

To calculate the volume fraction of the ceramic phase and the porosity, the obtained ceramic preforms were measured and weighed. To examine the density of the powders used in the thermal degradation of coal the calculations was also necessary, using the automatic gas pycnometer Micrometrics Accu Pyc II 1340.

Fractographic examinations of obtained ceramic porous preforms were made on the Zeiss Supra 35 scanning electron microscope.

3. Experimental results and their discussion

On the base of laser particle size measurement of pores forming agent and Al_2O_3 powder heir particle size distribution was determined (Figs 4-8). Table 1 shows the average particle size of the tested powders.

Table 1. Average grain size of Al_2O_3 powder and coal material

Material	Average grain size [μm]
Coal sieved through a sieve of 0.5 mm	121.56
Coal milled 5 min 400 rpm	21.32
Coal milled 10 min 400 rpm	17.29
Charcoal milled 5 min 40 0rpm	8.86
Al_2O_3 powder	8.21

The samples sintered at 1000°C, the temperature suitable for the glassy phase resulted from the degradation of coal, were characterized by low strength properties that dissable their use for further research. Too low mass fraction of the glassy phase, approx. 3%, resulted that it is not a sufficient binder in the obtained porous ceramic skeleton.

Measurements of geometry and weight of preforms manufactured by sintering ceramic powder with pores forming agents as the carbon materials allow the calculation of theoretical content of ceramic phase in a volume of porous material, as presented in Table 2. To the calculation were used, previously measured in an automatic helium picnometer, Al_2O_3 powder density, which was 3,99g/cm^3. Shrinkage of skeletons during sintering was also estimated.

Table 2.
Content of glassy phase and shrinkage of preforms

Composition of sintered powder	Ceramic content, %	Shrinkage, %
70% Al_2O_3 + 30% coal	43.03	22.86
60% Al_2O_3 + 40% coal	43.08	37.92
70% Al_2O_3 + 30% charcoal	27.63	0.15
60% Al_2O_3 + 40% charcoal	19.81	0.34

For samples manufactured with charcoal addition the porosity rises with increases the content of pores forming agent in the sample before the sintering process and in tested samples is about 72-80%. It follows from this that the carbon material degradates leaving in ceramic skeleton only a small amount of ash (5% by weight of the carbon material). During sintering there was no significant shrinkage of the samples.

The porous ceramic skeleton containing coal has a porosity of approx. 57% and does not change, irrespective of the content of carbon material in the sample before sintering. In addition, the preform has demonstrated significant shrinkage during sintering, the value of which rises with increasing initial carbon content in the mixture. This is due to the presence of a glassy phase as a product of the degradation of coal. Al_2O_3 sintering temperature (1500°C) is much higher than the melting point of the glassy phase, hence sintering powders followed by the liquid phase, which is the cause of samples' shrinkagein the process. Additionally, glassy phase may cause undesired closed pores in the final ceramic skeleton.

Observations of fractures of preforms manufactured by sintering Al_2O_3 powders made in scanning electron microscope (Fig. 5) allow to estimate two basic types of pores. The first of them, larger sizes, formed as a result of degradation of carbon materials, while the second, smaller, occur around single ceramic particles and result of a purposeful lack of their compaction.

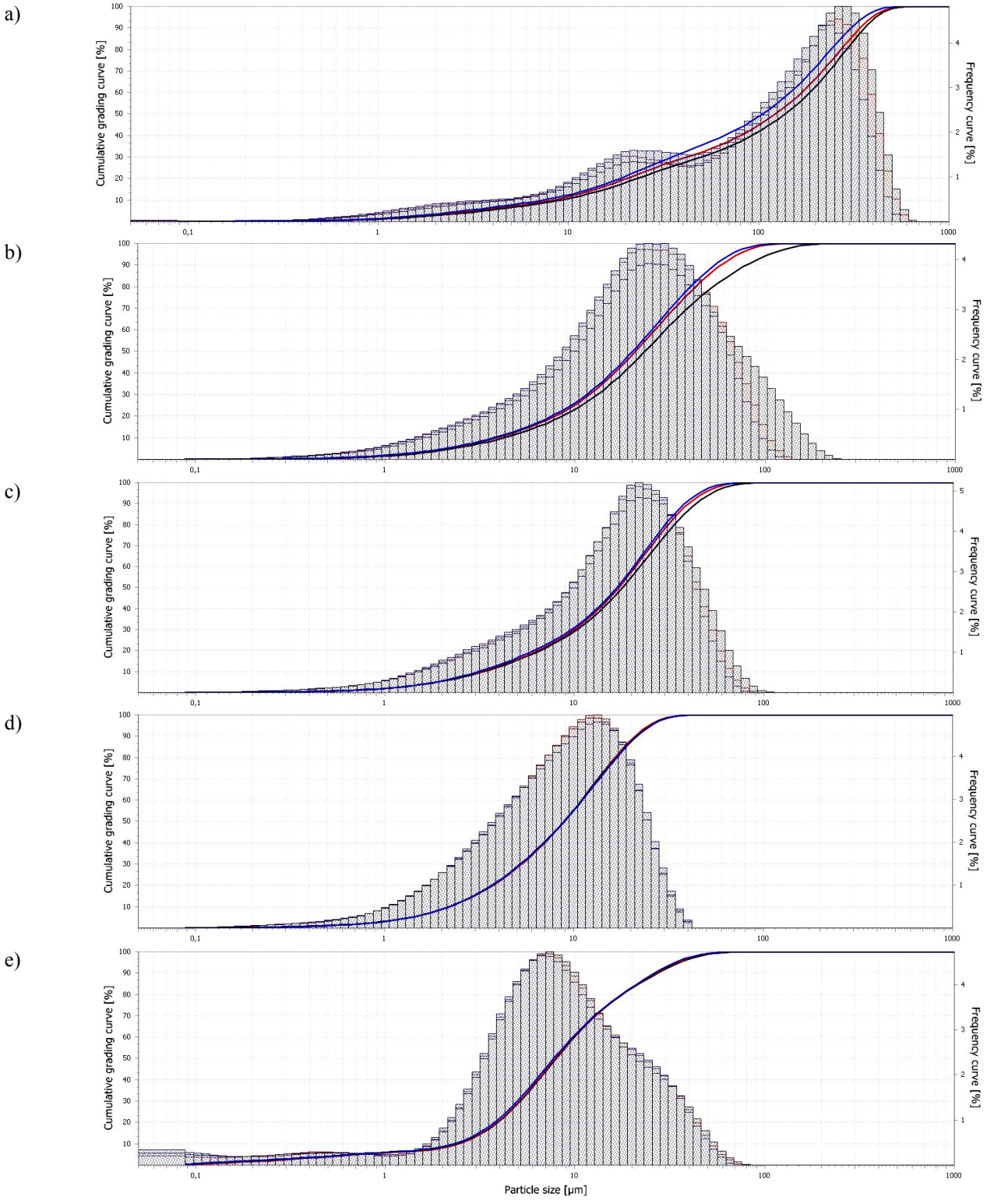

Fig. 4. Particle size distribution of: a) coal sieved through a sieve of 0.5 mm, b) coal milled for 5 min c) coal milled by 10 min, d) charcoal milled for 5 min, e) Al_2O_3 powder in the supply state

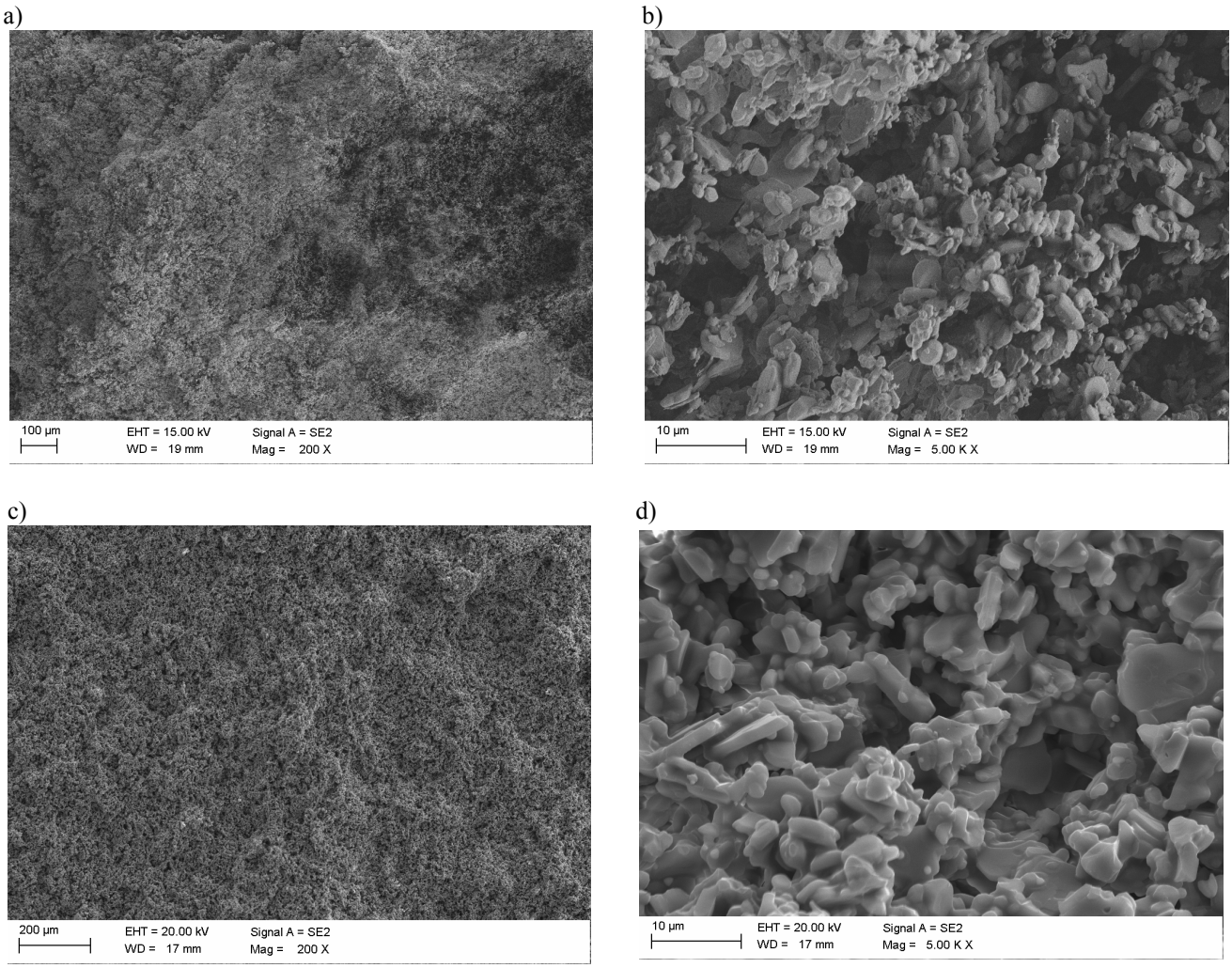

Fig. 5. Microstructure of fractures of ceramic preforms based on Al_2O_3 powder with addition: a), b) 30% charcoal, c), d) 30% coal

4. Conclusions

Carbon materials are very good as pores forming agent, because undergo total thermal degradation. The product of their degradation is mainly CO_2, then a high purity process can be obtained. Furthermore, the use of coal and charcoal as an alternative to laboratory-produced preparates is economically justified.

Charcoal is a better pores forming agent than coal where the product degradation is the ash-containing glassy phase. This phase at the Al_2O_3 sintering temperature (1500°C) is presented in the liquid state causing the high shrinkage of the porous ceramic skeleton.

The measurements of geometry and mass of ceramic materials can be concluded that there is a possibility for manufacturing preforms of a porosity up to 80% volume fraction

Examination of the structure in the scanning electron microscope shown a uniform distribution of the channels created after oxidized carbon materials and the presence of micropores around the ceramic particles.

The technology of sintering porous ceramic skeletons consisting Al_2O_3 powder with the addition of pores forming agents as coal and charcoal allows to produce a skeleton of the desired structure and properties. Researches are being done on developing a composite materials on the base of obtained semi-products.

Additional information

Selected issues related to this paper are planned to be presented at the 22nd Winter International Scientific Conference on Achievements in Mechanical and Materials Engineering Winter-AMME'2015 in the framework of the Bidisciplinary Occasional Scientific Session BOSS'2015 celebrating the 10th anniversary of the foundation of the Association of Computational Materials Science and Surface Engineering and the World Academy of Materials and Manufacturing Engineering and of the foundation of the Worldwide Journal of Achievements in Materials and Manufacturing Engineering.

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