

CONTROL OF GRADED STRUCTURE IN GASARS**STEROWANIE STRUKTURĄ W HIPOTETYCZNYCH
GAZARACH GRADIENTOWYCH**

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Abstract

Development of instruments for micro- and macrostructure design in functionally graded materials is a challenge for the modern industry. In this path, mathematical modeling and numerical simulation are extremely helpful techniques for design and investigations of functionally graded materials. The aim of this paper is to show the possibility for production of gasars with graded porous structure. This is done by means of comprehensive mathematical model of fered by the authors in a previous work. The way to control the structure by means of processing parameters is discussed on the basis numerical experiments related to copper/hydrogen system.

Key words: gasars, highly porous materials, designing, functionally graded materials, mathematical modeling, numerical simulation, copper-hydrogen system, oriented structure

Streszczenie

Rozwój metod projektowania mikro- i projektowanie makrostruktury materiałów określanych mianem funkcjonalnych gradientowo stanowi wyzwanie dla nowoczesnego przemysłu. Modelowanie matematyczne i symulacja numeryczna są technikami niezmiernie pomocnymi w zakresie wyznaczania wytycznych technologicznych dla opracowania i przewidywania właściwości danej klasy materiałów. Celem danej pracy jest analiza możliwości wytwarzania nowego rodzaju materiałów – hipotetycznych gazarów – charakteryzujących się gradientową strukturą porowatą. Opierając się na wynikach poprzednich prace autorów, zaproponowano model matematyczny, umożliwiający sterowanie strukturą mediów porowatych o ukierunkowanej strukturze por. Poczynione założenia przedyskutowano na podstawie numerycznych eksperymentów dotyczących układu miedź – wodór. Stwierdzono, że podstawowymi parametrami technologicznymi procesu wytwarzania gazarów o strukturze gradientowej jest parcjalne ciśnienie gazu, wywierane na ciekły metal przed i w trakcie krzepnięcia. Zmiana ciśnienia prowadzi do zasadniczych zmian w ilości zarodków i por w odniesieniu do jednostki powierzchni. Uzyskiwana porowatość może się wahać w szerokim zakresie 0-40%, co pozwala na wytworzenie materiału o wysokim gradiencie właściwości.

Słowa kluczowe: gazar, materiały wysokoporowate, projektowanie, materiały funkcjonalne gradientowe, modelowanie matematyczne, symulacja numeryczna, układ miedź – wodór, struktura ukierunkowana

Introduction

Functionally graded structures, their characterization, properties and methods for making are new rapidly developing branch in materials science. The idea of Functionally Graded Material (FGM) is introduced in the early 1980s in Japan, where this new material concept has been proposed to increase adhesion and minimize the thermal stresses in metallic–ceramic composites developed for reusable rocket engines [1]. Meanwhile, FGM concepts have triggered world-wide research activities and are applied to metals, ceramics and organic composites to generate improved components with superior physical properties [2-4].

Functionally graded materials are characterized by gradual space changes in their composition, structure and, as a result, in their properties. Usually they are composites (not only metal matrix composites) but graded structures can be obtained also in non-composite materials on the basis of variety of microstructures formed during some kind of material processing. These materials do not contain well-formed sharp boundaries or interfaces between their different regions as in case of conventional composite materials. Because of this, such materials possess good chances reducing mechanical and thermal stress concentration in many structural elements, which can be developed for specific applications. The structure is not simply inhomogeneously but this inhomogeneousness is performed in one direction, typical for entire volume of a material. Development of instruments for micro- and macrostructure design in functionally graded materials is a challenge for the modern industry. In this path, mathematical modeling and numerical simulation are extremely helpful techniques for design and investigations of functionally graded materials, which, in fact, are typical representatives of knowledge-based materials.

The aim of this paper is to show the possibility for production of gasars with graded porous structure. The way to control the structure by means of processing parameters will be discussed.

1. Gas diffusion problem in gasar technology

Gasars are specific class of porous materials [5, 6], which are obtained by unidirectional solidification of gas saturated melt. The gasar structure can be characterized by local porosity, average pore diameter at certain cross-section, pore number per unit area, pore direction and some others. Gasar production technologies offer different instruments for structure control. They are connected with changes of gas pressures during solidification and direction of solidification front moving and its velocity. In fact, all the structure-determining phenomena take place on solid/liquid interface. Most essential are gas diffusion and gas concentration in the melt just ahead of the solidification front. The natural internal interconnections between the phenomena and the formed structure can be seen more clearly through the explicit mathematical model, which is largely discussed in [7]. Here only the gas diffusion problem will be shortly described.

As it is well known [7, 8], for purpose to provide more flexible structure control, mixture of active gas (hydrogen or nitrogen) and argon is used in this technology. Argon

does not dissolve in metal melts but it affects pressure in gas pores and by this reason controls pore size and local ingot porosity.

In the case here considered gas diffusion in the melt appears because of the non-uniform gas distribution formed during solidification and pore growth. The dynamic of gas concentration in liquid, $C=C(x,y,z,t)$, can be determined as solution of 3-D diffusion equation:

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \frac{\partial^2 C}{\partial z^2}\right), \quad t > 0, \quad x \in (0, X_0), \quad y \in (0, Y_0), \quad z \in (Z_{S/L}, Z_0) \quad (1)$$

with initial condition

$$C(x,y,z,0) = C_0 \quad \text{where} \quad C_0 = \eta(T)\sqrt{P_H} \quad (2)$$

and boundary conditions

$$\frac{\partial C(0,y,z,t)}{\partial x} = \frac{\partial C(X_0,y,z,t)}{\partial x} = 0 \quad (3)$$

$$\frac{\partial C(x,0,z,t)}{\partial y} = \frac{\partial C(x,Y_0,z,t)}{\partial y} = 0 \quad (4)$$

$$C(x,y,Z_0,t) = C_0 \quad (5)$$

$$C(x,y,Z_{S/L},t) = C_b \quad \text{for } (x,y) \text{ on pore/melt interface where } C_b = \eta(T)\sqrt{P_b} \quad (6)$$

$$-D \frac{\partial C(x,y,Z_{S/L},t)}{\partial z} = C(x,y,Z_{S/L},t) \cdot (1-k) \cdot v_{cr} \quad \text{for } (x,y) \text{ on solid/melt interface} \quad (7)$$

For simplicity we consider unidirectional cooling which results in planar solidification front. In equations above $Z_{S/L}$ is the coordinate of solidification front, P_H is partial pressure of active gas above the melt, k is distribution coefficient for metal-gas system considered, and v_{cr} is solidification velocity. P_b is the pressure in bubble which is defined as follow:

$$P_b = P_H + P_{Ar} + P_\sigma + P_g \quad (8)$$

Here P_{Ar} is argon partial pressure in gas mixture above the melt, P_g is hydrostatic pressure on solid/melt interface and P_σ is an extra pressure due to the curvature of gas/melt interface [9] and can be expressed by the relation

$$P_\sigma = \frac{2\sigma_{gl} \cdot \cos(\theta_w - \theta_C)}{r_g} \quad (9)$$

Here r_g is pore radius, θ_w and θ_C are contact angle of melt on solid nucleation site and half angle of conical pit, respectively.

The function $\eta(T)$ in (6) expresses the temperature dependence of Sievert's law and looks like this

$$\eta(T) = A \exp\left(-\frac{B}{T}\right) \quad \text{where } A, B \text{ are constant} \quad (10)$$

Let us mention that solidification velocity in v_{cr} (7) is the connection between the thermal and the diffusion problem. This velocity expresses the natural relation between solidification and gas transfer in the melt for the process discussed here. The means for gasar structure control are schematically shown in Fig. 1.

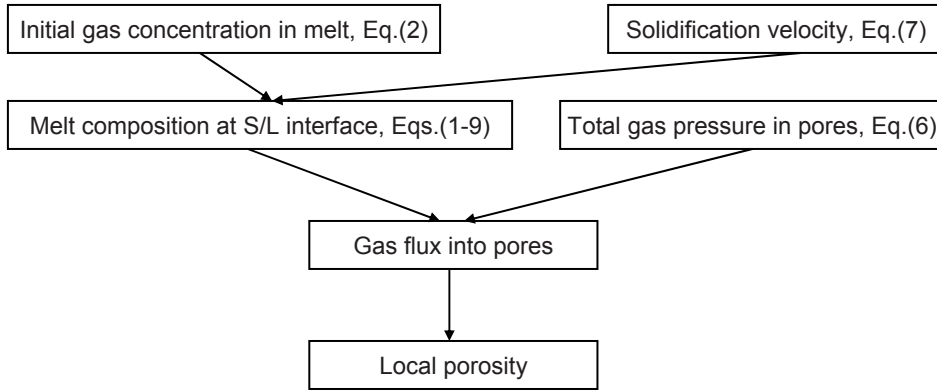


Figure 1. Chain of structure (local porosity) control parameters

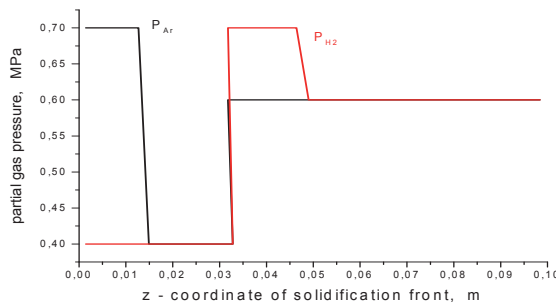
2. Numerical experiments

For purpose to demonstrate the potential for obtaining of gasars of graded structure a number of numerical experiments are carried out. All simulations discussed below are done by the model offered in [7] and are related with copper/hydrogen system. Physical parameters for copper, which are used, can be found in Table 1. Gasar ingots are obtained in a metal mold predominantly cooled at the bottom. The active gas is hydrogen and melt saturation is realized by gas mixture of hydrogen and argon. Initial melt temperature is 1473 K and solidification started after complete melt saturation. Equilibrium gas concentration in melt is determined by relation (2).

Table 1. List of values of some basic copper characteristics used in simulations

Symbol	Quantity	Value	Units
D	Coefficient of diffusion for hydrogen		
	in liquid (average)	$1.7 \cdot 10^{-8}$	m^2/s
	in solid (average)	$0.4 \cdot 10^{-8}$	
λ	Thermal conductivity		
	in liquid (average)	270	W/m^2K
	in solid (average)	350	
ρ	Density		
	liquid	8000	kg/m^3
	solid	8400	
c	Specific heat		
	for liquid (average)	470	$J/kg K$
	for solid (average)	515	
L_v	Latent heat of fusion	205 000	J/kg
σ_{gl}	Surface tension	1.285	J/m^2

First experiment is aimed to demonstrate possibility for obtaining a layered ingot, which consists of three areas of: graded porosity; “zero” porosity; and the last one of constant local porosity. The local porosity here is controlled only by partial gas pressures of hydrogen and argon. The partial pressures in gas mixture above the melt are changed as shown in Fig. 2. The rapid decreasing of partial argon pressure, P_{Ar} ,

Figure 2. Changes of partial Ar and H_2 gas pressure versus position of solidification front

when solidification front passed through $z = 0.015$ m, causes rapid increase in porosity and average pore diameter, Fig. 3a). In the same time pore number per unit area and nuclei number per unit area decrease also rapidly. The reduction of nuclei number per unit area is greater than reduction of pore number per unit area, Fig. 3b). This is because of the gas concentration at solid/melt interface is lower due to increasing of porosity. When solidification front passed through $z = 0.033$ m both partial argon, P_{Ar} , and hydrogen, P_{H_2} , pressure increase rapidly, the local porosity and average pore diameter decrease and reach zero values. The very high gas pressure values in the system cause reduction of gas volume in pores and all pores close. Then entire gas quantity that is rejected on solid/melt interface remains in the melt ahead of solidification front and increases probability for gas pore nucleation. A lot of pores form and start to grow, which can be seen in Fig. 3b). In this case again number of pore nuclei increases quicker than number of pores. This is because some nuclei do not become pores. They simply remind as very small gas bubbles in solid. When solidification front passes through $z = 0.05$ m P_{Ar} decrease from 0.7 MPa to 0.6 MPa. This reduction does not have sensible effect on the structure. The porosity and the average pore diameter slowly increase because the gas volume in the pores increases, Fig. 3a). At constant gas pressures nucleation and pores per unit area become approximately constant at relatively low level, Fig. 3b). The ingot local porosity in the range 0.004 m $< z < 0.015$ m is about 0.09. From $z = 0.015$ up to $z = 0.025$ the local porosity constantly increases up to 0.4 and forms an area of graded porosity. Then, because of the partial pressures increase, the local porosity sharply decrease and a narrow non-porosity zone appeared in the ingot. At values of z greater than 0.06 m the local porosity reached approximately constant value of 0.37. This experiment demonstrates the possibility for production of ingot, which local porosity varies in wide range (in our case from 0 up to 0.4, see Fig. 3a) and can be controlled by the partial gas pressures. This points out that materials of graded porous structure and properties can be successfully produced by this technology.

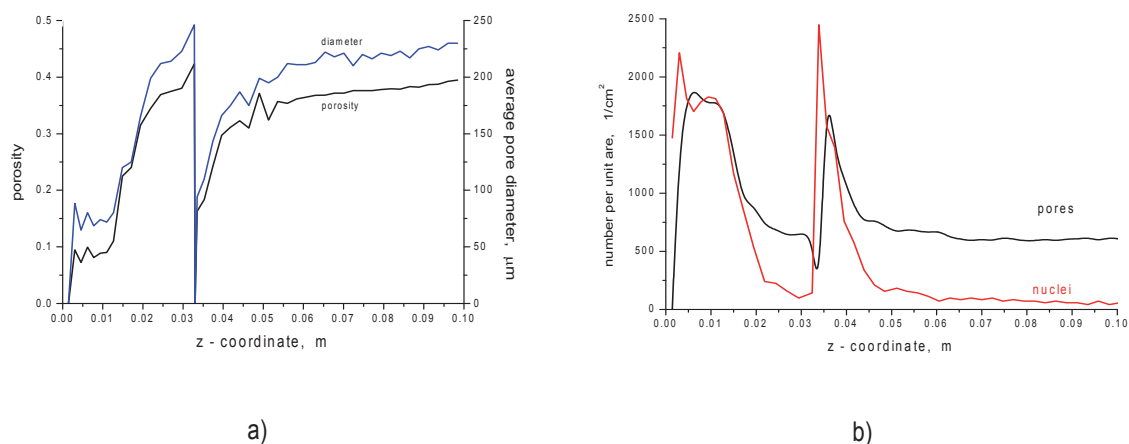


Figure 3. Change in structure caused by changes of partial gas pressures: a) porosity and average pore diameter at different cross-sections; b) pores per unit area and nuclei per unit area

The follow numerical experiment has been carried out to show another way for control of graded porosity in gasar ingot. In this case the partial gas pressures of argon and hydrogen are changed as shown in Fig. 4 a). The changes are chosen to provide graded porous structure in the ingot obtained. By means of partial hydrogen pressure, P_{H_2} , gas concentration in the melt ahead the solidification front can be controlled, and partial argon pressure, P_{Ar} , control pore size and gas flux trough gas/liquid interface. When solidification front moves from $z=0$ m till $z=0.038$ m the total pressure decrease slowly as result of slow reduction in P_{H_2} and P_{Ar} . The local porosity, Fig. 4 b), and the pore number per unit area, Fig 4 d), increase. After this stage, when partial argon pressure increases and partial hydrogen pressure decrease, local porosity becomes zero. High value of argon pressure does not allow formation of porosity. It must be mentioned that in this case average pore diameter does not vary in so large interval, Fig. 4 c). In this case a gasar ingot of graded porosity is also obtained.

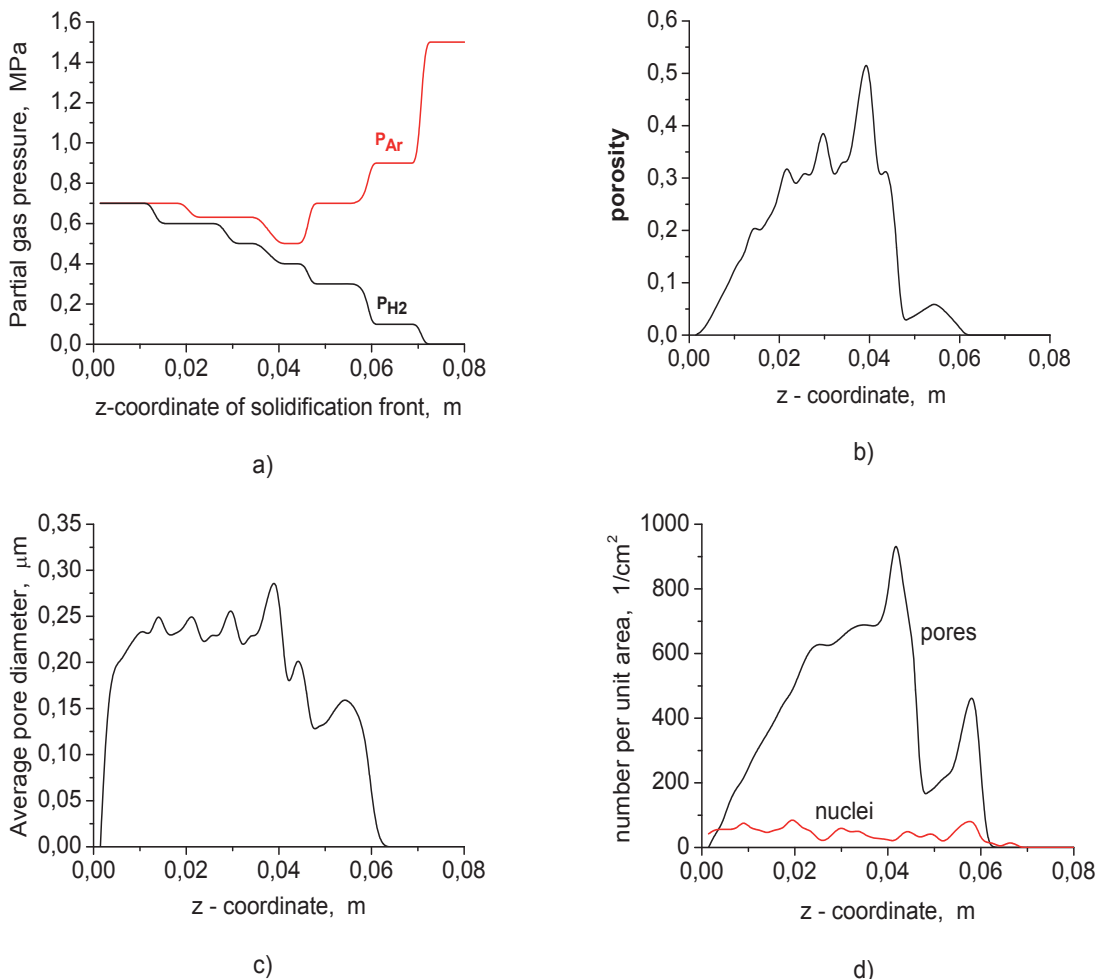


Figure 4. Changes in structure characteristics due to changes of partial gas pressures: a) partial gas pressures versus position of solidification front; b) local porosity; c) average pore diameter as function of vertical coordinate of the gasar ingot; and d) pore number and nuclei number as function of vertical coordinate of the gasar ingot

Conclusion

It can be concluded that partial gas pressures applied on the melt before and during solidification are basic driving parameters for structure control. During ingot solidification the porosity can be managed in a wide range, in our case from 0 up to 0.40 (even max. 0.70 for some metal-gas systems analyzed by authors previously [7]). This allows production of functionally graded materials of specific characteristics. Change in partial gas pressures cause essential changes in nucleation and pore number per unit area. Nucleation is more sensitive to these changes.

The technology considered is a flexible method for production of functionally graded porous materials.

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