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Dielectric Properties of Plasma Sprayed Silicates Subjected to Additional Annealing

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ABSTRACT

Several silicate materials were plasma sprayed and characterized by the authors in recent years from the point of view of their chemical and phase compositions, microstructure and mechanical as well as thermal properties. The present work is concerned with selected dielectric properties of these deposits. Synthetic mullite and steatite as well as natural olivine-forsterite were plasma sprayed using the water-stabilized plasma system (WSP®). The deposits were striped-out, ground and polished to produce samples in form of plates with a smooth surface. Part of samples was later annealed in air. These samples-after coverage by metal electrodes functioning as monoblock capacitors -were tested in the alternative low voltage electric field to measure capacity and loss factor in the frequency range from 200 Hz to 1 MHz. Relative permittivity was calculated from the measured capacity. Volume resistivity was measured in the direct electric field. It is shown that the relative permittivity of as-plasma sprayed silicates is less stable compared to bulk in the whole studied frequency range. However, thermal annealing modifies the structure much closer to the sintered bulk which is also reflected in dielectric properties. Insulating ability of plasma-sprayed silicates with and without annealing is discussed in consequence with chemical changes and phase changes induced by annealing. Prog. Color Colorants Coat. 10 (2017), 105-114© Institute for Color Science and Technology.

1. Introduction

The family of SiO_2-based ceramics is widely used in electrical industry, especially as insulators. Sintered silicates exhibit excellent volume resistivity and minimal dielectric losses under a wide range of conditions. These two parameters are strongly connected together and they also reflect the structure features such as the crystallinity and grain size [1-3].

In recent times large number of silicates was examined in the Institute of Plasma Physics (IPP) from the viewpoint of the ability to be successfully processed by the water-stabilized plasma system

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(WSP®) [4-6]. Resulting deposits were studied to gain basic characterization of such materials and evaluate all differences in structure and properties induced by plasma spraying process in comparison to conventional furnace processes used in ceramic industry. The group of silicates is wide and important and therefore it is necessary to carry out experiments on many different compositions before successful generalizations could be made. The present work is only a small piece in such a mosaic.

After successful spraying of zircon [4], the IPP group continued to look for potential inexpensive natural silicate materials for spraying. Garnets [5] were successfully applied, but their composition was too complicated and some structural features of deposits were so extraordinary that the authors preferred to continue with more simple chemical compositions in the following works. Synthetic silicates such as wollastonite, mullite, cordierite and steatite were tested and the results were published [6]. After their spraying more detailed study of them started and in the same time, spraying of other silicates was performed. Here, we tried to search on one hand for inexpensive natural, but relatively simple materials and on the other hand for complementary materials with earlier tested synthetic (based on fused feedstock) oxide deposits [7]. This series of experiments include natural olivine-forsterite, synthetic mullite and several SiO_2-based glassy compositions. However, this paper does not focus on the comprehensive study of their plasma spraying.

Subsequently, the authors selected chemical compositions with lower melting points, especially in the MgO-Al_2O_3-SiO_2 system, c.f. the phase diagram in [6-9], which is in present time almost completely covered by spraying activity done at IPP.

In the present work, we are focused on synthetic mullite $3Al_2O_3$-$2SiO_2$ – pure and also in mechanical mixture with 15wt% of glass, steatite $MgSiO_3$ as well as on natural olivine having near-forsterite (Mg_2SiO_4) composition. All these materials are frequently used in electric industry. Mullite is a refractory material undergoing no transformation in solid state [10]. It exhibits perfect dimension stability during thermal cycling [11, 12]. Its use in electrical industry is limited to mechanically supporting parts due to its high loss factor [13]. Steatite is used in high-frequency devices (as coil cores, supporting parts of switches and insulations, in general) thanks to its low loss factor and good mechanical properties [14]. Forsterite has a very high thermal expansion coefficient. It is frequently used in the same way as steatite in electric industry as insulator, especially in vacuum devices where forsterite and its high thermal expansion is utilized in combination with metallic parts. Loss factor of forsterite is low and independent of frequency [14].

The mentioned character of each of these materials in commercial bulk form is obtained thanks to their small grain sizes, low and uniformly distributed closed porosity and moreover thanks to a relatively low glassy phase content. Conventional furnace processes like sintering or manufacturing of glass-ceramics [10, 11] lead to production of the above-described structures. Plasma spray process represents rapid heating and very fast cooling (including massive undercooling) which results in metastable state of the product. The character of the spray process is responsible for specific lamellar character of majority of as-sprayed ceramics. The thermal history of the powder in plasma is responsible for chemical and phase changes (often complicated and not easily manageable by set-up settings change) compared to starting material. Additional annealing is used to ensure crystallization, phase transformations, annihilation of several types of fine defects and to sinter the existing deposits to certain extent. Associating structural changes induced by the annealing with changes in dielectric properties is relatively a new topic and for selected silicates is it a goal of this paper.

2. Experimental procedure

2.1. Raw materials

All synthetic materials were obtained in the form of tablets of industrial purity, produced by the sintering or reactive sintering of previously calcinated powder. Natural forsterite was in the form of raw mineral, received as blocky pieces. All materials were crushed and sieved to obtain feedstock powder for spraying (Table 1). It is necessary to point out that they contain (with exception of natural forsterite) a certain amount of alkali or other metallic impurities due to the previous fabrication of the tablets. In addition, high Fe content in the powders was due to the wearing of steel parts in crushing apparatus.

2.2. Plasma spraying

Mullite and steatite were selected from silicate deposits described in [5] as materials having lowest open and total porosity. Mullite was now sprayed using shorter stand-off distance, SD, compared to earlier experiments [9]. The samples were manufactured using high-throughput water-stabilized plasma spray system WSP® 500 (IPP, Prague, Czech Republic). This system operates at about 160 kW arc power and can process high amounts of material per hour. This system with external anode enables independent adjusting of stand-off distance (SD) and feeding distance (FD). In the presented experiment, feedstock throughputs of 22 to 24 kg/h were used, i.e., about 50% of maximum available throughput. Main spray parameters of this system, i.e. FD and SD, were optimized before deposition and optimum preheating temperature of the substrate was also determined [5, 9]. Stand-off distances tested were in the range of 300 to 550 mm. Metallic coupons were used as substrates and the powder was fed in the plasma plume by compressed air through two injectors. Other parameters are listed in Table 1. Deposited thickness was about 2.5 mm. The deposits were then stripped from the substrate by releasing agent or by thermal cycling at about –100/+100°C to form self-supporting ceramic plates.

2.3. Annealing

Annealing of plasma sprayed deposits was carried out in a laboratory furnace at different temperatures typically for two hours in an air atmosphere. Heating as well as cooling speed was 7 °C/min in all cases. The conditions are summarized in Table 2. The annealing temperature was selected with respect to sintering temperature, which is usually considered to be 0.7 of the melting point [13]. Mullite with glass addition was annealed at higher T_A/T_M ratio because of multicomponent nature of this material and difficulty of estimating the required temperature for inter-diffusion to make considerable changes in the microstructure.

Table 1: Feedstock materials and spraying parameters.

Material	Feedstock size [μm]	FD [mm]	SD [mm]
Mullite	60-100	29	300
Mullite + 15% glass	60-100	28	350
Olivine - forsterite	63-90	31	400
Steatite	70-120	25	350

Table 2: Parameters of annealing of studied silicates.

Material	Melting point T_M [K]	Annealing temperature T_A [K]	T_A / T_M
Mullite	2083	1673	0.80
Mullite + 15% glass	approx. 1873	1673	0.90
Olivine - forsterite	2163	1673	0.77
Steatite	1830	1393	0.76

2.4. Measurements

Both sides of the stripped-off ceramic samples were then grounded to produce smooth plates with parallel surfaces. Such specimens represent in principle monoblock capacitors with dimensions of 10×10×1 mm. A thin layer of aluminium was sputtered subsequently on both sides of the grounded plates at 2×10^{-3} Pa.

Electric measurements were carried out at the CTU in Prague, Faculty of Electrical Engineering. The electric field was applied parallel to the spraying direction (i.e. perpendicular to the substrate surface).

Capacity was measured in the frequency range from 200 Hz to 1 MHz using programmable LCR-meter (PM 6306, Fluke, USA). The frequency step was 100 Hz between 200 and 1000 Hz, 1 kHz between 1 kHz and 10 kHz, 10 kHz between 10 kHz and 100 kHz and 100 kHz between 100 kHz and 1 MHz. Test signal voltage was 1V AC, the stabilized electric LCR-meter was equipped with a micrometric capacitor as recommended in the relevant standard [15]. Relative permittivity ε_r was calculated from measured capacities and specimen dimensions.

LCR-meter (PM 6306) was also used to measure the loss factor tg δ at the same frequencies used for capacity measurment. Electric resistance was measured with a special resistivity adapter – Keithley model 6105. The electric field was applied from a regulated high-voltage source and the values read by a multi-purpose electrometer (617C, Keithley Instruments, USA). The magnitude of the applied voltage was 100±0.05V DC [16]. Volume resistivity was calculated from the measured resistance and specimen dimensions. Measurements are performed for 4 to 5 specimens and the mean values are presented.

Porosity was studied by the optical microscopy on polished cross sections. Micrographs were taken via CCD camera and processed using the image analysis (IA) software (Lucia G, Laboratory Imaging, Czech Rep.). In all cases, 10 images of microstructures, taken from various areas of the cross section for each sample, were analyzed. The over-all chemical composition of powders was measured by X-ray fluorescence (XRF) apparatus LINK XR 200 (Link analytical, UK). Phase analysis was done by X-ray diffraction (XRD) on the diffractometer D 500, Siemens, Germany, using the filtered Cu K_α radiation in the diffraction angle interval from 5 to 90° 2 theta.

3. Results

3.1. Permittivity

Relative permittivity results are shown in Figure 1. In general, plasma deposits have slightly higher permittivity than the bulk ceramics, as can be seen in Table 3. This effect was partly suppressed by the annealing.

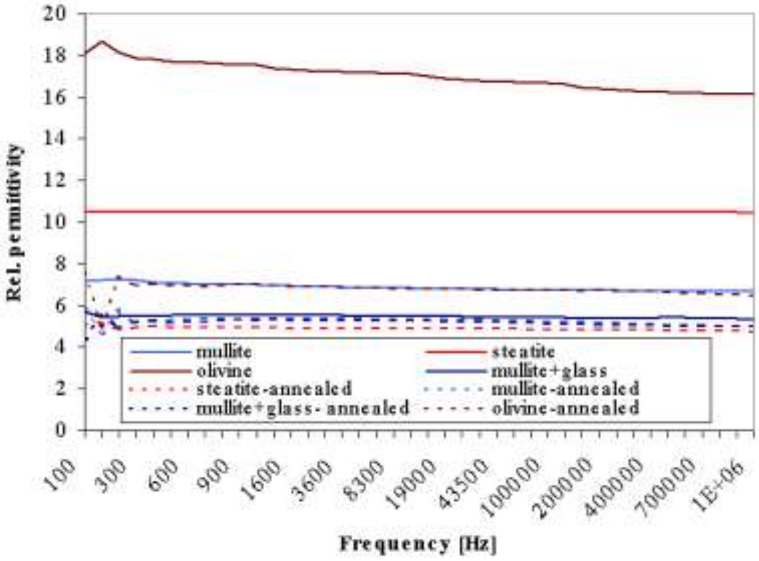

Figure 1: Relative permittivity of as-sprayed and annealed silicates.

3.2. Loss factor

Measured values of the loss factor are summarized in Figure 2 and reference values for bulk ceramics are given in Table 4. Losses in plasma-sprayed materials are in general similar to those of the bulk. Loss factor of plasma deposits exhibits certain decrease with increasing the frequency. This is also typical for bulk ceramics [17], while an increase with frequency is more typical for plasma sprayed dielectrics [18]. Annealed plasma deposits have higher losses with more pronounced decrease in the frequency range of 800 to 8000 Hz, where the values are rather low but not very stable. Above 8 kHz, the losses of annealed deposits start to grow – only slightly for steatite but markedly for all other materials with an extreme for mullite.

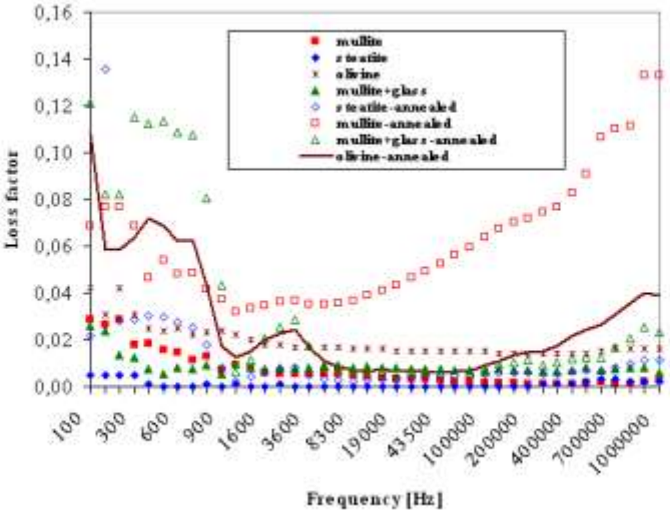

Figure 2: Loss factor of as-sprayed and annealed silicates.

Table 3: Comparison between relative permittivity of plasma deposits and literature values of bulk at frequency 1 MHz Corresponding reference is added in brackets.

Material	PLASMA SPRAYED	BULK	ANNEALED PLASMA DEPOSITS
Mullite	6.7	4 [10]	8.4
Mullite +15% glass	10.2	n.a.	8.4
Olivine - forsterite	14.7	6-8.5 [11]	11.9
Steatite	10.3	6 [10]	8.4

Table 4: Comparison between loss factor of plasma deposits and literature values of bulk at frequency 1 MHz Corresponding reference is added in brackets.

Material	PLASMA SPRAYED	BULK	ANNEALED PLASMA DEPOSITS
Mullite	$2.6x10^{-3}$	$1.6x10^{-2}$ [10]	$2.7x10^{-3}$
Mullite +15% glass	$5x10^{-3}$ at 70 kHz	n.a.	10^{-3}
Olivine - forsterite	$1.6x10^{-2}$	$4x10^{-4}$ [8]	n.a.
Steatite	$8.3x10^{-3}$	$1.5\text{-}2x10^{-3}$ [15]	$4.5x10^{-3}$

3.3. Volume resistivity

The resistivity results together with the porosity are summarized in Table 5. Plasma deposits have resistivity in a very wide range. On one hand, values for mullite and mullite-glass mixture are comparable with bulk material. On the other hand, steatite deposits show values approximately four orders of magnitude lower than that of the bulk. After annealing, the resistivity grows markedly. It is even higher than that of sintered materials.

4. Discussion

4.1. Influence of crystallinity and chemical purity on permittivity

It is well known that first of all the presence and the amount of the amorphous phase controls the relative permittivity of silicates [13]. Annealing is always a very prospective way to obtain crystallized SiO_2-based materials [20-22].

In Figure 1**,** we can see a certain relaxation, i.e. permittivity decrease with increasing the frequency. This corresponds to the fact that in bulk silicates the relaxation is caused by the alkali ions in the amorphous phase, which could shift themselves in the electric field and contribute to the polarization [24]. The higher the amount of amorphous phase in the material, the higher the polarization and therefore the permittivity (for detailed discussion see [24, 25]). We also found that amorphous phase is dominant in the as-sprayed state. Steatite deposits are completely amorphous while mullite is substantially amorphous with traces of crystalline mullite and gamma alumina [5]. Crystalline forsterite feedstock was fully amorphized, only traces of original Mg_2SiO_4 phase (PDF 34-189) remained in the deposit. The amorphization of originally crystalline feedstock during the spraying process [5] is probably associated with very narrow interval between solidus and liquidus in these materials (approx. 40 K at steatite [23]).

Chemical composition investigation by X-ray fluorescence analysis method proved that impurities like K, Ca, Ti and Fe were presented in the feedstock powders. In as-sprayed deposits, their content decreases but certain amounts remain in all samples. Annealed deposits exhibit relative permittivity comparable with the sintered silicates used frequently in electrical and electronic industry. The values are stable as the samples are crystalline (Figure 3 and 4) – similar to bulk silicates. This corresponds to the fact that the two materials with the highest permittivity in as-sprayed state are crystalline after annealing. Steatite is completely crystalline with $MgSiO_3$ enstatite as the main phase with semi-quantitatively estimated content around 70%, and the remaining is silica and small amount of ferroan enstatite (Figure 3). Olivine in annealed state consists of forsterite-fayalite $Mg_{1.8}Fe_{0.2}(SiO_4)$, whereas traces of magnetite Fe_3O_4 and ferrosilite (Fe, Mg)SiO_3 are present (Figure 4). Olivine has the highest permittivity among other materials and the values after annealing are even higher than the bulk. This is probably due to the presence of iron impurities.

Table. 5: Volume resistivity of as-sprayed plasma deposits, annealed plasma deposits and bulk; porosity of plasma deposits. BULK: Values without reference in brackets were measured by the authors.

Material	Resistivity [Ωm] PLASMA SPRAYED	Resistivity [Ωm] BULK	Resistivity [Ωm] ANNEALED	Porosity [%] PLASMA SPRAYED	Porosity [%] ANNEALED
Mullite	$5.34x10^{10}$	10^{10} [10]-10^{11}	$7.18x10^{13}$	7.8	n.a.
Mullite + glass	$2.28x10^{10}$	n.a.	$1.87x10^{14}$	4.2	4.8
Olivine - forsterite	$3.87x10^{9}$	10^{11}	$6.78x10^{11}$	4.4	9.9
Steatite	$8.09x10^{6}$	10^{11}	$2.18x10^{12}$	3.7	7.3

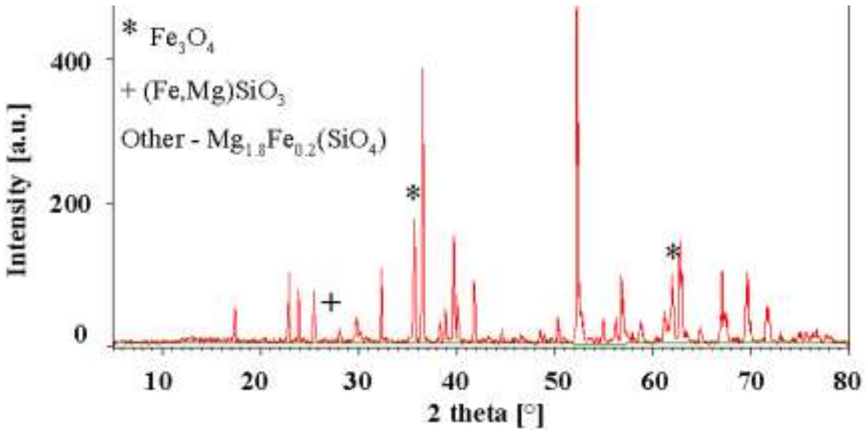

Figure 3: XRD pattern of annealed olivine.

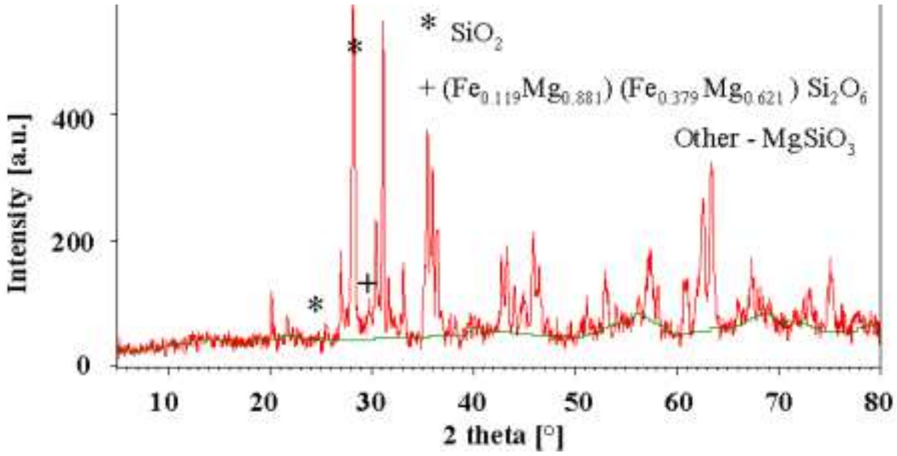

Figure 4: XRD pattern of annealed steatite.

4.2. Loss factor as indication of polarizing mechanisms

Dielectric losses represent the portion of the electric field energy dissipated to heat in the ceramic body. Decrease of loss factor with increasing the frequency is typical for bulk silicates [19]. But the as-sprayed deposits have also similar character of losses. It indicates that the ions shifted by the AC field consume for this movement certain energy. This phenomenon is well pronounced at low frequencies. At higher frequencies, where the fast changes of the field direction enable the shift in limited extent only, the loss factor decreases. The part of the polarization, which is associated with these ions, disappears. At 1 MHz, and above, the polarization of the void-filling medium or other frequency-independent mechanisms (electron polarization) are predominant in deposits, similarly like in [18].

Annealed plasma deposits have higher loss factor compare to as-sprayed ones. Moreover an increase with frequency - to the level of low frequency losses or even more - was observed. Origin of additional boundaries between grains and probably also of some ultra fine microcracks due to crystallization is supposed to be responsible for that effect.

4.3. Volume resistivity-influence of crystallinity on transport of impurities

The resistivity results, Table 5, show that mullite and mullite-glass mixture have excellent resistivity similar to the bulk material. In the case of forsterite, there is certain difference between plasma sprayed and bulk ceramics and plasma sprayed steatite has insufficient resistivity. The authors suppose that in steatite, ionic conductivity via the transport of the impurities must be activated. Steatite deposits and more or less the others, Figure 5, exhibit a particular microstructure without any splats, flat pores and cracks. Such a structure represents 'barrier-free' environment for transport of impurities. Annealed deposits have very high resistivity. The mobility of impurities was diminished by the crystallization and origin of new grain boundaries that caused improvements in insulating ability of the materials.

4.4. Structure and porosity

The same microstructure as described in [5], i.e. lack of "lamellar structure" attributes were found in our samples as well. In the case of steatite, rather bulk-like structure with few boundaries and spherical pores was found in deposits and only partial appearance of rapidly solidified lamellas (lamellas are typical for plasma sprayed coatings) was observed in other coatings (Figure 5a, c, e). After annealing, some changes are visible. In the case of mullite with glass, which forms a composite structure, certain dissolving of the heterogeneous areas is visible. Effect of new very fine globular pores is probably less pronounced compared to pervious features.

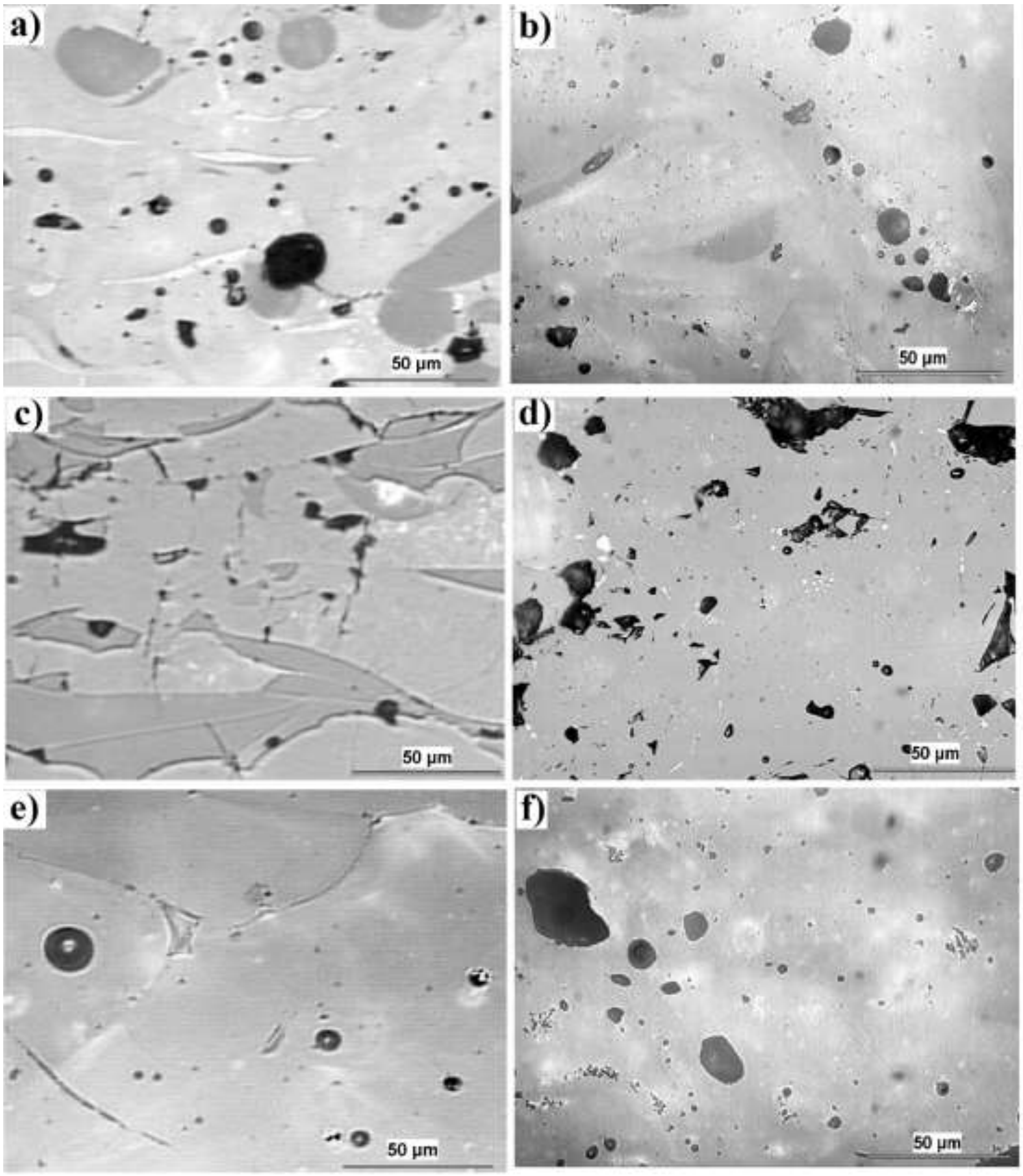

Figure 5: As-sprayed (a, c, e) and annealed (b, d, f) microstructure of mullite with glass, forsterite and steatite; light microscopy.

Similar results were obtained for the mullite deposits, whose images are not given here. In the case of forsterite, certain amount of new pores with rather sharp edges and large size were found. The pores are also larger than those in the as-sprayed microstructure. The considerable porosity increase (Table 5) is mainly attributed to this type of pores. Their feature is probably associated with the high thermal expansion of the forsterite. In the case of steatite, certain amount of new - but fine and globular - pores is responsible for porosity increase (Figure 5 and Table 5).

5. Conclusions

Quality of silicates deposited by plasma spraying could be examined from various points of view. The present work is concentrated on their dielectric properties. Plasma deposits exhibit several differences compared to the bulk. Relative permittivity is in general higher than that of the bulk and is more frequency-dependent. Similarly, the loss factor of the deposits is higher and strongly frequency-dependent in majority of studied deposits. Volume resistivity of the majority of studied plasma deposits is lower than that of the bulk counterparts. After annealing, most of dielectric properties typical for bulk silicates are restored. The only exception is the loss factor that remains rather high and unstable. It has been found that the phase composition and the presence of impurities can markedly affect the resulting values while the influence of porosity seems to be less important.

The application potential of the annealed deposits is promising because of their low crystallization temperatures. This fact suggests that annealing of these coatings can be done on special metallic substrates without any damaging of the metals. Insulating ability of plasma sprayed silicates could be markedly improved by annealing while the polarization behavior is still anomalous and further investigation on this topic (variation in annealing speed and delay on maximum temperature) could improve existing understanding of the involved phenomena.

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