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Utilizing the energy of kinetic friction for the metallization of ceramics

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Abstract. The paper is concerned with the metallization of ceramic materials using the friction-welding method in which the mechanism of the formation of a joint involves the kinetic energy of friction. The friction energy is directly transformed into heat and delivered in a specified amount precisely to the joint being formed between the metallic layer and the substrate material. The paper describes the ceramic metallization process, which has been developed by the present authors based on the friction-welding method.

The stress and temperature fields induced in the joint during the metallization process were determined using the finite element method with the aim to optimize the process parameters. The results were verified experimentally. The structures of the metallic coatings thus obtained were examined and the results are discussed in the paper.

Key words: metallization of ceramics, ceramic-metal joints, FEM, friction welding.

1. Introduction

When using ceramic materials in advanced industrial products it is often required that a metal/ceramic joint should be formed or the surface of the ceramic should be modified [1–7] so as to combine the properties, intrinsically different, of the ceramic and the metal and to obtain a product with the desired properties and functions. The ceramic/metal joints have been made by two methods. In one method the two materials are joined directly, but here the possible pairs of the materials is considerably limited [5, 8–20]. The other method, used more often, consists of producing a thin metallic coating on the surface of the ceramic which can then be relatively easily joined with metals. Both methods give products of high quality but they involve considerable costs and the technological process is much complicated since it requires high temperatures (the entire volume of the ceramic to be metallized is heated) and a protective vacuum or hydrogen atmosphere. Moreover the process is time-consuming. The methods most frequently used in the industrial practice are powder metallurgy, active brazing, PVD, CVD and IPD [17, 21, 22]. Another known method consists of the activation of the ceramic surface by metal ion implantation [1, 7, 14, 20, 23].

The difficulties encountered in joining ceramics with metals are associated with the extreme differences between the properties of these materials, such as:

- lack of wettability of ceramics by metals and lack of interactions between them (the bonds in ceramics are ionic, covalent, and mixed, whereas in metals they are metallic,
- no mutual solubility,
- weak diffusion of metals into ceramics,
- differing crystallographic lattices,
- considerable difference between the melting temperatures, and
- great differences in hardness, brittleness, heat conductivity, and thermal expansion coefficients.

Advanced methods used for joining ceramics with metals are chiefly based on diffusion and usually require high temperatures, expensive additional chemically-active materials, and long time to complete the process.

Most generally, the condition for a directed diffusion stream to be achieved is the existence of a gradient of chemical potential (Gibbs' mol free enthalpy) the occurrence of which depends on the following factors:

- the chemical composition and structure of the material,
- kind and properties of the substrate material,
- ratio of the diameters of the matrix ions and the diffusing ions,
- corpuscular radiation,
- temperature,
- temperature gradient,
- kind and magnitude of the external load,
- deformation degree.

In the friction-driven metallization method proposed in the present paper the last three of the above mentioned factors are very favorable since they have relatively high values and, thus, enable the process time to be shortened.

In all joining processes it is necessary to deliver energy to activate the process. In practice, it is usually thermal energy, which is supplied to the surface atoms so that they can overcome the barrier of activation of the joining process.

In the technology proposed in the present paper the required activation energy is delivered to the system in a me-

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chanical way, utilizing the transformation of the energy of dynamic friction into heat directly in the region of the formation of the joint. There are other literature reports describing the use of mechanical energy for enhancing the joining process [12, 24].

2. Idea underlying the friction-driven metallization

A thin metallic layer was produced on the ceramic surface utilizing the friction force active between the front faces of a titanium cylinder and an Al_2O_3 ceramic cylinder. Compared to other joining methods, the proposed method has advantages in that the joint is formed between materials in the solid state, its efficiency is high, the reproducibility of the joint formation conditions is good, and it permits joining materials with greatly differing properties which often must be joined in the solid state. The method has occasionally also been employed for producing metallic coatings on metals [15].

The variation of the couple of the friction force well illustrates the course of the friction-welding process [18, 25–27]. It depends on several factors such as pressure, rotational speed, the properties of the materials to be joined, and the geometry of the friction surface. At the beginning of the cycle the friction is dry which results in a violent increase of the friction couple. At this stage, we observe the occurrence of first welds (acted upon by shear stresses), and then some particles of the more plastic material are torn out and plastically deformed so that the irregularities of the ceramic surface are leveled. The plastic deformation is accompanied by an increase of the temperature, which stimulates adhesion of the particles. As a result of the adhesion, frictional wear, and internal friction, the material between the surfaces subjected to friction undergoes displacements. The friction couple is increasing until the internal friction acts on the entire surface area of the contact. Local welds, which are formed between the two surfaces being joined are sheared in the plane parallel to the friction plane. When depositing a Ti coating on a ceramic substrate by the friction method the shear plane lies in the near-surface zone of the titanium cylinder front face. In general, in joining two unlike materials with greatly differing R_e, the thorn out particles are transported onto the surface of the harder material [24, 28]. At the sliding stage the friction couple is stable and a layer of high plasticity is formed on the friction surface. This, in consequence, may result in the internal friction passing into external friction. In classical brazing, whose aim is to simply join two parts, this effect is undesired since it leads to the occurrence of quasi-stationary sliding friction when less heat is dissipated on the two surfaces. When the aim is to metallize the surface of a ceramic material, this effect is advantageous and even desirable (Fig. 1), since thanks to it the metallic coating bonded with the ceramic is separated from the solid metallic body. At appropriate values of the load pressure and rotational speed, as the friction time increases a layer of plastic metal is formed on the ceramic surface. With the friction time further increasing, the metallic coating begins to flow out in the radial direction. This is the moment when the process must be stopped and the ceramic component covered with the metallic coating must be separated from the metallic body.

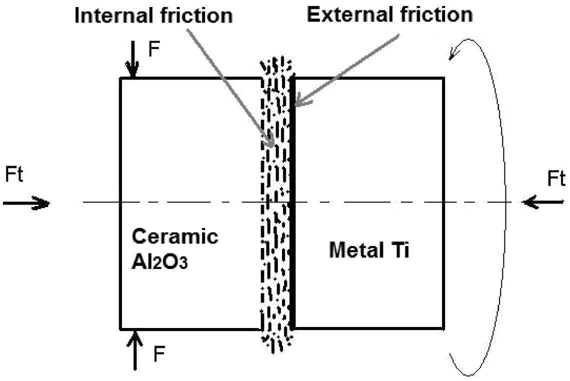

Fig. 1. Simplified schematic representation of the formation of a metallic coating during the transition from internal to external friction

3. Friction-driven metallization of Al_2O_3 with titanium

The experiments were conducted in an apparatus for friction welding (Harms und Wende). The ceramic cylinder was installed in the stationary holder, whereas the titanium cylinder – in a rotational holder. The first series of experiments were devoted to determining the optimum process parameters, which will ensure the formation of a homogeneous Ti coating on the entire surface of the ceramic cylinder front face. In general, the so-called window of the technological parameters is narrowed to the point in the Cartesian space, where the rotational speed, loading pressure, and friction time are the x, y, z coordinates. Table 1 gives the parameters, which in this experiment, ensured the best results of the metallization process.

Table 1
Parameters of the friction-driven metallization of an Al_2O_3 substrate with titanium

Parameter	Parameter value
Rotational speed	2550 [rev/min.]
Pressure on the surface in friction	13.4 [MPa]
Friction time	14 [s]
Ceramic sample diameter	9.5 [mm]
Titanium sample diameter	9.5 [mm]
Length of the samples	20 [mm]

4. Modeling the physical phenomena which occur during the friction-driven metallization of Al_2O_3 with titanium

The temperature, stress, and strain distributions during the friction-driven metallization of ceramic, factors crucial for the formation of a joint between the materials with greatly differing properties such as Al_2O_3 and titanium, were determined and analyzed using the numerical modeling methods. The temperature and stress fields were determined by the finite element method (FEM) with the use of the ADINA THERMAL AND ADINA STRUCTURE (8.6.1. version) software. The numerical simulation was performed based on the conditions, which prevailed during the technological process. In modeling the mechanical fields, we assumed that the contact was between the front faces of the two components being joined, whereas in modeling the temperature fields the assumption was that the temperature of the front faces of both components was the same. The physical and mechanical properties of the two materials were assumed to be dependent on temperature. The modeling results described the thermo-mechanical phenomena that took place during the friction welding process and delivered information necessary for analyzing the mechanism, which governed the formation of the joint.

The surfaces subjected to friction were cylinders 9.5 mm in diameter and 20 mm long, one made of corundum ceramic with 99% of Al_2O_3 and the other - of titanium. The aim of the process was to produce a uniform titanium coating on the front face of the ceramic cylinder. The front faces of the two cylinders were heated by friction under the load $p = 13.4$ MPa. The titanium cylinder was rotated with the speed $n = 2550$ rev/min rubbing the front face of the ceramic cylinder mounted in the immovable holder. The friction coefficient was assumed to be constant (independent of the temperature) and equal to $\mu = 0.46$. The heating time was 14 s.

4.1. FEM thermo-mechanical model. In the numerical modeling of the thermo-mechanical phenomena it was for simplicity assumed that the thermal and mechanical deformation induced during the individual time steps was quasi-stationary. This approach is known in the literature [26–30].

The temperature distributions in the components to be joined were calculated using the equations, which describe the non-stationary heat flow. The boundary conditions adopted in the modeling the heat flow were: the front faces of the cylinders (the interface between the two materials) are loaded with a known heat flux generated by friction and linearly increasing along the cylinder radii, the temperatures of the contact faces of the cylinders are equal, and the side cylindrical surfaces of the two components give out heat to the environment through convection and radiation. The elements used in the FEM analysis were 9-node axisymmetric conductive elements in both the materials, whereas at the edges, they were 3-node axisymmetric boundary convection and boundary radiation elements.

In the mechanical analysis the assumed initial and boundary conditions were: the contact is on the front faces of the two components, axial displacement (z) occurs on the front face of the ceramic component, radial displacement (y) occurs on the axes of both components (condition resulting from the symmetry), pressure acts upon the front face of the Ti component, and the temperature fields are as obtained from the previous solution of the thermal problem in the successive time steps. Figure 2 shows the geometry of the thermo-mechanical model superimposed on the FEM mesh with marked boundary conditions.

4.2. Results of calculation. The calculated distributions of the temperature and stress, factors which stimulate the formation of the ceramic/metal joint during the friction-driven deposition of a Ti coating on an Al_2O_3 substrate, are shown in the form of maps and diagrams plotted for some selected time steps. Figure 3 shows the distribution of the temperature fields determined at the times of (a) 1 s, (b) 5 s, (c) 9 s, (d) 112 s, and (e) 14 s. It is noticeable that the temperature increases with increasing friction time and its maxima during the successive time steps occur in the vicinity of the contact between the front faces of the two components in friction. The maximum calculated temperature achieved during the metallization cycle was 1300°C, which is much below the melting temperature of titanium.

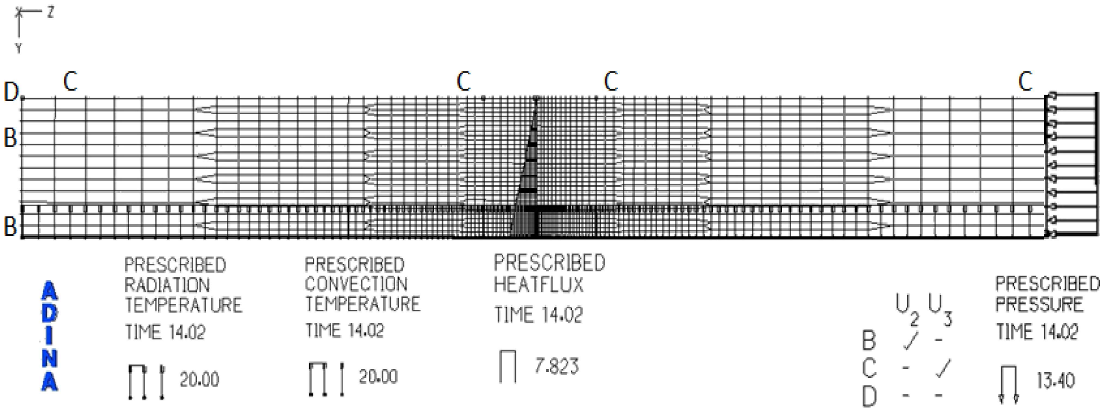

Fig. 2. Geometry of the thermo-mechanical model and the FEM mesh with marked boundary conditions

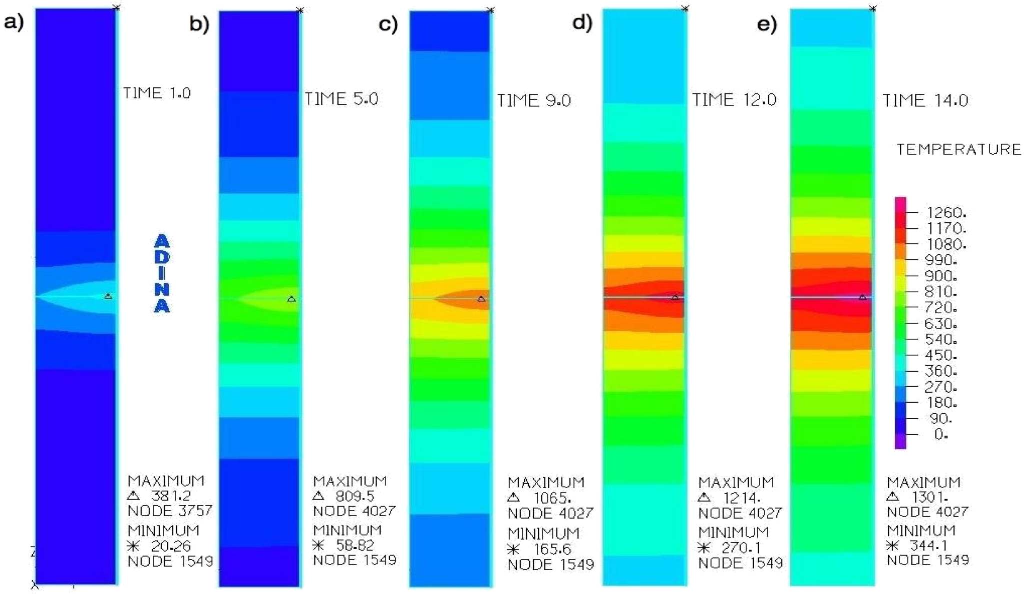

Fig. 3. Temperature distribution during the friction-welding process, calculated for (a) 1 s, (b) 5 s, (c) 9 s, (d) 12 s, and (e) 14 s of the process duration

The time-variation of the temperature within the region of the interface between the two materials has an essential influence on the formation of the joint. Figure 4 shows the temperature distribution along the radius of the contact surface, calculated for various time steps.

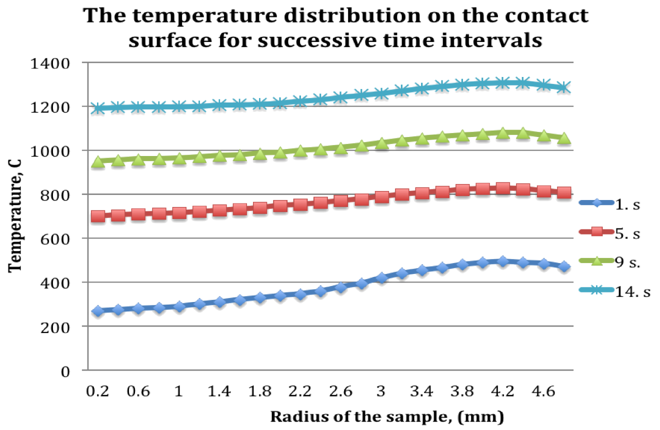

Fig. 4. Temperature distribution on the contact surface during the successive time steps

It can be seen from Fig. 4 that, during the entire process, the temperature of the regions near the axis of the two components is below the maximum temperature by about 100°C. The maximum temperature occurs on the contact surface and in its vicinity at a radial distance of about 4.2 mm from the axis of the cylindrical components.

Besides the temperature distributions, also the stress distributions exert an essential influence on the formation of the joint. In both the ceramic and titanium components, the axial stresses active during the friction process near the contact surface should be negative (compressive) since they stimulate diffusion processes, which promote the formation of the joint. Figure 5 shows the axial stress distributions in titanium during various time steps. As can be seen from these distributions, the axial compressive stresses induced in the region most distant from the component axis, i.e. at a radial distance between 3 and 4.75 mm, are low which is because of the considerable deformation of the titanium cylinder in this region. Figure 6 shows the distribution of the axial, radial, and circumferential stress components active in the Ti cylinder near the contact surface, determined after 1 s and 14 s of the friction heating. The stress components are distributed in an irregular manner, which indicates that the deformation in this region is inhomogeneous. Under favorable thermodynamic conditions, the joint being formed may be based on atomic bonds. In the process described in the present paper, the heating time is relatively short (14 s) and the temperature of the contact surface at the final stage of the process reaches a value between 1200 and 1300°C.

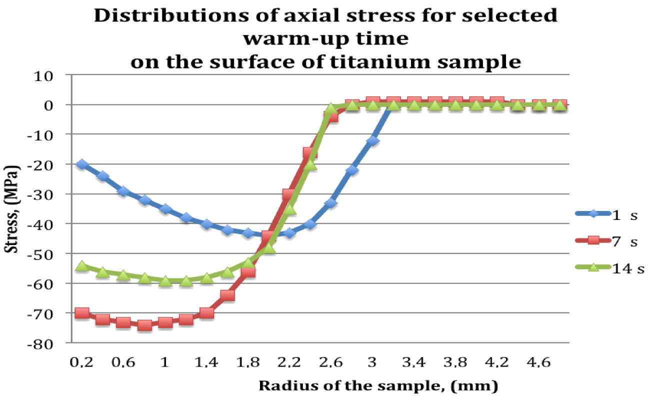

Fig. 5. Axial stress distributions in titanium in the vicinity of the contact surface, calculated at various heating times

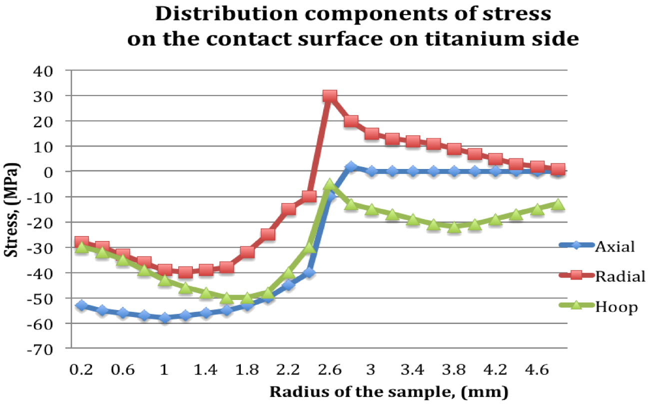

Fig. 6. Distributions of the stress components in the titanium cylinder near the contact surface and the temperature distribution after 14s of heating by friction

The mechanism of the formation of the joint also depends on the character, magnitude, and distribution of stresses induced in its vicinity. The factor deciding about the volumetric changes is the stress axiator. The volumetric changes due to octahedral (average) stresses are given by the equation:

$$\sigma = 1/3(\sigma_1 + \sigma_2 + \sigma_3). \qquad (1)$$

If at a high temperature, the average stress is positive (tensile), the crystalline lattice of the two bodies being joined is stretched which may promote diffusion. The diagram shown in Fig. 7 represents the distributions of the average stresses active along the radius of the front face of the ceramic cylinder at various time moments.

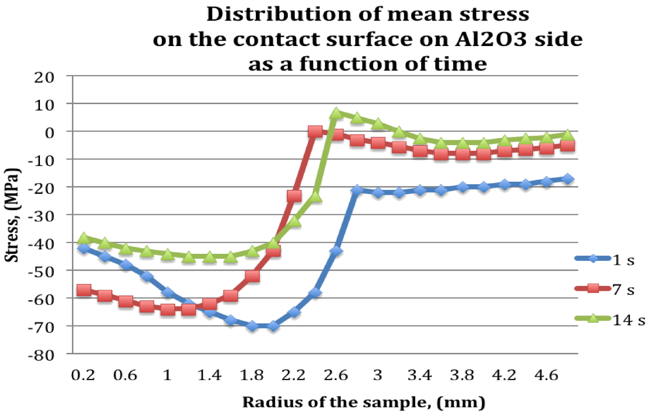

Fig. 7. Distribution of the average stresses in the ceramic near the contact surface, determined during various time steps

5. Some properties of the metallic coating and its joints with the ceramic substrate

An image of the surface of the Ti coating formed on an Al_2O_3 substrate is shown in Fig. 8. The surface has a regular stereometric structure with well-marked directionality, which is due to the directionality of the friction forces active between the two components during the metallization process. The thickness of the Ti coating ranges between 3 and 7 μm. The micrograph shown in Fig. 8 reveals some structural components of the surface layer, namely plastified titanium crystals whose presence is evidence of the high pressure applied to the friction surface and its high temperature.

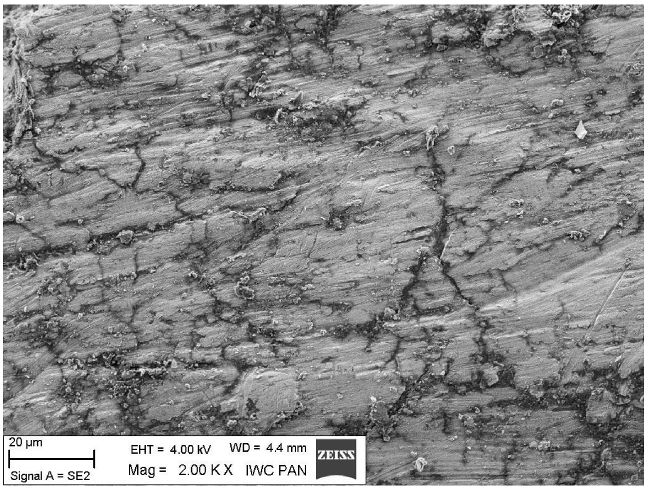

Fig. 8. Surface of Ti coating friction-welded to an Al_2O_3 substrate

Figure 9 is the image of a fracture of a Ti coating/Al_2O_3 substrate system. The fracturing of the sample, which was primarily intended for examination of the fracture surface, was also a severe test of the adherence of the metallic coating to the ceramic substrate.

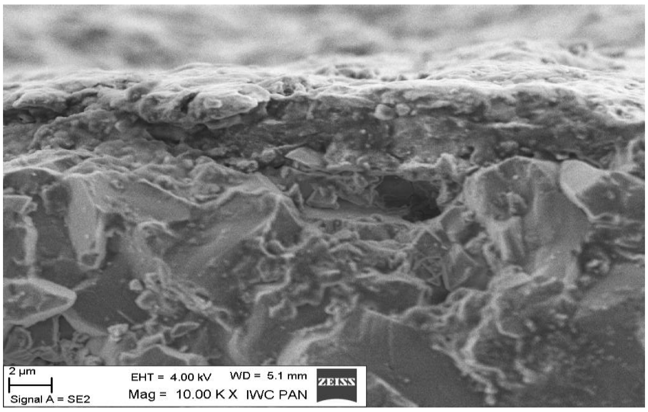

Fig. 9. Fracture of Ti coating/Al_2O_3 substrate system showing the joint region

As can be seen, the coating material is strongly bound with the substrate and fully fills the irregularities of its surface. No chipping was observed in the joining region. Figure 10 shows the surface distributions of oxygen, aluminum, and titanium on a cross-section of the Ti coating/ceramic substrate system. The presence of the Ti coating a few μm thick can be here clearly seen. Figure 11 shows a diffraction image of the Ti coating and of the front face of the Al_2O_3 ceramic cylinder friction-metallized with titanium. The results of the phase analysis indicate that titanium was not oxidized during the process. It is only titanium crystals and aluminum oxide from the substrate, which are observed. There are however no signs of the presence of a transition diffusion layer which would form during the friction welding process.

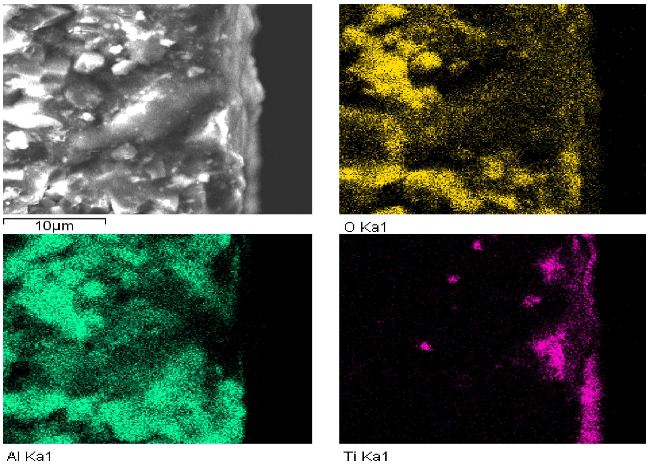

Fig. 10. Surface distributions of oxygen, aluminum, and titanium on a cross-section of Ti coating/Al_2O_3 substrate system

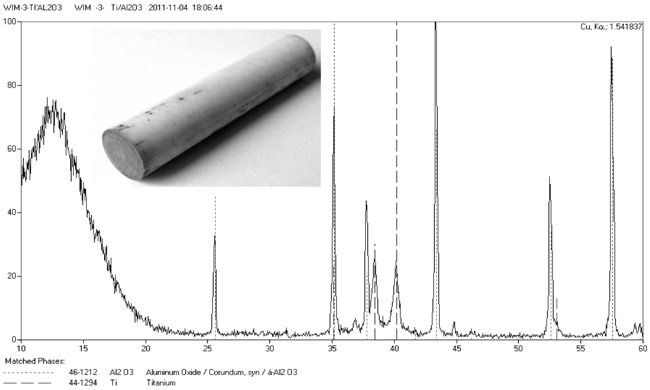

Fig. 11. Diffractogram of the near-surface zone of Ti coating friction-welded to a ceramic substrate

Figure 12 shows the microstructure of the joint between the titanium coating and the ceramic substrate, together with the linear distribution of the elements (Ti, Al, O) on a cross-section of the joint. The Al_2O_3/Ti interface was examined in a scanning electron microscope (SEM) on a cross-section perpendicular to this interface. The figure shows the microstructure of the joint and the linear distributions of the element concentrations. The profiles of the Al and Ti concentrations show no sharp changes, but there are no signs of the presence of the so-called diffusive transition layer.

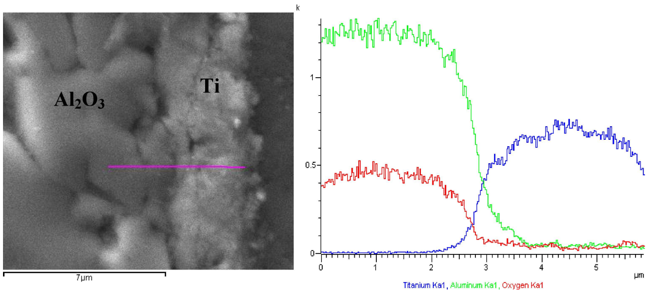

Fig. 12. Microstructure of the Ti coating friction-welded to an Al_2O_3 ceramic substrate and linear distribution of the elements (Ti, Al, O) on a cross-section of the joint

6. Summary and conclusions

Described experiments have shown that the energy delivered to the system in the mechanical way participates very effectively in the metallization of ceramic materials. Thanks to their specificity the mechanical methods permit delivering a specified portion of energy to the precisely defined place in the system where a new interface between the two phases (ceramic/metal joint) is to be formed.

Moreover, the mechanical methods have an additional advantage in that, during the process, various impurities and adsorbed films are removed from the ceramic surface. As a result, its free surface energy is increased which can be regarded as a sort of physical activation that decreases the energy barrier to be overcome in the metallization process.

The numerical simulation permitted determining approximately the temperature and stress fields induced during the metallization process. The calculations were confined to the stage of friction omitting the cooling stage, which has an influence on the generation of residual stresses but does not affect the formation mechanism of the joint.

Summarizing the results of the numerical calculations can be concluded that:

- The temperature distribution on the contact surface is non-uniform. Near the axis of the system the temperature is lower by about 100°C than its maximum value i.e. 1300°C.
- The highest temperature occurs at a radial distance of 4.2 mm from the axis of the two cylinders
- During the entire metallization process, the average stress values (deciding about volumetric deformations) active in the ceramic cylinder near the interface between the two materials are negative

The specific properties of ceramic materials, differing so much from the properties of metals, and the resulting difficulties in joining these two materials still constitute a serious technological problem. Although, thus far, a variety of methods and techniques have been developed for joining ceramics with metals, the possibility of the production in the industrial scale of high-quality and high-performance metal/ceramic joints is still extensively investigated since, in mass production, all the known methods appear to be unsatisfactory and inconvenient.

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