JTTEE5 18:988–995
DOI: 10.1007/s11666-009-9351-0
1059-9630/$19.00 © ASM International

Influence of the Definition of the Representative Volume Element on Effective Thermoelastic Properties of Thermal Barrier Coatings with Random Microstructure

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(Submitted January 29, 2009; in revised form April 17, 2009)

Coatings produced by air plasma spraying (APS) are characterized by a lamellar structure with high porosity, numerous microcracks and present a random microstructure. The process parameters influence not only the porosity but also the form of the microstructure defects. Different methods have been developed and used to investigate the influence of such random microstructures on the effective mechanical properties. Homogenization methods, based on asymptotic expansion of the involved fields, assume the periodicity of the microstructure. For materials with periodic microstructure they predict accurately and cost efficiently their effective properties. But, for non-periodic microstructures like APS thermal barrier coatings (TBC), the geometrical definition of the so-called representative volume element (RVE) plays a crucial role. In fact, these microstructures are approximated by periodic ones, whose characteristics (extension and phase distribution) are obtained by statistical methods. In this work, several approaches were applied to investigate the influence of the RVE position and geometry on the resulting effective properties of the TBC.

Keywords APS coatings, coatings for gas turbine components, hardness and (visco)elastic properties, influence of process parameters, influence of spray parameters, properties of coatings

1. Introduction

Air plasma spraying (APS) is a widely used technology for production of protective coatings against wear, heat and corrosion. The microstructure of APS coatings is defined by the (partially) molten powder particles solidifying on the substrate. Such microstructures are characterized by high porosity and microcracks. The investigation of the polished samples provides the evidence of so-called mesopores with a diameter between 0.5 and 10 μm together with micropores in the order of magnitude of 10-100 nm. These pores are typically connected to some open porous net of finest microcracks starting frequently from a mesopore and having a length between 10 and 100 μm. The characteristics of the microstructure defects (average crack length, pore diameters and shapes, distributions of the defects) as well as porosity are influenced by process parameters of the APS.

Mathematical homogenization was applied to take into account the effects of the micro-heterogeneities of the coating in the structural simulations. Direct simulation would include the representation of the coating microstructure of the whole turbine blade. Instead, the coating material was considered being homogeneous with effective properties taking into account the coating microstructure. It conforms to the common tendency in the field of the simulation of non-homogeneous media. The aim here is to adopt a macroscopic point of view, working within the continuum theory and representing of non-homogeneous material as a homogeneous medium with overall, effective properties to be determined. Experimental determination of these properties itself is quite difficult and can result in a big amount of experimental work to describe the properties dependency on the process parameters.

A big variety of methods was developed in the last decades to predict the effective properties of heterogeneous materials theoretically on the base of their microstructure and the properties of their components. These methods, usually called homogenization methods, simulate the microscopic scale state in the non-homogeneous materials and its influence on the macroscopic behavior. Ideally, a possibility to investigate all kinds of overall material models and their dependency on the geometry and properties of the component phases, on the loading and its history, state on the microscopic scale etc. should be provided. In fact, the homogenization methods tend to

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simplify the original microstructure as well as the behavior of ''homogenized'' material, which can be partially justified by a limited understanding of the behavior of the single microstructure components and their interaction.

Two methods are usually applied for the calculation of effective properties from microstructural descriptions: virtual testing and mathematical homogenization methods (Ref 1, 2). Whereas the first methods provide a straight comparison between simulation and experiment by a direct description of the microstructure, which results usually in an enormous calculation cost, the second methods, based on multiple scale calculations, result in a noteworthy models reduction. Application of these methods for investigation of the coatings obtained by APS is the topic of this article.

2. Investigation Procedures: Fundamentals

Virtual testing is the simplest way to obtain the effective properties of inhomogeneous material. This approach reproduces numerically the experimental methods used for material diagnosis and consists of the following steps:

1. Reproduction of the sample microstructure.
2. Application of the test loading by proper boundary conditions to calculate the stress-strain state, measured during the real test.
3. Evaluation of the properties similarly to the method used for the experimental results.

This method is physically transparent and provides results, usually corresponding well to the experimental ones. At the same time, the accuracy of the calculation with this method strongly depends on the accuracy of the microstructure representation. The calculation of big samples with a complex microstructure results in enormous calculation cost. Sometimes, researchers take some smaller part of the material and assign the behavior of this part to the whole material. The predicted properties, obtained such way, are extremely sensitive to the choice of the representative volume element (RVE) of the microstructure.

The *mathematical homogenization methods* (or simply homogenization methods) (Ref 1, 2) also suppose statistical homogeneity of the material—the existence of some smaller statistically representative volume containing a very large (mathematically infinite) set of microstructure elements but having smaller dimensions than the original sample. However, these methods go further and investigate the asymptotic behavior of media with such RVEs. Moreover, the definition of the RVE becomes the first of three steps of these methods:

1. Definition of the RVE;
2. Definition of macroscopic quantities starting from microscopic ones through an averaging process over the defined RVE;
3. Derivation of microscopic quantities from macroscopic ones using a localization procedure.

In the case of structure periodicity, common for classic composite materials, the choice of the RVE is equal to the definition of the unit cell, representing the geometrical structure of the material. The definition of the RVE for non-periodical structures entails special procedures, connected mostly to the analysis of the random data. More details will be given in Section 3.

The second step is seen usually as the homogenization itself. It works quasi-simultaneously on two scales corresponding to the two levels of the description. The first one, the macroscopic scale, relates to the whole structure, in which the dimensions of the heterogeneities are very small. The microscopic scale, relative to the single RVE, is the scale of the heterogeneities. Here, the dependence of the macroscopic material characteristics on the constitutive laws of the material components is established. Two common approaches are usually pursued: the asymptotic theory like firstly showed in Ref 1, 2 and the mean field theory (Ref 3).

The localization methods of the third step are preconcerted by the methods of the second step, where the dependency of the macro on the micro fields is investigated. The localization problem can be understood as the inverse dependency and will not be considered here.

In this paper the asymptotic homogenization approach for periodic structures (Ref 1, 2, 4-8) was applied. Here, the macroscopic solution is treated as the limit of microscopic solutions of the family of differential operators depending on a small, scale ratio parameter ε (see e.g. Ref 9 for a brief overview).

In the case of the calculation of the effective mechanical properties of the elastic materials, the method deals with problems in the form of

$$\sigma^{\varepsilon}_{ij,i} + f_j(x) = \underline{0} \qquad \text{(Eq 1)}$$

$$\sigma^{\varepsilon}_{ij}(x) = C_{ijkl}(x) e_{kl}(\underline{u}^{\varepsilon}) \qquad \text{(Eq 2)}$$

$$e_{kl}(\underline{u}^{\varepsilon}) = \frac{1}{2}\left(u^{\varepsilon}_{k,l} + u^{\varepsilon}_{l,k}\right) \qquad \text{(Eq 3)}$$

where x is the coordinate vector, $\sigma^{\varepsilon}_{ij}, e_{ij}$ are the components of the stress and strain tensors, respectively, u_i are the components of the displacement vector, C^{ε}_{ijkl} are the components of the Hooke matrix. The index ε denotes the dependence on this small parameter. Equation 3, describing the strain-displacement dependence, is considered further as the definition of the function (operator) $\underline{\underline{e}}(\cdot)$, Eq 2 describes the material law (the elasticity model) and Eq 1 represents the static equilibrium of the body. The system (Eq 1)-(Eq 3) is completed by periodicity boundary conditions (BC).

An additional scales separation is done by introducing a ''microscopic'' coordinate y:

$$x = \{z = x;\ y = x/\varepsilon\} \qquad \text{(Eq 4)}$$

The final homogenized problem (see e.g. Ref 4-10 for a detailed description of its derivation) has the same

structure, as the original one and the effective elastic constants are calculated as

$$C^{E}_{ijkh} = \left\langle C_{ijlm}\left[\delta_{lk}\delta_{mh} + e_{lmy}\left(\underline{w}^{kh}\right)\right]\right\rangle \quad \text{(Eq 5)}$$

with

$$\langle F\rangle = \frac{1}{\|Y\|}\int_{Y} F(\underline{x})dY \quad \text{(Eq 6)}$$

$$e_{kly}(\underline{v})\frac{1}{2}\left(\frac{\partial v_k}{\partial y_l} + \frac{\partial v_l}{\partial y_k}\right) \quad \text{(Eq 7)}$$

The unknown microscopic displacements w are the solutions of the following boundary problem on the RVE Y:

$$-\frac{\partial}{\partial y_i}\left[C_{ijlm}(y)e_{lmy}\left(\underline{w}^{kh}\right)\right] = \frac{\partial C_{ijkh}(y)}{\partial y_i} \quad \text{(Eq 8)}$$

$\underline{w}^{kh}$ periodic on ∂Y

Being calculated once, the effective Hooke matrix is used in all macroscopic calculations. They do not depend on the boundary conditions of the original problem (Eq 1)-(Eq 3) and possess the properties of the stiffness matrix known from the continuum mechanics (Ref 1, 2). However, they do depend on the size and geometry of the RVE. Further, this dependence will be considered more in detail.

Another peculiarity that has to be mentioned here is the necessity to describe all the phases of the microstructure by their own mechanical properties. In the case of APS coatings, two phases are present: the ceramic phase, representing the powder material, and the ''void'' phase, representing the defects and pores of the microstructure and consisting of the mixture of air, plasma and process gases. The last phase is therefore gaseous, and some additional assumptions have to be made about the ''void''. These assumptions are only weekly connected to physical phase model and thus described more precisely in Section 5 related to numerical model parameters.

3. Microstructure Investigation: Definition of the RVE Size and Geometry

Usually not the microstructure dependency of the material properties is interesting for the application, but their dependency on the process parameters. The dependency of the microstructure on the process is investigated mostly separately: a set of microstructures or their characteristics in dependence on the process parameters (or different processes used to produce the material) builds the basis for the homogenization procedure.

The influence of process parameters of the APS on the characteristics of the microstructure defects (average crack length, pore diameters and shapes, distributions of the defects) was investigated by analysis of SEM photographs of polished samples forming a basis for the homogenization method (Ref 10). For that, a set of coatings was deposited with a special variation of the process parameters (Ref 11). Then, two aforementioned microstructure phases, the ceramic phase and the ''void'' phase, were separated by a special threshold value in the obtained SEM pictures. Since this value has no physical meaning, more information about its choice is given in Section 5.

The SEM images were used directly to build the FE-mesh of the compound and to define the size and geometry of the RVE. As it was mentioned before, one usually cuts out a part of the microstructure and considers it being the RVE. The dimensions of the RVE have to be chosen much smaller than the dimensions of original microstructure, but still being big enough to contain a sufficient number of microstructure elements (pores, inclusions, grains etc.). Here, the RVE is defined by the characteristic period of the microstructure.

A similar problem is treated in the digital signal processing. There, spectral analysis is applied to find the characteristic periods of stochastic digital signals. This method, adopted for treatment of spatial stochastic functions, was used to determine the dimensions of the characteristic period of the coating microstructure (Ref 11-13). Within the earlier investigations of the APS TBCs (Ref 4-10), the real microstructure of the coating was approximated by an artificial periodic one with a simplified geometry of the microstructure defects. Such simplification of the RVE geometry allowed the representation of the ''critical'' characteristic of the coating—its porosity. Further investigations with more complicated RVE geometry (Ref 10) showed results, better corresponding to the experimentally obtained results.

Alternatively, a cut-out of the original microstructure with the dimensions, defined by spectral analysis, can be adopted as RVE. Such approach is completely reasoned for the case of pure periodic structures, where the definition of the unit cell size allows obtaining the same geometry by cutting out the proper volume from any place in the sample. In the case of microstructures with no obvious periodicity, like the microstructures of APS coatings, it causes dependency of the RVE geometry (and consequently of the predicted coating properties) on the position of the cut-out. To work out this dependency in detail, homogenization calculations on RVE located not only in the sample center but also at positions with maximum and minimum porosity are carried out, leading to the definition of a scatter range of the thermoelastic effective properties as it will be outlined in Section 6.

4. Definition of Model Parameters

In this section, non-physical parameters of the presented investigation method are considered. They are needed to adjust experimental data to the here used mathematical model.

First of all, a threshold value is used to separate two phases of the microstructure in SEM pictures of the coatings must be specified. It is used to convert the obtained greyscale microstructure images, where the phase

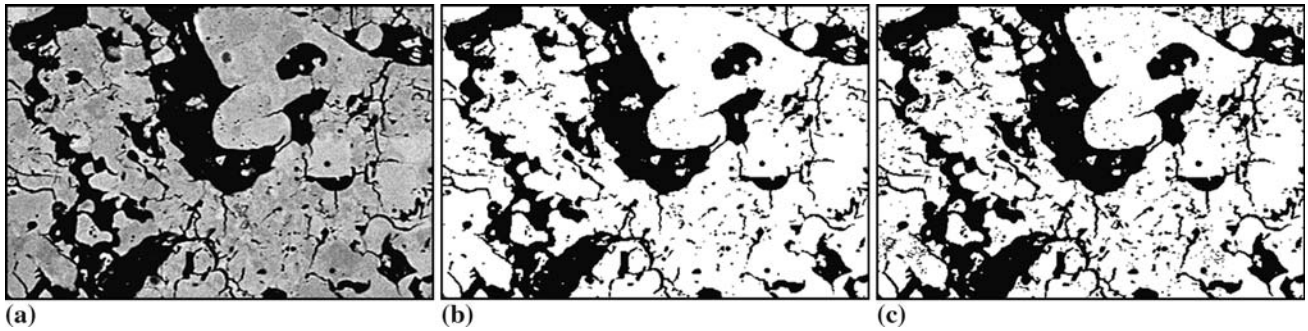

Fig. 1 Original SEM image (a) and its discretizations with thresholds 80 (b) and 120 (c)

Table 1 Variation of the threshold value

Threshold	Porosity	Specific heat, J/kg K	Density, kg/m^3	Young's modulus, GPa
120	0.1702	400.026	5009.79	89.3
80	0.1349	400.020	5222.82	98.7
40	0.1057	400.015	5398.77	97.2

distribution is represented continuously, into a black-white microstructure representation. This value has no physical meaning. As described in Ref 4-8, 10, the digitalization of the images was made on the base of the simple optical images consideration. In the case of the APS coating, when the variation of the threshold value does not influence the most important microstructure characteristic, porosity, significantly (see Fig. 1), such an approach is still warrantable. Additional proof of this was done by calculation of the effective properties for pictures, obtained by different thresholds. Density and specific heat are calculated directly accordingly to the rule of mixture for the whole image from the properties of ceramic ($\rho = 6037$ kg/m^3, $c_p =$ 400 J/kg K) and ''void'' ($\rho = 1.2$ kg/m^3, $c_p = 1050$ J/kg K) materials. The effective Young's moduli were calculated by virtual testing since this method usually gives results corresponding mostly to the experimental ones. Table 1 represents the values calculated for sample 11243 digitalized with the threshold values 120, 80 and 40. Although the difference of the porosity values reaches 20%, the variation of the density does not exceed 4%. The difference of the specific heat values is less than 0.002% and is therefore negligible. The variation of the Young's modulus is less than 6% and still small enough to legitimate the chosen approach of threshold estimation. All further calculations were made with a threshold value of 120.

Next unknown parameter is the Young's modulus of the ''void'' phase, required by the homogenization method for calculation of the effective Young's modulus of the coating. In previous work, when microscopic fields on the RVE were calculated with the commercial FEM solver MSC Marc, the ''voids'' were represented as elastic media with Young's modulus E_{void} much smaller than the Young's modulus of ceramic ($E_{PYSZ} = 2.2 * 10^{11}$ Pa and $E_{void} = 1$ Pa). Such small stiffness value of one part of the RVE can result not only in numerical instability of the microscopic problem (7) but also to erroneous microscopic displacements. For calculations with the software

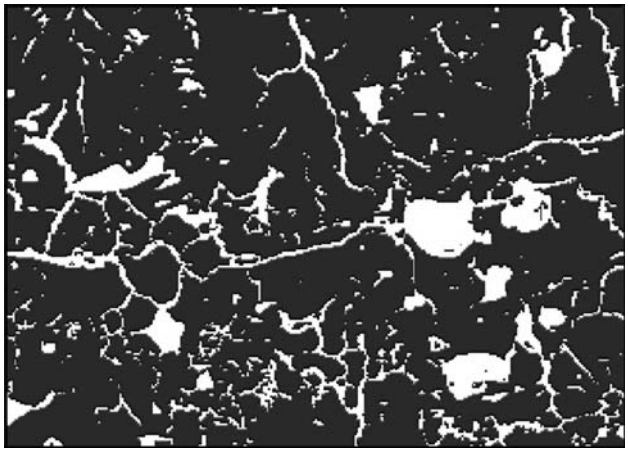

Fig. 2 RVE geometry

HoMat (Ref 12) an additional investigation was performed to find out the minimal acceptable value of the Young's modulus of the ''void'' phase. The RVE of 74.8×42.9 μm^2 with cut-out geometry presented in Fig. 2 was used for this analysis. Experimental data were used for the Young's modulus of ceramics. The results of these calculations are presented in Fig. 3 and 4.

The smoothness (absence of the obvious jumps in the neighbor values) of the functional dependency of the effective Young's modulus on the temperature $E(T)$ was taken as characteristic of the method stability. One can see local fluctuations of the function with magnitude of 1-2% for $E_{void} < 100$ MPa. They increase up to 3% for $E_{void} < 50$ MPa. At the same time, the difference of the effective properties is negligible for $E_{void} < 50$ MPa. Since the last value is much less than the Young's modulus of ceramic (220 GPa), further calculations were made with $E_{void} = 50$ MPa.

5. Effect of the RVE Location on the Effective Young's Module of the APS Coating

According to the investigation procedure described before, the asymptotic homogenization method was

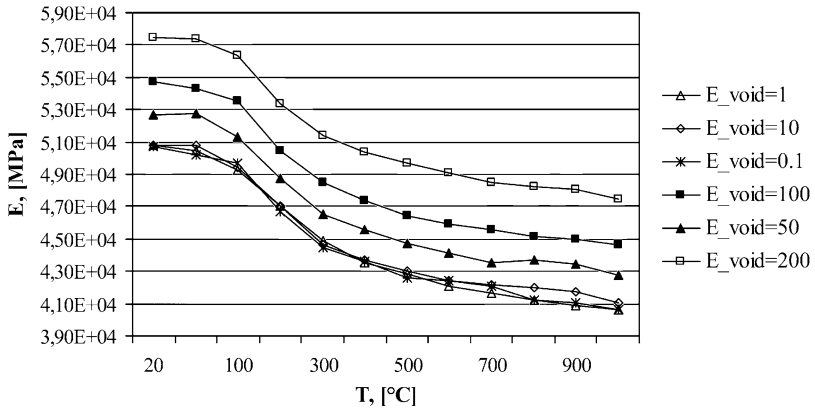

Fig. 3 Dependency of the effective Young's modulus E on the Young modulus of the "void" phase E_{void}

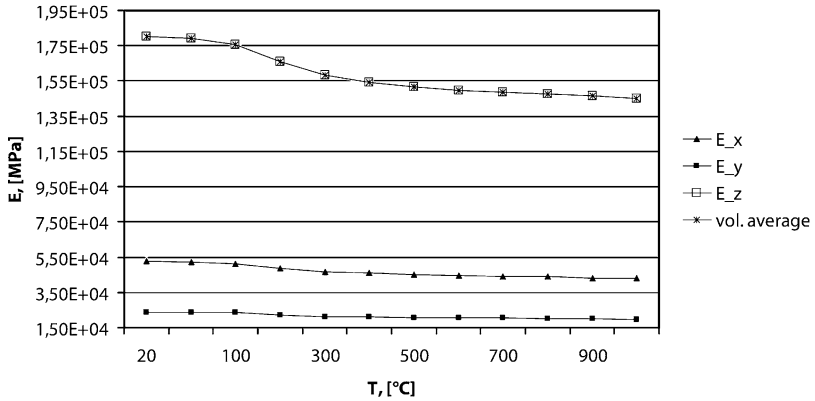

Fig. 4 Effective Young's module E, calculated for different void Young's module E_{void}, compared with volume averaging

applied for a set of coatings made of partially yttria stabilized zirconia ($ZrO_2 + 7\%\ Y_2O_3$), sprayed with an additional variation of some APS process parameters (Ref 11). The description of the process parameters, according to the sample number, can be found in Ref 4. The calculations were carried out assuming elastic behavior of both microstructure phases with Young's moduli of $2.2 * 10^{11}$ Pa for ceramics (PYSZ) and 50 Pa for the "void" phase.

The size of the RVE was defined for every microstructure by the spectral analysis (see Section 3 and Ref 4-10). Firstly, the geometry of the RVE was simplified as shown e.g. in Ref 9. It was shown that variation of the approximated RVE microstructure by assuming more complex defects structure can cause a modification of approximately 20% of the effective thermal conductivity and lead to a smaller gap to the experimental measurements (see Ref 8, 13). At next, cut-outs of the real microstructure were used to define the geometry of the RVE sized to $32.03 \times 64.42\ \mu m^2$. To work out the dependency of the effective Young's modulus, calculated by asymptotic homogenization, on the RVE position, multiple RVEs, distributed along the diagonal of the sample 11283 of $224 \times 128\ \mu m^2$ with porosity of 25.4% were considered here (see Fig. 5). The resulting dependency is outlined in Fig. 6. The RVE porosity varies between 24.4 and 25.5% while the effective Young's modulus of the coating, calculated for these RVEs, varies from 80 GPa up to 136 GPa. It can also be seen that the dependency of the effective Young's modulus on the porosity is not linear, as it is usually predicted by the rule of mixture (volume averaging).

To estimate the complete range of porosity values and the resulting variations of the effective Young's modulus, 50 RVEs sized by $32.03 \times 64.42\ \mu m^2$ were distributed randomly over the sample 11290 (see Fig. 7). Primary, calculations were done using asymptotic homogenization. First of all, the RVEs with maximal and minimal porosities were examined. The cut-out with porosity of 21% gave the effective Young's modulus $E = 126$ GPa, whereas the one with porosity of 25.5% gave $E = 111$ GPa. Figure 8 represents the dependency of the porosity on the coordinates of the cut-out. Here, the coordinates of the left bottom corner are understood as the coordinates of the cut-out. The mean value of the calculated porosity distribution (24.1%) varies slightly from the porosity of the whole sample (24.5%). The standard deviation of the porosities makes 0.018.

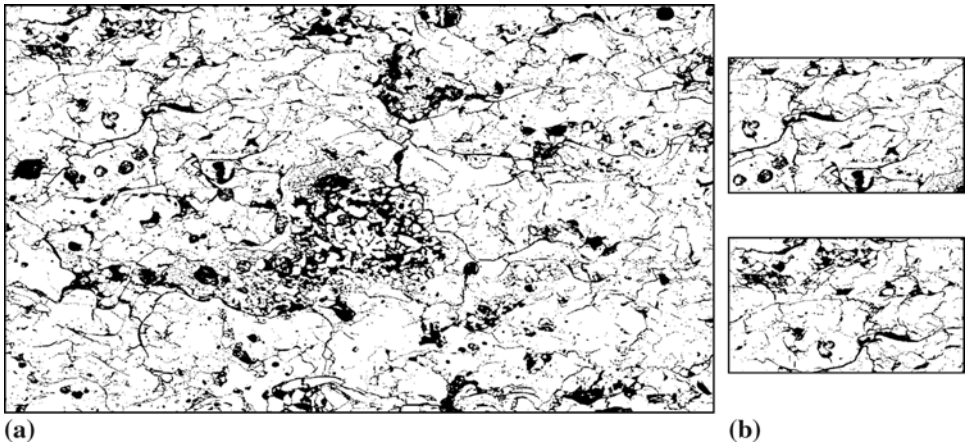

Fig. 5 Original sample (a) and RVE (b)

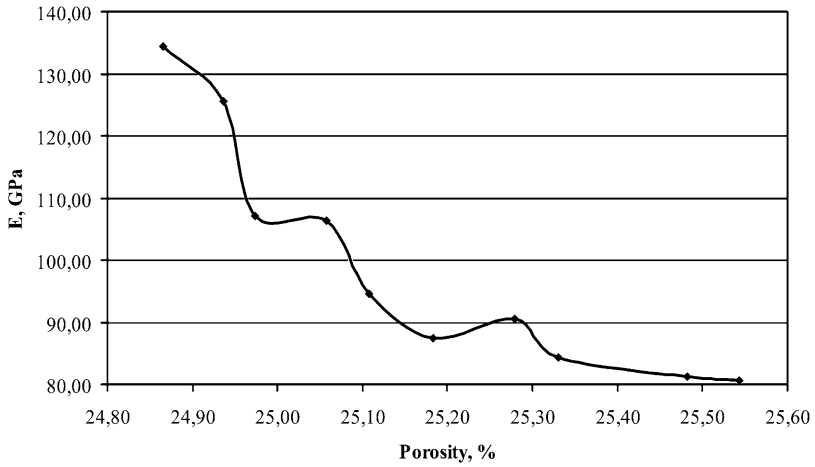

Fig. 6 Dependency of the effective Young's modulus E on the porosity of the RVEs

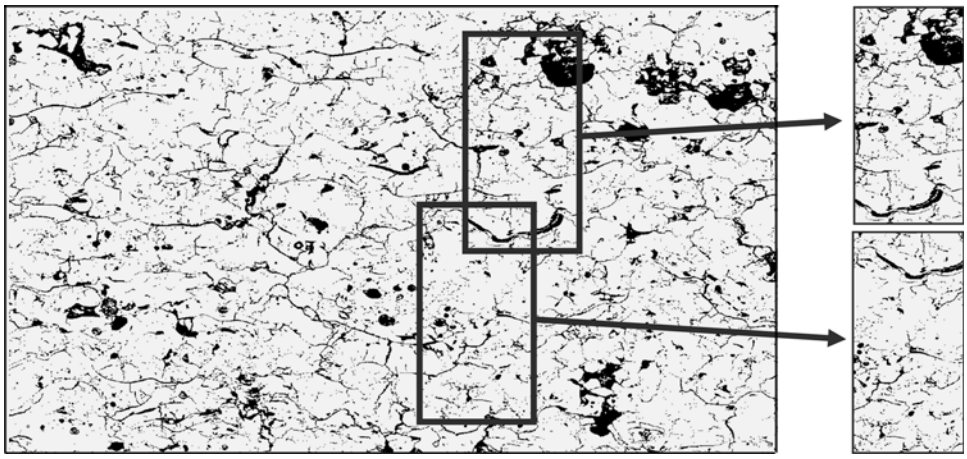

Fig. 7 Original sample and the RVEs with maximal and minimal porosities

The entire dependency of the effective mean Young's modulus, calculated by the asymptotic homogenization method, on the RVE position is represented in Fig. 9. This distribution is characterized by the mean value of 121 GPa and standard derivation of 3.8 GPa (~3% of the mean value). One sees that the character of this dependency

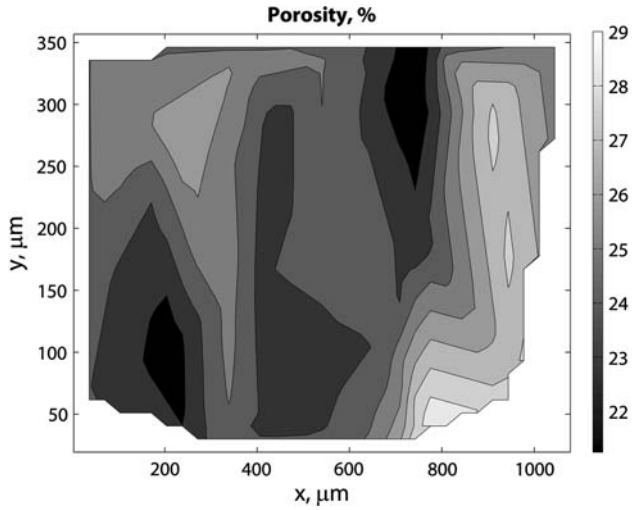

Fig. 8 Porosity distribution over the sample

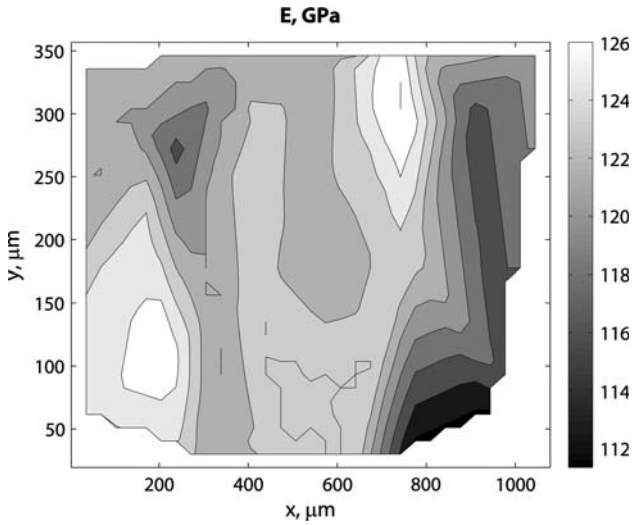

Fig. 9 Distribution of the effective Young's modulus E over the sample (homogenization results)

agrees mostly with the character of the porosity dependency. It means the RVE with the largest porosity produces the lowest effective Young's modulus and vice versa. At the same time, not all RVEs with the same porosity define the same effective Young's modulus. Here, the form of the ''voids'', presented in the RVE, influences the results noticeably.

This influence becomes significant in calculations by virtual testing. The values, obtained in such way, are represented in Fig. 10. One sees here that the values are much lower than the ones calculated by asymptotic homogenization. The mean value of this distribution is 30 GPa and the standard deviation is 16 GPa. So, it is possible to say that the asymptotic homogenization is more robust to the choice of RVE than the virtual testing.

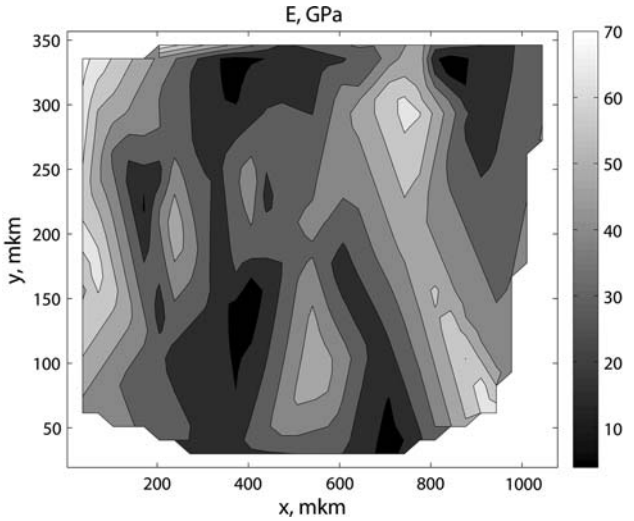

Fig. 10 Distribution of the effective Young's modulus E over the sample (virtual tests)

6. Conclusions

Mathematical homogenization was applied to APS coatings in order to avoid the representation of the whole coating microstructure in FEM simulations, where the influence of the coating microstructure on its elastic properties is investigated.

The available homogenization methods like the asymptotic ones are based on the assumption of the existence of RVE—a smaller statistically representative volume containing a very large (mathematically infinite) set of microstructure elements but having smaller dimensions than the original sample. Application of the virtual testing also frequently uses smaller parts of the original microstructure to reduce the calculation cost. In both cases, the importance of the proper treatment of the input microstructure description and RVE definition cannot be neglected.

The description of the microstructure was separated from the description of the micro-macro coupling, namely from the calculation of the effective material properties on the base of the simulation of the RVE behavior. The microstructure images of the polished coating samples obtained by scanning electron microscopy (SEM) were digitalized. In this step it was shown that this artificially introduced threshold value separating the two phases—ceramics and ''void''—has no significant influence on the characteristics (porosity, density, specific heat) of the obtained microstructure representations.

To define the size of the RVE, the spectral analysis technique was applied as statistical approach. It allows the definition of the most characteristic period of the random coating microstructure per structural direction. Then, the influence of the location of RVE on the digitalized images of the TBC sample was investigated. Both porosity and effective Young's modulus have quite low standard deviations, what confirms the validity of the application of the

spectral analysis for definition of the RVE dimensions. The variation of the effective Young's modulus, calculated for these RVEs by asymptotic homogenization, is about 3%, which enables to take its mean value as ''the'' effective Young's modulus of the structure. Note that Kanit et al. (Ref 14) suggest to adopt a variance of 1% of the effective properties to specify the dimensions of the physical RVE. But we consider here that the obtained 3% variance is accurate enough, so that the dimensions of the RVE, defined by spectral analysis, do not be enlarged. At the same time, the values, calculated by virtual testing, are much more sensitive to the variation of the RVE microstructure. They lead to a large variance of the effective Young modulus which implies to enlarge the RVE dimensions in order to be predictive. This result illustrates that the mathematical homogenization method is more suitable than the virtual testing approach to predict the effective mechanical behavior of APS coatings with random microstructures.

Further development of the computing technology allows the numerical description of the whole microstructure formation process. The material data for individual phases and interfaces, which can be obtained e.g. from ''ab initio'' calculations or molecular dynamics simulations, and the knowledge of microstructures being simulated along the entire process chain will then build a basis for the homogenization method. So, further calculations will use the simulated 3D coating microstructures to calculate their effective properties.

Acknowledgments

The authors would like to thank the German Research Foundation DFG for the support of the depicted research within the Cluster of Excellence ''Integrative Production Technology for High-Wage Countries''.

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