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Effective permittivity and permeability of coated metal powders at microwave frequency

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ABSTRACT

Recent developments of the microwave heating of compacted metal powders bring a demand on calculation of corresponding effective optical parameters. Since the existing models cannot be applied to such powders, in this paper new effective dielectric permittivity and magnetic permeability are introduced by means of the combination of Mie theory and Bruggeman's effective medium. It is demonstrated that both an insulating shell and the skin effect should be taken into account. The shell drastically modifies the effective permittivity, while ignoring the skin effect brings to the permittivity an error as large as 30% at 30 GHz for copper powder with grains of 50 μm radius. The induced currents are shown to affect the magnetic properties of the powders. The strong skin effect substantially reduces the permeability of magnetic grains, and thus the intrinsic permeability measured in experiments can be much less than the real one. The strong dependence of the effective parameters on the microstructure of powders suggests a variety of possible heating scenarios in experiments.

Recently, the utilization of microwaves for heating was extended to compacted metal powders [1]. Bulk metal is known to mainly reflect microwaves since an alternating incident electromagnetic field induces macroscopic current, which in turn generates a strong secondary field oppositely directed to the incident one. In many metal powders irradiated by microwaves, macroscopic currents are not induced since the conductive grains are coated by an insulating oxide layer. As a result, microwaves can penetrate deeply inside the sample and induce microscopic currents strong enough to heat the cores by means of the Joule heating.

Though the microstructure of a composite determines its macroscopic properties, sometimes this information is complicated or may not be available. However, composites frequently behave like a homogeneous material on the scales relevant to an experiment. In this case, approximate homogeneous effective parameters are introduced.

There are different ways to introduce the effective parameters [2–6]. Among them the effective-medium approximation (EMA) [7] is frequently used. A number of numerical and experimental works prove that EMA is a good approximation for the estimations of optical functions of aggregate composites [8–10]. In this approach, the interactions between grains are taken into account by self-consistent manner and thus a multi-body problem is reduced to a single body one. In particular, each constituent of the powder along with pores is considered as equivalent inclusions embedded in effective Bruggeman's medium. Shape of the inclusions is usually chosen to be spherical, but it also can take other convenient forms. Parameters of the medium are determined self-consistently. For example, the space averaged far electromagnetic field due to these inclusions is required to vanish.

In this study expression for the far field is obtained by means of Mie theory [11]. The Mie theory (or sometimes Lorenz–Mie–Debye theory) is a general analytical solution for the electromagnetic field inside and outside of a homogeneous and isotropic spherical particle irradiated by a plane wave. The solution is given in terms of infinite series. Further improvements of the theory extend it to the case of coated particles [12]. Due to the use of Mie theory, the magnetic response is assumed to be isotropic and linear. Questions related the microwave heating of materials with spontaneous magnetization are answered by Tanaka et al. [13].

The obtained expressions for the effective parameters take into account the eddy current and an insulating layer simultaneously, and this differs from the expressions in an existing literature.

This paper is mainly focused on the case of a thin shell compared with the radius of the core, which frequently appears in experiments. In this case, a strong effect of the shell on the

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effective parameters, and thus on the heating rate, is not *a priori* apparent. Also it is not clear how the finite wavelength of microwaves affects the effective dielectric permittivity. Indeed, an incident electromagnetic wave induces microscopic currents in grains which are known to cause magnetization and results in the magnetic permeability of the powder. At the same time, the relationship between the effective permittivity and the induced currents is not evident.

Another question to be considered is the influence of the skin effect on the effective permittivity of nonmagnetic or magnetic coated powders. Specifically, does the shell affect the magnetization of a nonmagnetic powder? What is the overall effect of the induced current and the intrinsic permeability on the effective permeability? These questions are to be answered in the following sections.

1. Theoretical background

To determine the effective optical parameters of metal powders, lets consider a spherical inclusion embedded into homogeneous medium with yet unknown dielectric permittivity $\varepsilon_{\rm eff}$ and magnetic permeability $\mu_{\rm eff}$. The inclusion is subjected to a plane time-harmonic electromagnetic wave (Fig. 1). Bruggeman's approximation states that in effective medium the average scattered field due to inclusions must vanish.

Components of the scattered field can be found by means of Mie theory [11]. Since in microwave heating experiments the grain size in powders is usually much smaller than the wavelength of microwaves in background medium, only the first term of the Mie series contributes to the solution. In this case, the components of the scattered far field $\mathbf{E}_s$ at distance r from the particle ($k_{\rm eff}r \gg 1$) are given in spherical coordinates (r, θ, φ) by

$$E_{s\theta} = E_0 e^{-i\omega t}\frac{3i}{2}\frac{e^{ik_{\rm eff}r}}{k_{\rm eff}r}(a_1+b_1\cos\theta)\cos\varphi, \tag{1}$$

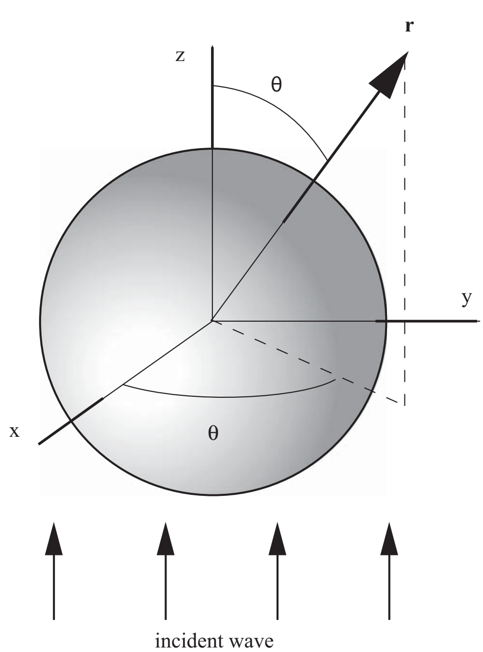

Fig. 1. Coordinate system used for calculation of scattered field. The x-polarized incident electromagnetic wave propagates along z-direction.

$$E_{s\varphi} = -E_0 e^{-i\omega t}\frac{3i}{2}\frac{e^{ik_{\rm eff}r}}{k_{\rm eff}r}(b_1+a_1\cos\theta)\sin\varphi, \tag{2}$$

where E_0 is the amplitude of the incident electric field at the inclusion position; $i=\sqrt{-1}$; $\omega=2\pi f$, f is the frequency of the electromagnetic wave; $k_{\rm eff}=\omega\sqrt{\varepsilon_{\rm eff}\mu_{\rm eff}}$; a_1 and b_1 are the Mie coefficients to be determined below. The self-consistent condition yields

$$\langle a_1\rangle = 0, \quad \langle b_1\rangle = 0, \tag{3}$$

where $\langle\,\rangle$ denotes averaging over various inclusions.

When the inclusion is a coated particle, the Mie theory gives the following coefficients a_1 and b_1 (see Appendix):

$$a_1 \approx \frac{2ix_2^3}{3}\frac{\mu_{\rm p}-\mu_{\rm eff}}{\mu_{\rm p}+2\mu_{\rm eff}}, \tag{4}$$

$$b_1 \approx \frac{2ix_2^3}{3}\frac{\varepsilon_{\rm p}-\varepsilon_{\rm eff}}{\varepsilon_{\rm p}+2\varepsilon_{\rm eff}}, \tag{5}$$

where

$$\mu_{\rm p} = \mu_2 F_2^{\mu}, \quad \varepsilon_{\rm p} = \varepsilon_2 F_2^{\varepsilon}, \tag{6}$$

with

$$F_2^{\mu} = 2\frac{1-(r_1/r_2)^3F_1^{\mu}}{2+(r_1/r_2)^3F_1^{\mu}}, \quad F_1^{\mu} = 2\frac{1-(\mu_1/\mu_2)F_0}{2+(\mu_1/\mu_2)F_0}, \tag{7}$$

$$F_2^{\varepsilon} = 2\frac{1-(r_1/r_2)^3F_1^{\varepsilon}}{2+(r_1/r_2)^3F_1^{\varepsilon}}, \quad F_1^{\varepsilon} = 2\frac{1-(\varepsilon_1/\varepsilon_2)F_0}{2+(\varepsilon_1/\varepsilon_2)F_0}. \tag{8}$$

Here, $r_{1,2}$ are the radii of the core and the shell; $\varepsilon_{1,2}$ and $\mu_{1,2}$ are the permittivity and permeability of the core and the shell, respectively (Fig. 2). The factor F_0 is

$$F_0(y) = 2\frac{-y\cos y+\sin y}{y\cos y-\sin y+y^2\sin y}, \tag{9}$$

with $y \equiv k_1 r_1$, where $k_{1,2}=\omega\sqrt{\varepsilon_{1,2}\mu_{1,2}}$. For highly conducting nonmagnetic materials $k_1r_1=(1+i)r_1/\delta$, where

$$\delta = \sqrt{\frac{2}{\omega\sigma\mu_0}} \tag{10}$$

is the skin depth ($\mu_0 = 1.2566\times10^{-6}$ H/m is the permeability of free space, σ is the static conductivity), and thus the factor F_0 takes into account the skin effect. When the skin effect is negligible ($r_1/\delta \ll 1$), the factor becomes $F_0=1$.

The assumptions used during derivation of Eqs. (4) and (5) are $|k_{\rm eff}r_2| \ll 1$ and $|k_2r_2| \ll 1$. Note that the magnitude of $|k_1r_1|$ is still unconstrained, and thus these expressions can be used for a dielectric core as well as for a highly conductive one.

When the inclusion is a pore filled by a gas with the permittivity $\varepsilon_{\rm g}$ and permeability $\mu_{\rm g}$, the coefficients a_1 and b_1

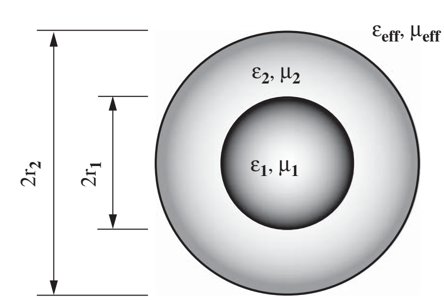

Fig. 2. Sketch of a coated particle.

are readily given by Eqs. (4) and (5) with ε_p and μ_p replaced by ε_g and μ_g, respectively.

Finally, from Eq. (3), the effective permeability μ_{eff} and permittivity ε_{eff} of Bruggeman's medium are determined by solving the equations

$$p\frac{\mu_p-\mu_{eff}}{\mu_p+2\mu_{eff}}+(1-p)\frac{\mu_g-\mu_{eff}}{\mu_g+2\mu_{eff}}=0, \tag{11}$$

$$p\frac{\varepsilon_p-\varepsilon_{eff}}{\varepsilon_p+2\varepsilon_{eff}}+(1-p)\frac{\varepsilon_g-\varepsilon_{eff}}{\varepsilon_g+2\varepsilon_{eff}}=0. \tag{12}$$

Here p is the volume fraction of particles.

The validity region of the obtained expressions is outlined by the more stringent assumption, namely $|k_{eff}r_2| \ll 1$ and $|k_2 r_2| \ll 1$. The lower limit comes from physical constraints. The grains should be large enough to use the macroscopic conductivity, permittivity and permeability. At microwave frequencies, metal powders with grains of a few tens of microns or less usually fall between these limits.

Eqs. (11) and (12) can be reduced to those derived in other works. First, when $r_1=r_2$ and $\mu_1=\mu_2$, then one obtains $\mu_p=F_0\mu_1$, which is the known result for the magnetic permeability of a homogeneous metal particle [14–16,8]. In this case, the induced eddy current causes magnetization of the particle, and therefore the effective relative permeability is not unity in spite of the fact that the particle itself may be nonmagnetic. Secondly, the obtained expressions correctly describe the optical behavior of dielectric coated particles ($F_0=1$) considered by Bohren et al. [17] and Buchelnikov et al. [18]. In the latter work, some comparison of the obtained effective permittivity with the existing expressions is made. In contrast to the works noted above, expressions (11) and (12) are more general since they take into account the skin effect and the insulating shell simultaneously.

It is interesting to note that, from Eq. (6) in the case of a homogeneous grain when $r_2=r_1$ and $\varepsilon_2=\varepsilon_1$, the "equivalent permittivity" of the grain becomes $\varepsilon_p=\varepsilon_1 F_0$ [19], though it is expected to be $\varepsilon_p=\varepsilon_1$. At microwave frequencies the imaginary part of the dielectric permittivity of a highly conductive core is much larger than the real one, and in this study the real part is neglected, i.e. $\varepsilon_1 \approx i\sigma/\omega$. The factor F_0 introduces a large and negative real part to the permittivity ε_p, which corresponds to free electron dielectric response at microwave frequencies in good conductors according to Drude and Sommerfeld models [20]. For example, for a copper grain of $r_1=5\,\mu m$ irradiated at 2.45 GHz the intrinsic permittivity is $\varepsilon_1/\varepsilon_0=i4.3\times10^8$, while one has $F_0\varepsilon_1/\varepsilon_0=(-1.1+i1.2)\times10^8$, where $\varepsilon_0=8.85\times10^{-12}$ F/m is the permittivity of free space.

In this study, the optical parameters of gas filling pores are assumed to be $\varepsilon_g=\varepsilon_0$ and $\mu_g=\mu_0$.

2. Effective permittivity

2.1. Effect of skin depth

With the Mie expressions the effect of skin depth on the effective permittivity ε_{eff} can be naturally taken into account. This effect was usually ignored in previous works. Numerical analysis shows that the estimation error caused by exclusion of the skin effect, defined by $[\varepsilon_{eff}-\varepsilon_{eff}(F_0=1)]/\varepsilon_{eff}$, increases with increasing frequency and the particle core radius, and decreases as the core conductivity increases (Fig. 3). Ignoring the skin effect introduces

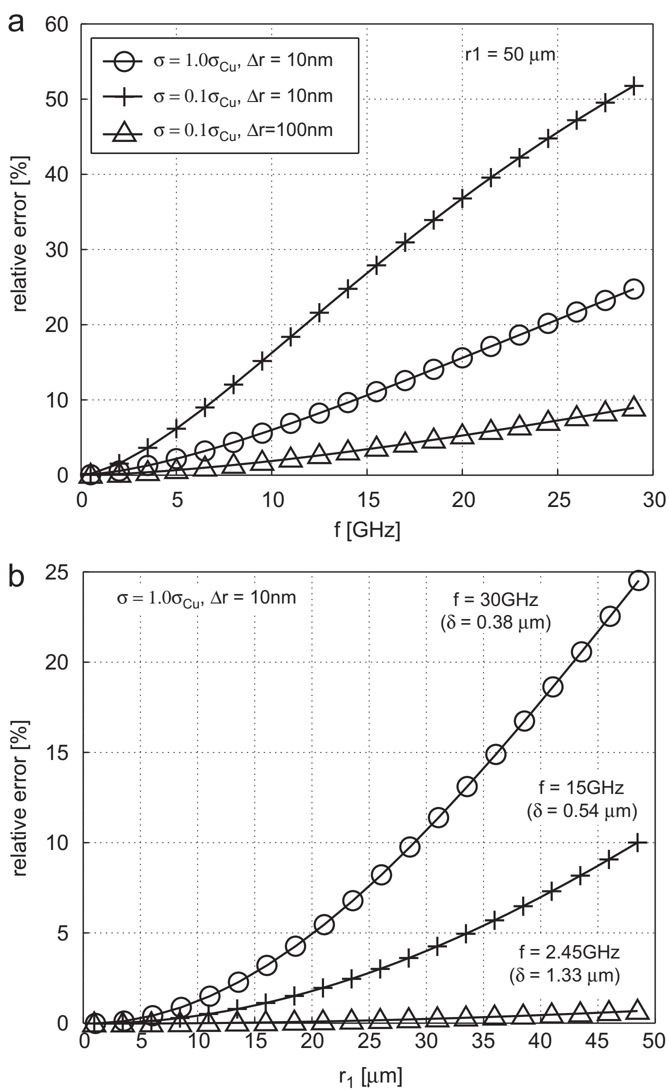

Fig. 3. Estimation error in $\varepsilon_{eff}=\varepsilon'_{eff}+i\varepsilon''_{eff}$ caused by ignoring the skin effect: (a) dependence on frequency and (b) dependence on the core radius. Here, $\Delta r=r_2-r_1$, $\sigma_{Cu}=5.8\times10^7$ S/m, $\varepsilon_2/\varepsilon_0=3+i0.3$, $p=0.9$. The relative error is defined by $[\varepsilon_{eff}-\varepsilon_{eff}(F_0=1)]/\varepsilon_{eff}$. Largest of errors of ε''_{eff} and ε'_{eff} is shown.

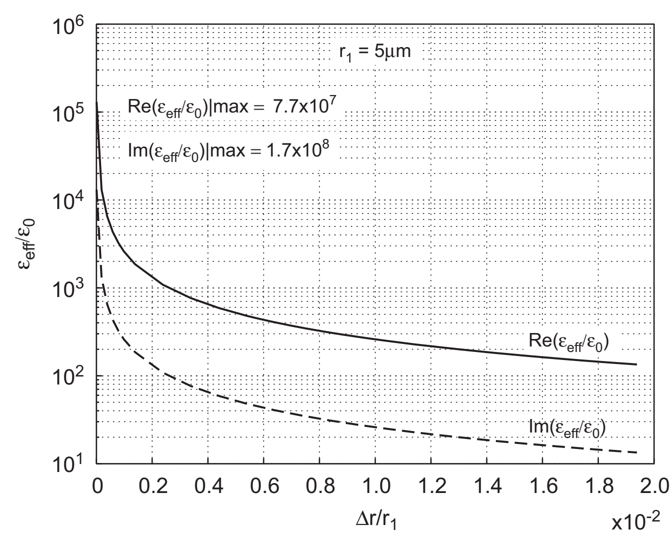

Fig. 4. Effect of the shell thickness on effective permittivity ε_{eff} for $\varepsilon_2/\varepsilon_0=3+i0.3$, $r_1=5\,\mu m$, $p=0.9$, $\sigma=5.8\times10^7$ S/m and $f=2.45$ GHz. The shell thickness Δr changes from 0.1 to 100 nm. Maximal values of the real and imaginary parts of the effective permittivity $\varepsilon_{eff}/\varepsilon_0$ are 7.7×10^7 and 1.7×10^8, respectively. The results demonstrate that even a thin shell drastically reduces ε_{eff}.

an error as large as tens of percents for compacted powders with grains of a few tens of microns or larger at $f = 30\,\mathrm{GHz}$ [Fig. 3(a)]. In the case of finer powders with a few microns radii, the influence of the skin effect on $\varepsilon_{\mathrm{eff}}$ is small [Fig. 3(b)]. Smaller thickness of the shell also magnifies the error.

2.2. Effect of shell thickness

In practice the radius of metal core r_1 is frequently much larger than the thickness of shells, i.e. $\Delta r \equiv r_2 - r_1 \ll r_1$, which raises a question of the influence of the shell on the effective parameters. Nevertheless, the calculations show that even a very thin shell $\Delta r / r_1 < 1 \times 10^{-2}$ significantly reduces $\varepsilon_{\mathrm{eff}}$ in comparison with the permittivity of the core (Fig. 4).

When the shell is thick, the effective permittivity tends to the permittivity of the shell ε_2. In the opposite case when the grains in a compacted powder are almost homogeneous, $\varepsilon_{\mathrm{eff}}$ is determined by the permittivity of the core ε_1. Since the difference between ε_1 and ε_2 can be many orders of magnitude, the effective permittivity changes drastically with the thickness of the shell.

3. Effective permeability of nonmagnetic powders

3.1. Effect of skin depth

When an uniform nonmagnetic metal particle ($\mu_1 = \mu_2 = \mu_0$) is subjected to microwave irradiation, an induced current causes magnetic moment and the particle effectively behaves like a magnetic one [14]. For particles coated by an insulating shell, electric current flows only in the core and thus "permeability of grains" $\mu_{\mathrm{p}} = \mu_0 F_0$ is determined merely by the core parameter r_1/δ (Fig. 5).

The result shows that the particles do not couple with the magnetic component when the grain cores are very small ($\mu_{\mathrm{p}} \to \mu_0$ when $r_1/\delta \ll 1$) or very large ($\mu_{\mathrm{p}} \to 0$ when $r_1/\delta \gg 1$). Between these cases, there is a region where the imaginary part of the "permeability of grain" μ_{p}'' has a maximum. The flow of the induced current changes here from volumetric to surface one [14].

Clearly, the powder composed of such particles also exhibits magnetic behavior. Position of μ_{eff}'' maximum directly corresponds to that of μ_{p}'' and it is almost independent of the particles

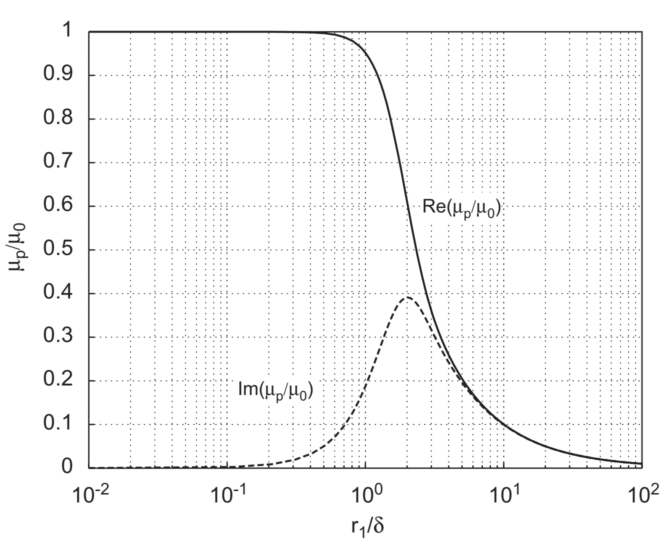

Fig. 5. The induced "permeability of grain" μ_{p} in the case of nonmagnetic homogeneous particle as a function of r_1/δ ratio. The result demonstrates that in spite of nonmagnetic material, the grains themselves show magnetic behavior.

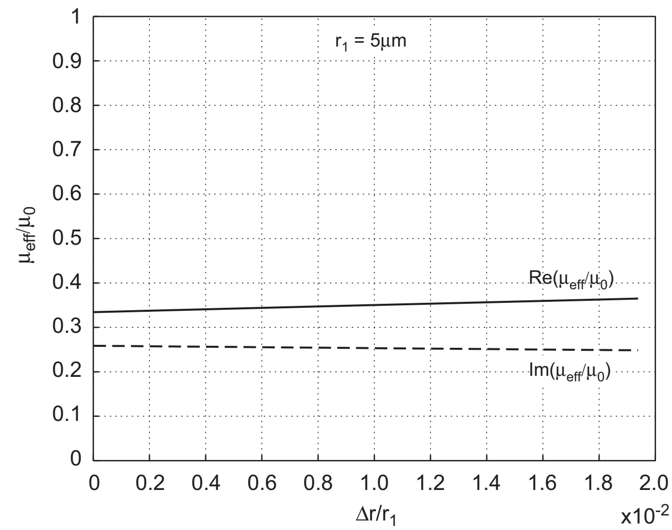

Fig. 6. Effect of the shell thickness on effective permeability μ_{eff} for $r_1 = 5\,\mu\mathrm{m}$, $p = 0.9$, $\sigma = 5.8 \times 10^7\,\mathrm{S/m}$ and $f = 2.45\,\mathrm{GHz}$. The shell thickness Δr changes from 1 to 100 nm. For the presented parameters, the effective μ_{eff}, on the contrary to $\varepsilon_{\mathrm{eff}}$, is almost insensitive to the thickness of shell.

volume fraction. Of course, the volume fraction should be high enough for the pronounced induced effective permeability.

3.2. Effect of shell thickness

In contrast to the effective permittivity, the effective permeability μ_{eff} is almost insensitive to the shell thickness when $\Delta r \ll r_1$ (Fig. 6). The magnetic permeability is caused by induced currents in a core, and therefore parameters of the core mainly determine it. Nevertheless, in the limiting case when the thickness of the shell is comparable or larger than the radius of the core, the effective permeability tends to the permeability of the shell (not shown in the figure).

4. Effective permeability of magnetic powders

The skin effect is known to modify the effective permeability of powders made of homogeneous grains [15,16,8]. In the case of coated grains, a shell contributes to the effective magnetic permeability along with a core, and a question arises about the significance of this contribution. Another question concerns with interaction of the induced permeability with the intrinsic one. To answer these questions, two cases are investigated: (i) grains consist of nonmagnetic cores covered by magnetic shells and (ii) grain cores are magnetic but coated by nonmagnetic shells.

In order to reduce the number of independent parameters, the "permeability of grain" μ_{p} is considered instead of the effective permeability μ_{eff}. To distinguish the induced permeability determined by Eq. (6) with $\mu_1 = \mu_2 = \mu_0$ from the intrinsic one, the former is denoted as μ_{ind} in this section.

In following calculations, the ratio $(r_1/r_2)^3$ is fixed at 0.9. The ratio r_1/δ is set to 0.1, 2 and 5, which corresponds to nonmagnetic, highly lossy and highly diamagnetic behavior of μ_{ind} (see Fig. 5, where $\mu_{\mathrm{p}} \equiv \mu_{\mathrm{ind}}$). The range of the intrinsic permeability is chosen to demonstrate the behavior of μ_{p}.

Dependence of the magnetic "permeability of grain" μ_{p} on that of the shell μ_2 in the case of nonmagnetic core is presented in Fig. 7. The results show that the shell weakly affects the μ_{p} for the presented conditions, which is contrasted to the case of dielectric permittivity. Note that the permeability μ_p can be roughly decomposed into the r_1/δ depending part and the μ_2 depending part. It means that the shell and the core interact with the magnetic field almost independently. This conclusion is supported by the empirical expressions for μ_p obtained based on Fig. 7:

$$\mu_p \approx \mu_{ind}\frac{r_1^3}{r_2^3} + 2\mu_2\frac{1-(r_1/r_2)^3}{2+(r_1/r_2)^3}. \tag{13}$$

This expression points out that the contribution of the shell is proportional to $1-(r_1/r_2)^3$. Therefore, a thin shell can significantly affect μ_p only in the case of large value of $|\mu_2|$.

Fig. 8 shows the magnetic "permeability of grain" μ_p in the case of a magnetic core coated by a nonmagnetic shell. The results indicate that the strong skin effect (large r_1/δ ratio) substantially reduces μ_p. Only in the case of "fine" powders when $r_1/\delta \ll 1$, the permeability μ_p restored from experimental data directly corresponds to the intrinsic permeability. In the case of "coarse" powders ($r_1/\delta \gg 1$), the measured μ_p is much less than the intrinsic one.

5. Dependence on volume fraction of particles

The dependence of the effective parameters ε_{eff} and μ_{eff} on the volume fraction p is shown in Fig. 9. In calculations, the powder is assumed to be nonmagnetic. The core conductivity is $\sigma = 5.8\times10^7$ S/m, which is the static conductivity of copper. Relative permittivity of the shell is arbitrarily set to $3+i3$ or $3+i0.3$. The shell thickness is fixed at 100 nm. The microwave frequency is 2.45 GHz.

Both the real and imaginary parts of ε_{eff} increases with the volume fraction. The magnitude of the effective permittivity ε_{eff} (especially the imaginary part) changes drastically by orders of magnitude at a percolation threshold. For Bruggeman's medium the percolation threshold is $p_c = 0.33$.

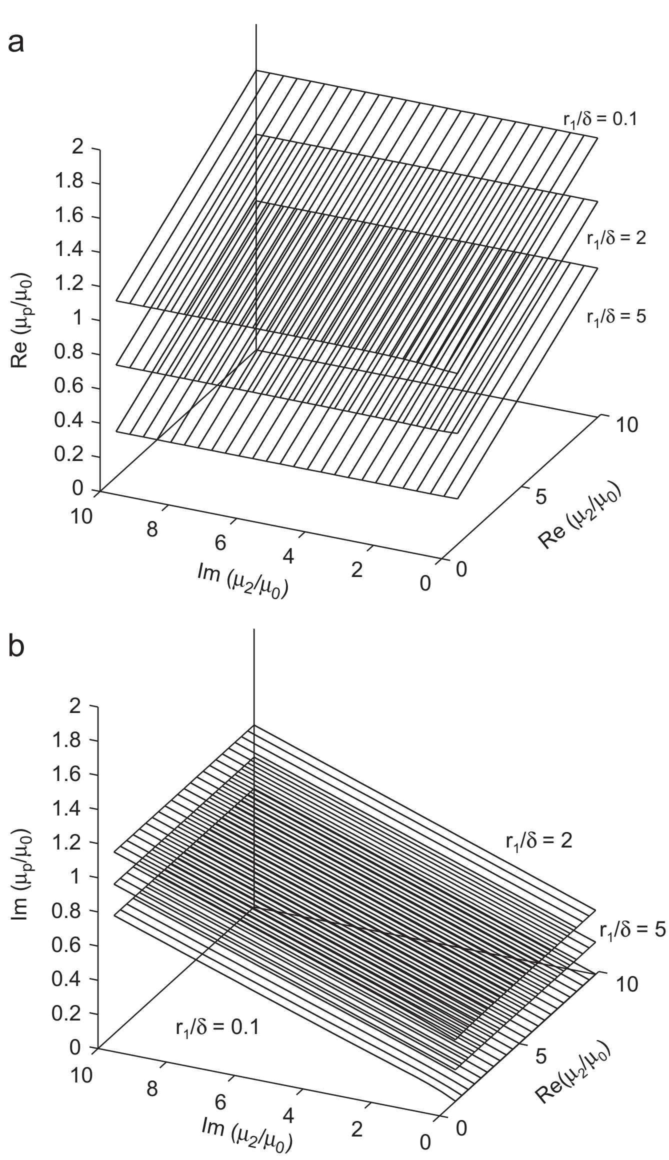

Fig. 7. Permeability μ_p as a function of intrinsic permeability of the shell μ_2, (a) real part and (b) imaginary part. Core is assumed to be nonmagnetic. To demonstrate the skin effect, ratio r_1/δ is set to 0.1, 2 and 5. Within the used parameters the resultant μ_p is a sum of the term depending on r_1/δ ratio and the term depending on μ_2.

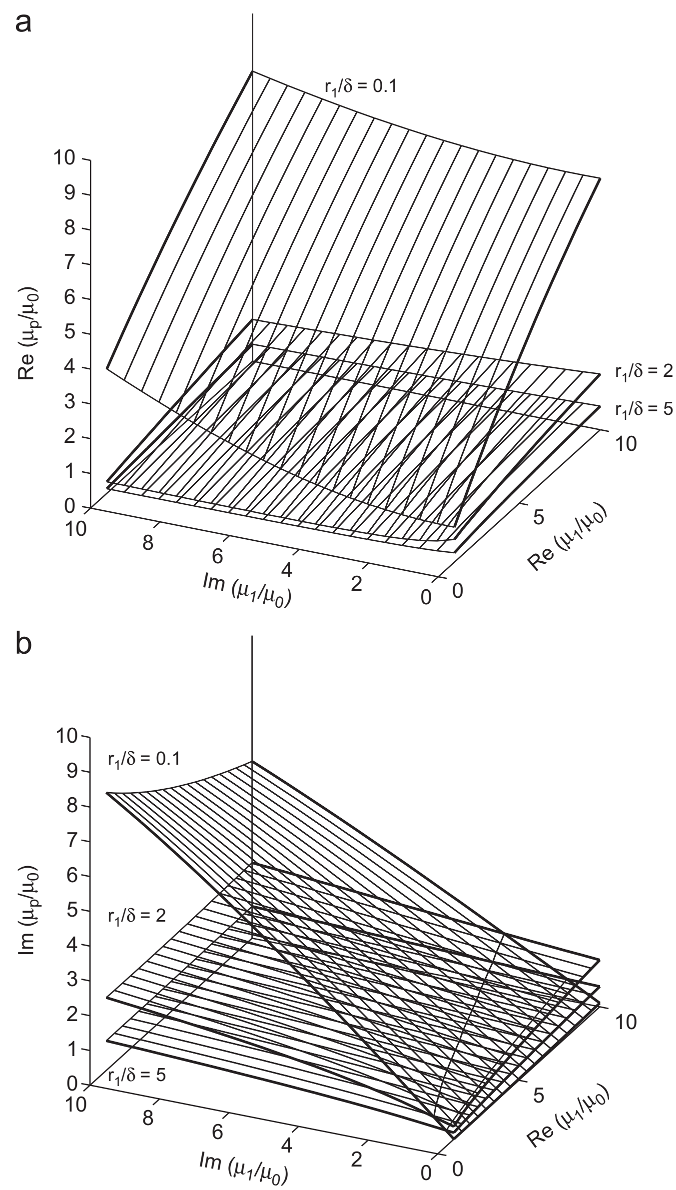

Fig. 8. Permeability μ_p as a function of intrinsic permeability of core μ_1, (a) real part and (b) imaginary part. Shell is assumed to be nonmagnetic. To demonstrate the skin effect, ratio r_1/δ is set to 0.1, 2 and 5, where δ is the skin depth of the corresponding nonmagnetic core [see Eq. (10)].

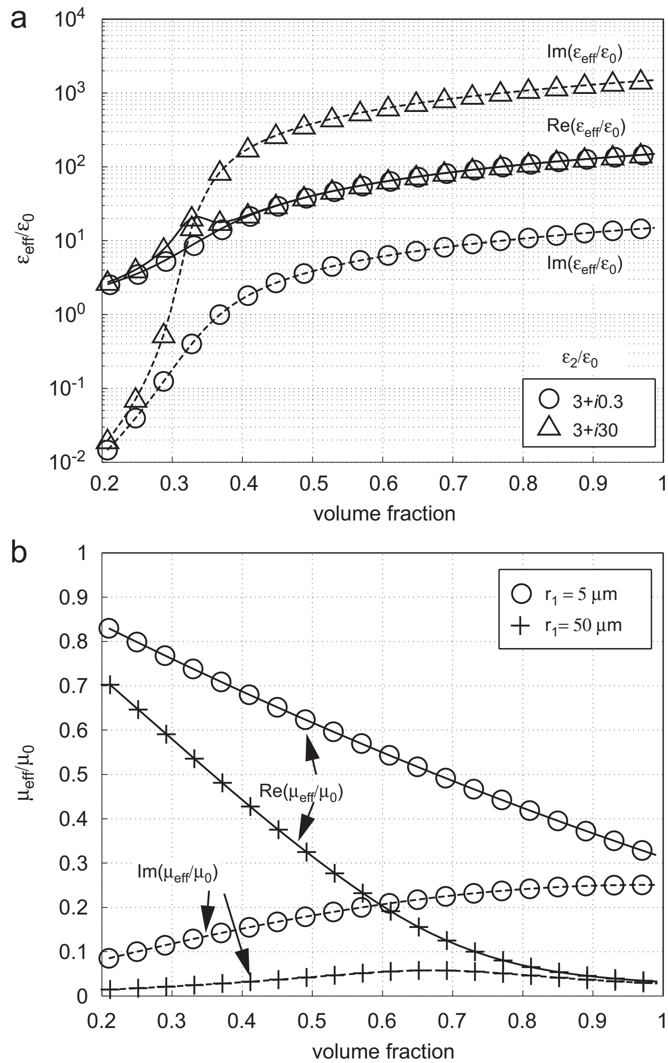

Fig. 9. Effective parameters as a function of volume fraction of particles p: (a) effective permittivity, $r_1 = 5\,\mu\text{m}$ and (b) effective permeability. Core conductivity $\sigma = 5.8 \times 10^7$ S/m and $f = 2.45$ GHz, shell thickness is 100 nm.

Results also demonstrate that above p_c when $p \gtrsim 0.5$, the loss tangent $\varepsilon''/\varepsilon'$ of a compacted powder is approximately close to that of the shell.

Magnetic permeability changes much smoother and less in magnitude with volume fraction. The real part of μ_{eff} decreases as volume fraction increases while the imaginary one increases. At large r_1/δ ratios the imaginary part of μ_{eff} has almost unnoticeable maximum.

6. Conclusion

Optical parameters of a composite made of compacted metal particles and irradiated at microwave frequencies are investigated. The corresponding expressions are obtained by means of self-consistent Bruggeman's approximation combined with Mie theory. The use of Mie theory allows natural account for finite frequencies of incident signals as well as the dielectric coating of metal cores. The region of validity of obtained expressions is roughly outlined in Section 1.

Numerical analysis in Section 2 shows that the error caused by exclusion of the skin effect increases with increasing frequency and the particle core radius, and decreases as the core conductivity increases. Ignoring the skin effect introduces an error as large as tens of percents for compacted powders with grains of a few tens of microns or larger at $f = 30$ GHz. In the case of finer powders with a few microns radii, the influence of the skin effect on ε_{eff} is small.

In the case of nonmagnetic metal grains, their magnetic behavior is solely determined by induced currents and thus the skin effect is again important (Section 3).

Next factor to be taken into account is the thickness of the shell. This paper is focused on the case of a thin shell compared to the radius of the core. In spite of that the shell is much thinner than the core radius, the shell is shown to drastically reduce ε_{eff} in comparison with the dielectric permittivity of the metal core ε_1. At the same time, the μ_{eff} is not so sensitive to the shell thickness.

The account for a magnetic shell or a magnetic core modifies the effective permeability (Section 4). When a nonmagnetic core is coated by a thin magnetic shell, the contribution from the shell to "permeability of grain" is scaled as $1-(r_1/r_2)^3$. Therefore this contribution is significant only for large values of magnetic permeability of the shell $|\mu_2|$. In the case of the magnetic core coated by a nonmagnetic shell, strong skin effect $(r_1/\delta \gg 1)$ substantially reduces μ_p.

The effective dielectric permittivity strongly depends on volume fraction (Section 5). The magnitude of ε_{eff} drastically changes at a percolation threshold. Results demonstrate that in the case of highly conductive cores, the loss tangent $\varepsilon''/\varepsilon'$ of a compacted powder is approximately close to that of the shell when $p \gtrsim 0.5$. The effective magnetic permeability changes much smoother with the volume fraction.

The calculations demonstrate that the effective optical parameters of metal powders strongly depend on many parameters: the relative thickness of the shell (r_1^3/r_2^3 ratio), the skin effect (r_1/δ), parameters of the core and shell, the volume fraction. Ignoring the complex behavior of ε_{eff} and μ_{eff} can lead to the misinterpretation of similar experiments conducted by different groups of researchers. For example, the parameters strongly depend on the thickness of a shell. The shell can be affected by a number of ways: the powder production process, powder storage, the type of gas atmosphere in the heating experiments. This leads to a wide range of magnitudes of the effective permittivity and, as a result, to different coupling of powders with microwaves even if particle size distributions are the same.

Another example concerns with the size of grains. The dependence of the effective parameters on the particle size makes microwave heating sensitive to the size distributions. In the heating experiments, the particle size distributions are frequently characterized by a single number like the average size of grains or the size of passed standard mesh. The obtained results strongly point out that the same average size of grains or that of passed mesh does not guarantee similar coupling of powders with microwaves. In addition, the sensitivity to the particle size suggests the sensitivity to the particle shape. It means that the powders made of spherical grains and those made of flake-shaped grains couple with microwaves differently.

The variety of factors strongly affecting the behavior of microwave heating leads to variety of possible experimental results. To make definite comparison of the results, the detailed description of the experimental conditions is required.

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Appendix A. Expressions for a_1 and b_1 for a small coated particle

In the case of a coated particle embedded in background medium with the dielectric permittivity ε_b and magnetic permeability μ_b, the Mie coefficients a_n and b_n, where $n=1,2,3,\ldots$, are given by [12]

$$a_n = -R_n^{(1)}(x_2)\frac{m_2\mu_b H_n^a - \mu_2 D_n^{(1)}(x_2)}{m_2\mu_b H_n^a - \mu_2 D_n^{(3)}(x_2)}, \tag{14}$$

$$b_n = -R_n^{(1)}(x_2)\frac{\mu_2 H_n^b - m_2\mu_b D_n^{(1)}(x_2)}{\mu_2 H_n^b - m_2\mu_b D_n^{(3)}(x_2)}, \tag{15}$$

with

$$H_n^a = \frac{R_n^{(2)}(m_2x_2)D_n^{(1)}(m_2x_2) - A_n D_n^{(2)}(m_2x_2)}{R_n^{(2)}(m_2x_2) - A_n}, \tag{16}$$

$$H_n^b = \frac{R_n^{(2)}(m_2x_2)D_n^{(1)}(m_2x_2) - B_n D_n^{(2)}(m_2x_2)}{R_n^{(2)}(m_2x_2) - B_n} \tag{17}$$

and

$$A_n = R_n^{(2)}(m_2x_1)\frac{m_1\mu_2 D_n^{(1)}(m_1x_1) - m_2\mu_1 D_n^{(1)}(m_2x_1)}{m_1\mu_2 D_n^{(1)}(m_1x_1) - m_2\mu_1 D_n^{(2)}(m_2x_1)}, \tag{18}$$

$$B_n = R_n^{(2)}(m_2x_1)\frac{m_2\mu_1 D_n^{(1)}(m_1x_1) - m_1\mu_2 D_n^{(1)}(m_2x_1)}{m_2\mu_1 D_n^{(1)}(m_1x_1) - m_1\mu_2 D_n^{(2)}(m_2x_1)}. \tag{19}$$

Here, $x_{1,2}=k_b r_{1,2}$, $k_b=\omega\sqrt{\varepsilon_b\mu_b}$, $\omega=2\pi f$, f is the frequency of the incident plane wave, $r_{1,2}$ are the radii of the core and the shell; $m_{1,2}=\sqrt{\varepsilon_{1,2}\mu_{1,2}/(\varepsilon_b\mu_b)}$ are the relative refractive indices, with $\varepsilon_{1,2}$ and $\mu_{1,2}$ being the permittivity and permeability of the core and the shell, respectively. The ratios $R_n^{(1)}$ and $R_n^{(2)}$ are defined as

$$R_n^{(1)}(x) = \frac{j_n(x)}{h_n^{(1)}(x)}, \quad R_n^{(2)}(x) = -\frac{j_n(x)}{y_n(x)}, \tag{20}$$

where j_n, y_n are the spherical Bessel functions, and $h_n^{(1)}$ is the spherical Hankel functions of the first kind. Logarithmic derivatives are given by

$$D_n^{(i)}(x) = \frac{[xz_n^{(i)}(x)]'}{xz_n^{(i)}(x)}, \tag{21}$$

where $z_n^{(1)} \equiv j_n$, $z_n^{(2)} \equiv y_n$ and $z_n^{(3)} \equiv h_n^{(1)}$.

If $|x_2| \ll 1$, then only terms with $n=1$ contribute to the final solution.

Additional condition $|m_2x_2| \ll 1$ makes possible the use of the only first terms in Taylor series of $R_1^{(j)}, j=1,2$ and $D_1^{(j)}, j=1,2,3$, i.e.

$$R_1^{(1)}(x) \approx \frac{ix^3}{3}, \quad R_1^{(2)}(x) \approx \frac{x^3}{3} \tag{22}$$

and

$$D_1^{(1)}(x) \approx \frac{2}{x}, \quad D_1^{(2)}(x) \approx D_1^{(3)}(x) \approx -\frac{1}{x}. \tag{23}$$

Here, the argument x can be x_2, m_2x_2 or m_2x_1 but not m_1x_1. Since the core is assumed to be highly conductive, its permittivity can be large at microwave frequencies, therefore the full expression for logarithmic derivative $D_1^{(1)}(m_1x_1)$ should be used:

$$D_1^{(1)}(m_1x_1) = \frac{2}{m_1x_1F_0(m_1x_1)}, \tag{24}$$

where

$$F_0(y) = 2\frac{-y\cos y + \sin y}{y\cos y - \sin y + y^2\sin y}. \tag{25}$$

In this case, a_1 and b_1 are equal to

$$a_1 \approx \frac{2ix_2^3}{3}\frac{\mu_p - \mu_b}{\mu_p + 2\mu_b}, \tag{26}$$

$$b_1 \approx \frac{2ix_2^3}{3}\frac{\varepsilon_p - \varepsilon_b}{\varepsilon_p + 2\varepsilon_b}, \tag{27}$$

where $i=\sqrt{-1}$; $\mu_p=\mu_2 F_2^\mu$ and $\varepsilon_p=\varepsilon_2 F_2^\varepsilon$ with

$$F_2^\mu = 2\frac{1-(r_1/r_2)^3F_1^\mu}{2+(r_1/r_2)^3F_1^\mu}, \quad F_1^\mu = 2\frac{1-(\mu_1/\mu_2)F_0}{2+(\mu_1/\mu_2)F_0}, \tag{28}$$

$$F_2^\varepsilon = 2\frac{1-(r_1/r_2)^3F_1^\varepsilon}{2+(r_1/r_2)^3F_1^\varepsilon}, \quad F_1^\varepsilon = 2\frac{1-(\varepsilon_1/\varepsilon_2)F_0}{2+(\varepsilon_1/\varepsilon_2)F_0}. \tag{29}$$

The most stringent among the two used conditions ($|x_2| \ll 1$ and $|m_2x_2| \ll 1$) determines the upper limit of appropriate particle size. The same conditions rewritten in terms of wavevectors and particle radius are $|k_b r_2| \ll 1$ and $|k_2 r_2| \ll 1$, where $k_{1,2}=\omega\sqrt{\varepsilon_{1,2}\mu_{1,2}}$.

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