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The effect of plasmonic nanoparticles on the electrophysical characteristics of triboelectric layers

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This paper examines the effect of spherical metal nanoparticles embedded in a triboelectric layer on its electrical properties. A hypothesis is proposed regarding the mechanism by which surface plasmon resonance excited in metal inclusion particles influences the charge density on the contacting surfaces of a triboelectric nanogenerator and its operational characteristics. The effective dielectric function of the spherical metal nanoparticles–polydimethylsiloxane composite is determined using the effective medium approximation. The frequency dependences of the effective dielectric function real and imaginary parts of the triboelectric layer are calculated in the frame of the classical and corrected Maxwell-Garnett models. It is established that the extrema of the dielectric function frequency dependence for the metal nanoparticle–polydimethylsiloxane composite correspond to surface plasmon resonance in the inclusion particles. The effect of the nanoparticle-inclusion size on the nature of the frequency dependences is revealed, namely, an increase in the amplitude of the maxima and their “blue” shift with decreasing particle radius. The polarization charge density on the surface of the triboelectric layer is calculated for spherical inclusion particles of different radii, made of different metals, and at different concentrations. A qualitative similarity is demonstrated between the frequency dependence curves of the effective dielectric function real part and the polarization charge surface density. It is shown that changes in the material of the inclusion particles and their volume content in the triboelectric layer have a significant effect on the amplitude and position of the frequency dependence extrema of the polarization charge surface density. It is proven that by embedding metallic nanoparticles into the triboelectric layer of a triboelectric nanogenerator, it is possible to obtain a polarization charge on the surface of the layer, the maximum density of which is approximately equal to the surface density of the polarization charge for PDMS with dielectric particles embedded in it, having permeability $\epsilon \approx 10$, with the same volume content as for metallic particles.

Keywords: spherical nanoparticles, nanocomposite, effective dielectric function, surface plasmon resonance, triboelectric nanogenerator.

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Introduction

The current stage of electronic technology development is characterized by the integration of intelligent, networked, and sensor solutions into a wide range of applications. The widespread adoption of the Internet of Things [1-2], autonomous data analysis systems, and augmented reality tools are driving the need for energy-efficient hardware platforms capable of operating for long periods without maintenance. In this context, technologies for converting ambient energy into

electrical power are seen as a key element in shaping the infrastructure of future electronic systems.

Among existing approaches to energy supply based on piezoelectric, pyroelectric, and other similar effects, triboelectric nanogenerators (TENGs) have proven to be the most promising, demonstrating significantly higher specific power output with minimal design complications [3-5]. Triboelectric systems can accumulate mechanical energy of various origins – from vibrations of liquid and gas media to the biomechanical movements of humans and animals – and convert it into an electrical signal [6-8]. Of conceptual importance is the ability of such devices to

function as sensor elements, which allows them to be integrated into portable medical, information, and security systems as autonomous sensors [9-11].

The physical mechanism underlying the operation of triboelectric systems is based on contact electrification processes occurring during the interaction of two triboelectric materials. Contact is accompanied by charge transfer [12, 13], which creates a potential difference between the contacting surfaces. The performance characteristics of triboelectric systems are determined primarily by the surface charge density, which depends on the dielectric parameters of the triboelectric layer, electrodes, and substrate, as well as the efficiency of the charge transfer mechanism.

One of the most effective approaches to improving the output characteristics of triboelectric nanogenerators is to increase the permittivity of their active materials [14]. For this purpose, polymer matrices are widely used, into which particles of ceramic materials with high permittivity are introduced, in particular titanates and other oxide compounds [15, 16]. An increase in the effective permittivity of the composite leads to an increase in the surface charge density and stabilization of the contact electrification process that affects the performance characteristics of TENGs.

Of considerable interest are composite systems formed by embedding metallic nanoparticles or conductive nanostructures into the triboelectric layer [17]. Such modifiers can lead to significant changes in the electrical properties of the tribomaterial, in particular due to the excitation of surface plasmon resonances (SPR) in the metallic nanoparticle-inclusions. In our previous studies [18-22], the properties of composites based on spherical nanoparticles of varying morphologies embedded in dielectric matrices were investigated. Thus, in [18, 19] the optical properties of composites with metal-oxide nanoparticles were investigated, in [20] – composites with metal nanoshells, in [21] – with metal nanoparticles coated with a J-aggregate layer, in [22] – with metal nanoparticles coated with a layer of surfactant (oleylamine). Studying the effect of metal nanoparticle-inclusions on the electrical characteristics of TENG's triboelectric layer is a natural development of these researches and their extension to an area promising from the point of view of practical application.

I. Basic relations

The positive effect of surface plasmonic oscillations in metal nanoparticles embedded in the triboelectric layer on the output characteristics of TENGs is experimentally confirmed [23, 24]. However, the theoretical explanation for this effect remains speculative, only more or less well-founded. Clearly, the cause lies in the increase in charge on the surface of the movable electrode when it contacts the surface of the dielectric layer, since it is this charge that forms the current during the TENG's operating cycle. We propose the following mechanism for the increase in charge density on the contacting surfaces due to SPR.

Let's consider the triboelectric layer of a nanogenerator, which is most often made of polydimethylsiloxane (PDMS), doped with spherical

metal nanoparticles (Fig. 1). This layer exhibits the properties of a metal-dielectric nanocomposite with a frequency-dependent effective dielectric function.

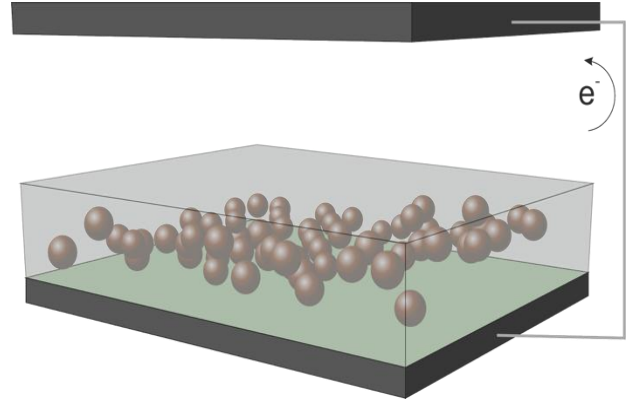


Fig. 1. Schematic representation of the TENG triboelectric layer.

Let's assume that a triboelectric layer is irradiated with sunlight. At a frequency corresponding to the SPR frequency, charge density oscillations occur in the inclusion particles. At each moment of this process, the particles can be considered as dipoles. The moments of these dipoles have a chaotic direction and magnitude, since the electric field of natural light changes chaotically in direction and phase within the triboelectric layer. The tribocharge field action causes the dipoles to turn in the direction of the field. The processes of chaotization in the light wave field and polarization in the triboelectric field occur simultaneously. There is reason to believe that the establishment of an ordered orientation of the dipoles in the triboelectric field occurs within a few femtoseconds and keeps up with the chaotic changes in the dipole moments.

As a result, at this stage of TENG operation, a constant polarization charge component should be expected to appear on the surface of the triboelectric layer. This surface charge, when the layer surface subsequently approaches the electrode, will lead to an increase in the charge on the electrode. Since this effect is caused by the generation of dipoles at the SPR frequency, we use the frequency-dependent dielectric function of the metal particles-PDMS composite to estimate the polarization charge surface density. Now let's move on to calculating this function.

We assume that the concentration of metal nanoparticles is low, so Maxwell-Garnett-type models can be used to describe the electrical and optical properties of the composite under study. In the conventional Maxwell-Garnett model, the effective dielectric function is

$$\epsilon_{\text{eff}}(\omega) = \epsilon_m \frac{(1+2\beta)\epsilon(\omega)+2(1-\beta)\epsilon_m}{(1-\beta)\epsilon(\omega)+(2+\beta)\epsilon_m}, \quad (1)$$

To take into account the size of nanoparticles, a modified (size-corrected) Maxwell-Garnett model is used. In this case [25]

$$\epsilon_{\text{eff}}^{\text{size}}(\omega) = \epsilon_m \frac{(1+2\beta)\epsilon(\omega)+2(1-\beta)\epsilon_m+(\epsilon_m-\epsilon(\omega))(1-\beta)\Delta}{(1-\beta)\epsilon(\omega)+(2+\beta)\epsilon_m+(\epsilon_m-\epsilon(\omega))(1-\beta)\Delta}. \quad (2)$$

In formulas (1) and (2) ϵ_m is the dielectric constant of PDMS; β is the volume content of metal in the composite; the correction factor and the wave parameter are

$$\Delta = \Delta_1 + i\Delta_2 = x^2 + \frac{2}{3}ix^3; \quad x = \sqrt{\epsilon_m} \frac{\omega R}{c}, \quad (3)$$

respectively; ω and c are the frequency and velocity of electromagnetic waves; R is the radius of the nanoparticle. The dielectric function of a particle in the Drude model has the form

$$\epsilon(\omega) = \epsilon' + i\epsilon'' = \epsilon^\infty - \frac{\omega_p^2}{\omega^2 + \gamma_{\text{eff}}^2} + i \frac{\omega_p^2 \gamma_{\text{eff}}}{\omega(\omega^2 + \gamma_{\text{eff}}^2)}, \quad (4)$$

where ϵ^∞ is the contribution of the crystal lattice to the permittivity of the metal; ω_p is the plasma frequency; γ_{eff} is the effective relaxation rate of electrons with three components – volume and surface relaxation and radiative damping

$$\gamma_{\text{eff}} = \gamma_{\text{bulk}} + \gamma_s + \gamma_{\text{rad}}, \quad (5)$$

$\gamma_{\text{bulk}} = \text{const}$, the rates of surface relaxation and radiative damping within the framework of the kinetic approach proposed by P. M. Tomchuk [26,27] are determined by the expressions

$$\gamma_s = \mathcal{S}(\omega, R) \frac{v_F}{R}, \quad (6)$$

$$\gamma_{\text{rad}} = \frac{V}{6\pi} \sqrt{\frac{\epsilon^\infty + 2\epsilon_m}{\epsilon_m}} \left(\frac{\omega_p}{c}\right)^3 \mathcal{S}(\omega, R) \frac{v_F}{R}. \quad (7)$$

In formulas (6) and (7), v_F is the Fermi velocity of electrons; $V = 4\pi R^3/3$ is the volume of a metal nanoparticle; an effective parameter describing the degree of coherence loss during electron scattering on the surface [26]

$$\mathcal{S}(\omega, R) = \frac{3}{4} \frac{1}{\epsilon^\infty + 2\epsilon_m} \left(\frac{\omega_p}{\omega}\right)^2 \left[1 - \frac{2v_s}{\omega} \sin \frac{\omega}{v_s} + \frac{2v_s^2}{\omega^2} \left(1 - \cos \frac{\omega}{v_s}\right)\right], \quad (8)$$

where $v_s = v_F/2R$ is the frequency of individual electron oscillations.

To estimate the polarization charge surface density, we use the formula

$$\frac{\sigma_{\text{pol}}}{\sigma_{\text{tr}}} = 1 + \frac{1}{1 + |\epsilon_{\text{eff}} - 1|}, \quad (9)$$

where σ_{pol} and σ_{tr} are the surface densities of the polarization and triboelectric charges.

Further calculations are performed using formulas (1), (2), and (9), taking into account relations (3)–(8).

II. Calculation results and their discussion

Calculations of the real and imaginary part frequency dependences of the TENG triboelectric layers effective dielectric function and the polarization charge surface density were performed within the framework of the Maxwell-Garnett model and the modified Maxwell-Garnett model for spherical metal nanoparticles of different radii, different metals and different concentrations embedded in a PDMS layer ($\epsilon_m = 2.7$). The characteristics of the particles-inclusions are presented in Table 1.

Table 1.

Metal parameters [21]			
Metals Parameters	Au	Ag	Cu
ϵ^∞	9.84	3.70	12.03
$\hbar\omega_p$, eV	9.07	9.17	12.6
$\hbar\gamma_{\text{bulk}}$, eV	0.023	0.016	0.024
v_F , 10^6 m/s	1.41	1.49	1.34

Figure 2 compares the frequency dependences of the

effective dielectric functions calculated using the Maxwell-Garnett model and the modified Maxwell-Garnett model. The difference between the results is that the amplitude of the extrema (maxima/minima of $\text{Re } \epsilon_{\text{eff}}$ and maxima of $\text{Im } \epsilon_{\text{eff}}$) is greater for the Maxwell-Garnett composites. Therefore, taking into account the size of the nanoparticles-inclusions leads to smoothing of the extrema of the effective dielectric function real and imaginary parts. Furthermore, the extrema of the dielectric function for the Maxwell-Garnett composite are shifted toward higher frequencies compared to the composite described by the modified model.

Importantly, the extrema of the frequency dependence of the composite's dielectric function real and imaginary parts correspond to the SPR frequency. Calculations show that in the region of the dielectric function's extrema, the condition $\text{Re } \epsilon(\omega) = -2\epsilon_m$ for SPR excitation at the metal-PDMS interface is satisfied with high accuracy, and small deviations from this equality are due to the content β of metal particles in the composite.

The frequency dependence curves of the effective permittivity real and imaginary parts of a nanocomposite with spherical Au nanoparticles of varying radii are shown in Fig. 3. The calculation results indicate an increase in the amplitude of the extrema and their shift to higher frequencies with decreasing particle radius. This fact can be explained by an increase in the surface area-to-volume ratio with decreasing nanoparticle radius. It is worth noting the presence of small-scale oscillations of the composite effective permittivity imaginary part in the near-infrared frequency range, which are a manifestation of classical size effects [19].

Figure 4 shows the frequency dependence of the polarization charge density on the surface of the triboelectric layer, calculated using formula (9), for different composite models and different particles-inclusions radii. As expected, the curves shown are similar to the frequency dependence curves $\text{Re } \epsilon_{\text{eff}}(\omega)$, since the

imaginary part of the composite's dielectric function is small. In particular, the extrema occur at almost the same frequency in the SPR excitation region.

The frequency dependences of the polarization charge surface density for nanoparticles of various metals and different volume contents embedded in the triboelectric layer are shown in Fig. 5. The amplitude of the extrema and their spectral position differ significantly for particles of different metals, as these metals have significantly

different plasma frequencies. As the volume content of embedded nanoparticles increases, the expected increase in the amplitude of the extrema is observed.

According to the calculation, by embedding metal nanoparticles into the triboelectric layer, it is possible to obtain a maximum polarization charge on its surface, the density of which is approximately equal to the surface density of the polarization charge for PDMS with dielectric particles embedded in it that have permeability

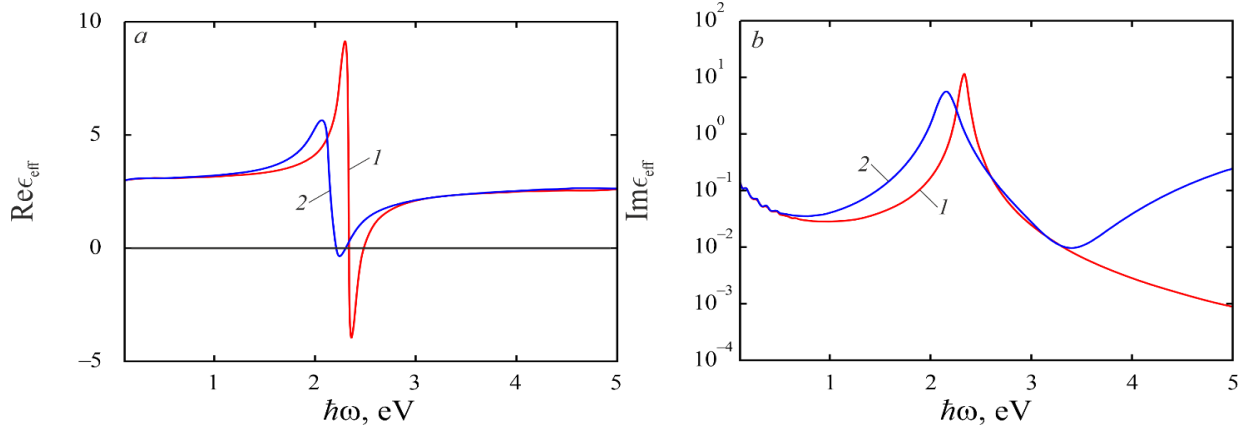


Fig. 2. Comparison of frequency dependences of the real (a) and imaginary (b) parts of the effective dielectric function within the Maxwell-Garnett model (curves 1) and the modified Maxwell-Garnett model (curves 2).

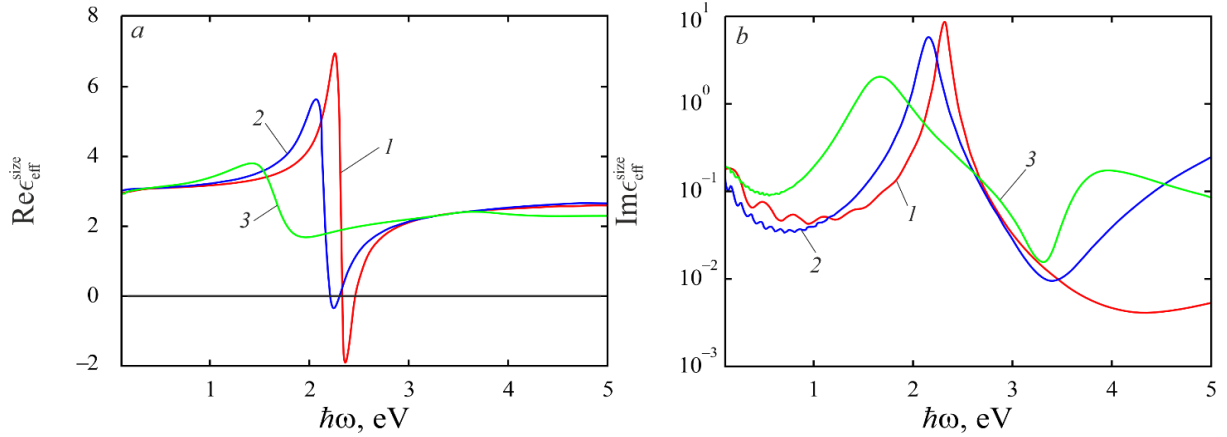


Fig. 3. Frequency dependences of the real (a) and imaginary (b) parts of the effective dielectric function within the modified Maxwell-Garnett model for a composite with Au nanoparticles of different radii: 1 – $R = 10$ nm; 2 – $R = 30$ nm; 3 – $R = 60$ nm.

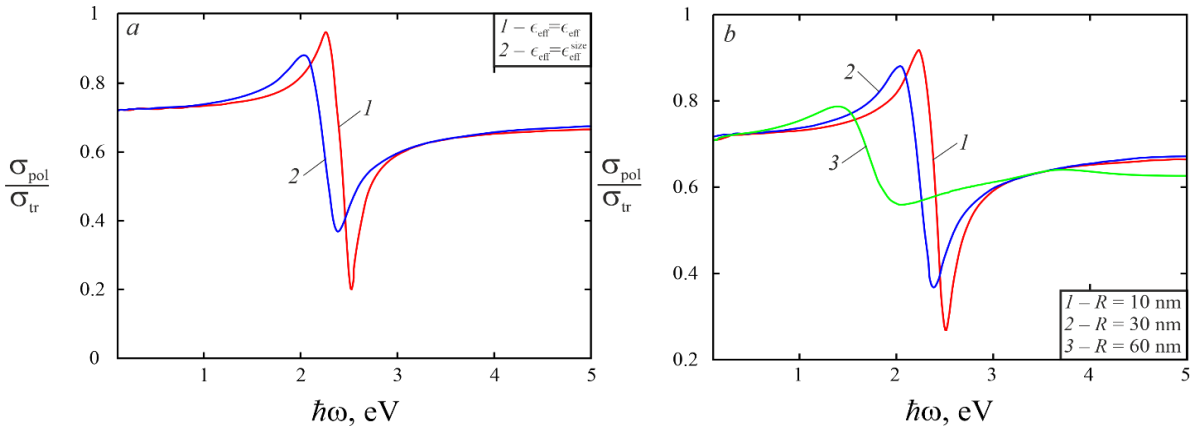


Fig. 4. Frequency dependences of the polarization charge density on the surface of the triboelectric layer: a – comparison of results for the Maxwell-Garnett model (curve 1) and the modified Maxwell-Garnett model (curve 2); b – within the framework of the modified Maxwell-Garnett model for Au nanoparticles of different radii.

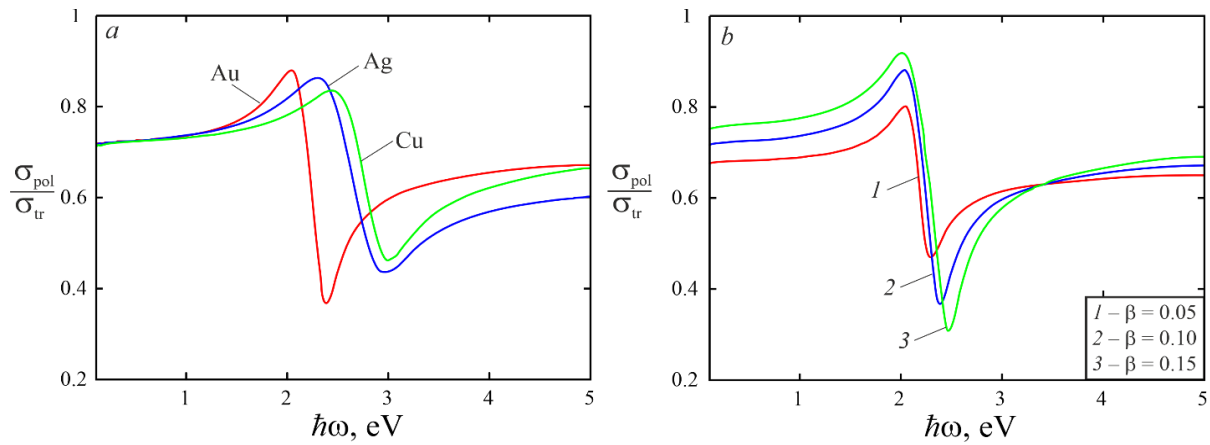


Fig. 5. Frequency dependences of the polarization charge density on the surface of the triboelectric layer: *a* – for nanoparticles of different metals; *b* – for Au nanoparticles with different volume contents.

$\epsilon \approx 10$, with the same volume content as for metal particles.

Formula (9) is approximate, since it assumes that all dipoles generated in sunlight have time to turn in the direction of the triboelectric charge field, and does not take into account the chaotic distribution of dipole moments by magnitude. Therefore, the SPR effect found using formula (9) should be considered overestimated.

Conclusions

An explanation is given of the influence of surface plasmon resonance excitation in metal nanoparticles embedded in a triboelectric layer on the TENG contacting surfaces charge density and TENG's performance characteristics.

Relationships are derived for the effective dielectric function of the spherical metal nanoparticles – PDMS composite and the polarization charge density on the surface of the triboelectric layer.

It was found that the extrema of the dielectric function frequency dependence of the metallic nanoparticles-PDMS composite correspond to surface plasmon resonance in the particles- inclusions.

It was established that taking into account the size of the particles-inclusions results in a "redshift" of the real and imaginary part frequency dependencies extrema and a decrease in their amplitude.

It is shown that with decreasing radius of the nanoparticles-inclusions, a "blue" shift of the frequency extremes occurs, their amplitude increases, and small-scale oscillations of the effective dielectric function

imaginary part appear, associated with classical size effects.

It is demonstrated that the amplitude and spectral position of the surface polarization charge density extremes depend on the material of the embedded nanoparticles and their volume fraction.

It is established that by embedding metallic nanoparticles into the triboelectric layer, a maximum polarization charge can be obtained on its surface, the density of which is approximately equal to the surface density of the polarization charge for PDMS with dielectric particles embedded in it that have permeability $\epsilon \approx 10$, with the same volume content as for metallic particles.

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Вплив плазмонних наночастинок на електрофізичні характеристики трибоелектричних шарів

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В роботі розглянуто задачу про вплив сферичних металевих наночастинок, впроваджених у трибоелектричний шар, на його електрофізичні характеристики. Висунуто гіпотезу про механізм впливу поверхневого плазмонного резонансу, збудженого у металевих частинках-включеннях, на густину заряду на контактуючих поверхнях трибоелектричного наногенератора та його експлуатаційні характеристики. В наближенні ефективного середовища знайдено ефективну діелектричну функцію композиту сферичні металеві наночастинок – полідиметилсилоксан. Розраховано частотні залежності дійсної та уявної частин ефективної діелектричної функції трибоелектричного шару в рамках класичної та скоригованої моделі Максвелл-Гарнета. Встановлено, що екстремуми частотної залежності діелектричної функції композиту металеві наночастинок – полідиметилсилоксан відповідають поверхневому плазмонному резонансу в частинках-включеннях. Виявлено вплив розмірів наночастинок-включень на характер частотних залежностей, а саме збільшення амплітуди максимумів та їх «синій» зсув при зменшенні радіусу частинок. Розраховано густину поляризаційного заряду на поверхні трибоелектричного шару для сферичних частинок-включень різного радіусу, з різних металів та за різної концентрації. Продемонстровано якісну подібність кривих частотних залежностей дійсної частини ефективної діелектричної функції та поверхневої густини поляризаційного заряду. Показано, що зміна матеріалу частинок-включень та їх об'ємного вмісту в трибоелектричному шарі суттєво впливають на амплітуду і положення екстремумів частотної залежності поверхневої густини поляризаційного заряду. Доведено, що за рахунок впровадження в трибоелектричний шар трибоенергетичного наногенератора металевих наночастинок можна отримати на поверхні шару поляризаційний заряд, максимальна густина якого приблизно дорівнює поверхневій густині поляризаційного заряду для PDMS з впровадженими в нього діелектричними частинками, які мають проникність $\epsilon \approx 10$, при тому ж самому об'ємному вмісті, що і для металевих частинок.

Ключові слова: сферичні наночастинок, нанокompозит, ефективна діелектрична функція, поверхневий плазмонний резонанс, трибоелектричний наногенератор.