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THE STRUCTURAL OPTIMIZATION OF CERAMIC-ORGANIC COMPOSITES

OPTYMALIZACJA STRUKTURALNA KOMPOZYTÓW CERAMICZNO-POLIMEROWYCH

The results of studies concerning of ceramic - polymer composites on the porous ceramic base are presented in the paper. Wide technological possibilities of fabrication of high-porous materials allows for an optimization of the structural parameters of ceramic-polymer composites in order to achieve their maximum mechanical properties. In the case of the transition from the single crack fracture to stochastic cracking, porous morphology, volume porosity, pore and particle size, properties of polymeric filler should be taking into account.

The influence of the porous structure and the porosity of the ceramic matrix on strength and the mechanical behavior of the ceramic-polymer composite is caused by the stress concentration near the pores. Introduced to the pores of the ceramic porous material of polymer leads to improve the strength and damping parameters of the composite obtained.

Keywords: porous ceramics, ceramic-polymer composites, structural optimization

W artykule przedstawiono i przeanalizowano właściwości mechaniczne kompozytów ceramika-polimer o osnowie z ceramicznego tworzywa porowatego. Wykazano, że optymalizacja struktury takich kompozytów stwarza możliwość uzyskania materiałów o bardzo dobrych parametrach mechanicznych. W procesie optymalizowania parametrów takich kompozytów należy wziąć pod uwagę porowatość, wielkość i rozkład wielkości porów oraz właściwości polimeru wprowadzonego do porów ceramicznego tworzywa porowatego. Wykazano że wprowadzenie polimeru do porów ceramicznego tworzywa porowatego prowadzi do wzrostu wytrzymałości mechanicznej oraz właściwości tłumiących takich kompozytów.

1. Introduction

In the previous work [1] the principles for the creation of a new class of materials on the basis of ceramic-polymer composites with high mechanical and damping properties have been developed. The idea of the creation of this class of materials was based on the well-known feature of high-porous materials, which is their tendency to fracture by stochastic cracking. Such fracture type implies multiple cracking of inter-porous bridges and, as a result, nonlinearity of the stress-strain diagrams of these materials.

For the optimization of ceramic-polymer composites, both the influence of ceramic matrix structural parameters (total porosity, pore size distribution, porous space morphology etc.) on the properties and the role of composition and the properties of the organic filler should be studied. The purpose of this work is an analysis of the basic principles for such structural optimization.

In article [2] the model for a construction of a nonlinear stress-strain diagram of high-porous materials at uniaxial tension (Fig. 1) and at uniaxial compression (Fig. 2) has been proposed. A thorough study of the stochastic fracture mechanism of high-porous materials inspired the creation of ceramic-polymer composites, since the organic filler with low Young's modulus forms an efficient barrier against the crack propagation. In this case, the strength and damping parameters of such composites are close to those of the best metallic dampers (e.g. foamed aluminium).

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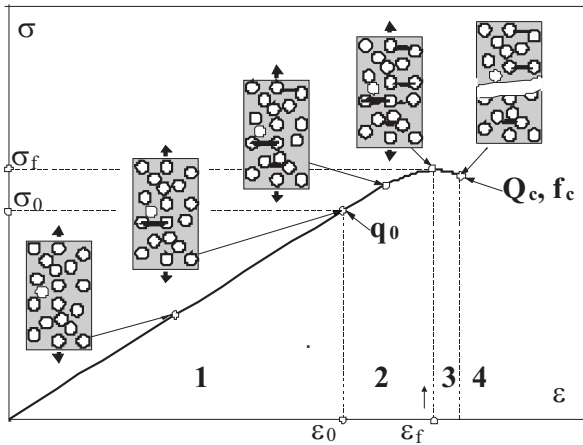
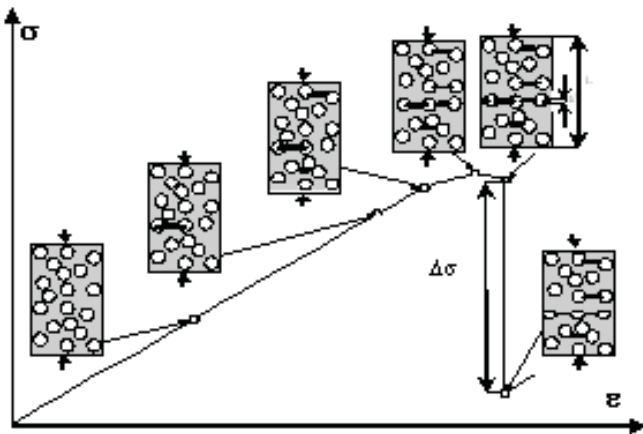Fig. 1. Stress-strain diagram of high-porous materials at uniaxial tension

Fig. 2. Stress-strain diagram of high-porous materials at uniaxial compression

2. Criterion of stochastic cracking

It is known that compact materials (ceramics and brittle metals) fracture by a catastrophic propagation of a single crack. Materials with small porosity also fracture by a propagation of a single crack, while materials with middle and large porosities fracture by stochastic cracking of separate inter-porous bridges.

The calculation of the tension field distribution around the crack was done in the work [3]. The authors of [4] made the formal specification $\sigma_p = f(\theta)$ closer to the experimental data. However, the physical reasons for the observed deviations was not discussed in this work.

It is necessary to take into account the fact that the mechanism of crack propagation in brittle high-porous materials changes with the increase of porosity (see Fig. 3). A brittle crack in a porous material has two alternative ways of propagation: by a linear trajectory, perpendicular to the loading axis, or by a curved trajectory via

the break of interparticle bridges and the confluence of the neighbouring pores.
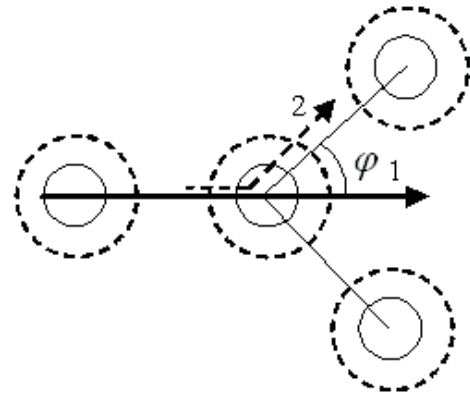

Fig. 3. Changes in crack propagation mechanism with the increase of porosity

TABLE 1

t	1	1.3	1.7	2.2	4
l/d	0	1/3	1	3	5

For the absolute brittle fracture, the calculation of the energy balance should be based taking into account of the total amount of the atomic bonds which will be broken during the crack propagation. Obviously, this amount, and consequently, the total fracture energy, are proportional to the area of the solid phase S_{ck} crossed by the crack during its propagation. In the first case, this area is equal to the specimen area minus the area of pores: $S_{ck} = S_0(1 - \theta)$, in the second case S_{ck} is equal to the area of broken contacts, which, according to [5], is equivalent to the contact section area: $S_{ck} = S_0(E_\theta/E_0)$.

The specific energy of fracture in the first case is $\gamma_{ck} = \gamma_0(1 - \theta)$, where γ_0 is true surface energy. In the second case $\gamma_{ck} = \gamma_0(E_\theta/E_0) \sec\varphi$, where E_θ/E_0 is the normalized Young's modulus, which determines, according to [5], the net cross-section, and φ is an angle of deflection of the crack.

The fracture mechanism changes when these energies are equal:

$$\gamma_0(1 - \theta) = \gamma_0(E_\theta/E_0) \cdot \sec \varphi. \tag{1}$$

According to Bal'shin, $E_\theta/E_0 = (1 - \theta)^2$. Inserting this ratio into Eq. 1 and putting $\varphi = 45°$, we get that the fracture mechanism changes at critical porosity in about $\theta_c = 0.3$. Thus, when porosity is lower than 0.3 the material fractures by single crack propagation and at larger porosity the stochastic cracking takes place.

3. The influence of pore morphology on mechanical properties

The calculations show that the porous structure only slightly influences the critical porosity θ_c. The strength and damping ability are considerably more sensitive to pore morphology. In the model proposed by the authors in [6], these mechanical parameters were related with the change of Young's modulus of the investigated materials. As it follows from the experimental data [7], Young's modulus depends on the porosity and the pore morphology (Fig. 4). In the figure, it can be seen that Young's modulus is different for different classes of materials of the same porosity.
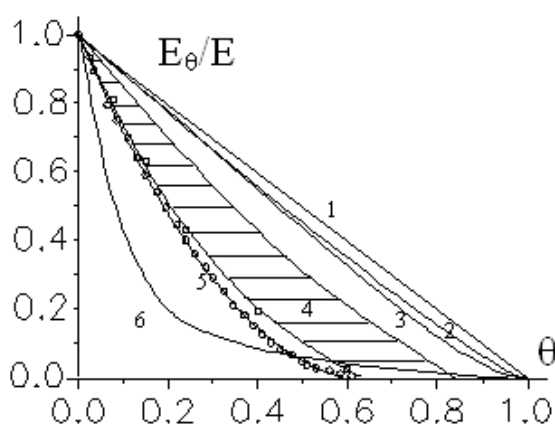

Fig. 4. The dependence of the Young's modulus on porosity for different classes of materials: 1 – gazars, 2 – foamed, 3 – metallic sponge, 4 – bi-porous, 5 – powder, 6 – fiber materials

In the case of a powder material, when a spherical particle is a structural element, according to Skorokhod et al. [8], the forming of open porosity starts at $\theta \approx 0.06$ and at $\theta \approx 0.2$ the porosity is always open. In the case of a material consisting of spherical pores, the transition takes place at the porosity range 0.35-0.50. This factor is fundamental for the creation of ceramic-polymer composites, in which the organic filler is injected into the ceramic matrix by extraction through the open pores.

Influence of particles and pores size

The strength theory of porous materials postulates that the pore and particle size do not influence the relative mechanical characteristics – Young's modulus E_θ/E_0 and yield stress of $\sigma_{T\theta}/\sigma_{T0}$. According to Bal'shin [5], the strength parameters of porous material $\alpha = \sigma_{p\theta}/\sigma_{p0}$, which was described within the frames of contact section concept, is also independent of the absolute sizes of the structure elements. However, this statement is not quite obvious. Indeed, the definition of σ_{p0} as the strength of nonporous material is rather relative, because in the absence of defects its value must be equal to the theoretical strength. Moreover, the strength of brittle material is described by Griffits equation, which contains the absolute size of defect: $\sigma_p = \sqrt{\frac{2E\gamma}{\pi c}}$, where E – Young's modulus, γ – surface energy, c – length of crack (critical defect). This critical defect size is associated with the crack size in the theory of fracture mechanics. In the real porous bodies atomic-sharp cracks are absent; however, the pores whose shape differs from the ideally spherical shape may be a critical defect. In this case, the fracture stresses decrease with the increase of the pore size.

Another important feature of the fracture of brittle materials is the fact that, in contrast to Young's modulus or the yield point, the key role in forming the brittle strength belongs to the maximal size of the structural element, e.g. the grain size for the compact polycrystalline materials or the minimum distance between the pores for porous bodies. The concept of the "weak link" is taken into account in our model of high-porous composite cracking by introducing the notion of local porosity. According to this model, with the application of load, at first the bridges break between the nearest pores, then, as the load increases, the pores located at a greater distance are involved in the cracking process, and so on up to the complete fracture of the material. Since the pores are stochastically located in the crystal, it is possible to assume that the distance between the pores follows some statistical distribution, e.g. long-normal distribution.

For the modeling of nonlinear stress-strain diagram we used the principle of local effective porosity [6]. Taking into account the stochastic distribution of pores in the material for each local volume, the value of effective porosity corresponds to the value of effective Young's modulus [9].

The influence of absolute crack and pores sizes shows up on the cracking stage, when the crack joins two neighbouring pores in the porous ceramic material. At this moment, the near-spherical defect is transformed into a ellipsoidal one. It substantially influences the stress distribution. To take this factor into account we have established the ratio of the pore size d to the total length of such defect l as: $l/d = (0,85/\theta)^{1/3} - 1$, where θ is the porosity. Moreover, because of the cracking condition, the long axis of the ellipsoid is always perpendicular to the loading axis.

The influence of the elliptic pores with random axes orientation on Young's modulus is described in [10]. However, this model works relatively well only for low-porous materials (0 to 0.3).

For the materials with near-maximal porosities (when the material remains continuous) the establishment of the mechanical property dependence on poros-

ity is a very current problem. For the analysis of such systems, the principles of the percolation theory are often applied. This theory is substantiated for conductivity in high-porous powder systems. The main relation of this theory is the connection between the effective property χ and the porosity near the percolation threshold: $\chi = (1 - \theta/\theta_0)^t$.

A similar tendency of the porosity dependence for the electric and mechanical properties allows the use of the percolation theory equations for the analysis of the mechanical properties of high-porous materials [11]. In the general theory of effective medium [12], the elliptic shape of the defect is taken into account by a correction to the percolation index. Using experimental dependences of Young's modulus on porosity for the anisotropic porous systems [9], the existence of a formal connection between the percolation index t and the ellipticity degree of the pores l/d was shown. This information is summarized in the Table. After an application of a corrected model, improved results were obtained (Fig. 5, curve 3).
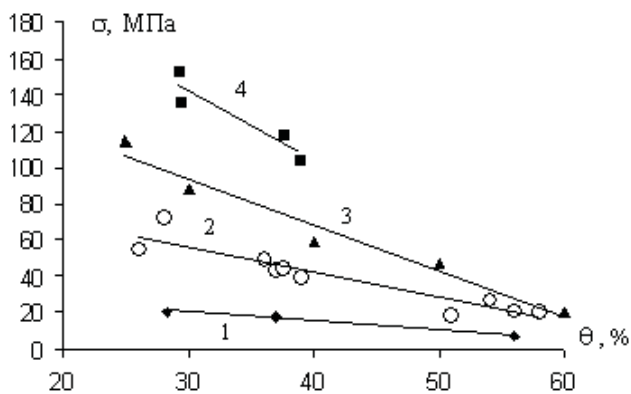

Fig. 5. The influence of porosity on bending fracture stress for porous ceramic and ceram-organic composites: 1, 2, 4 – experiment: 1 – without filler and compact layers, 2 – with narrow compact layers, 4 – with filler, 3 – calculation without filler

Thus, the improved model includes the shape and size of the pores and allows for a description of the stress-strain curve of the porous material more accurately. It should be noted that for the ceramic-polymer composites the size effect is substantially smaller, especially when there is a good adhesion between the ceramic matrix and the organic filler.

The analysis described above has shown that the problem of the structural optimization of ceramic-polymer composites is a multiparametric one. The calculation of the stress-strain curve and the determination of strength and ductility of the composites requires a complex solution with the use of a computer simulation. The example of the computer calculation of such diagrams, presented in Fig. 5, shows that the simulation agrees with the experimental data well.

4. Selection of organic filler

In the presence of an organic filler, the selection of the optimum structure also depends on the quality of the adhesion of the filler to the ceramic matrix. Therefore, for future investigations we will take into account such factors as: strength and modulus of the polymer, strength of the polymer-ceramics boundaries, viscosity of the polymers, the role of the ceramic pore size, medical compatibility.

5. Conclusion

1. Wide technological possibilities of fabrication of high-porous materials allows for an optimization of the structural parameters of ceramic-polymer composites in order to achieve their maximum mechanical properties. In the case of the transition from the single crack fracture to stochastic cracking, porous morphology, volume porosity, pore and particle size, properties of organic filler should be taking into account.
2. The influence of the porous structure and the porosity of the ceramic matrix on strength and the mechanical behavior of the ceramic-polymer composite is caused by the stress concentration near the pores.
3. After cracking, individual defects obtain an ellipsoidal shape. For this case, the dependence of the elastic modulus vs. porosity may be written in percolation terms. Such correction improves the correlation between the theoretical and experimental data.

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