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THE EFFECTIVE PERMEABILITY OF MIXTURES OF SOLIDS

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Summary

The effective dielectric constant is calculated for a material, consisting of a medium in which solid particles or empty holes with various dielectric constants are packed together and in which the individual particles or holes are assumed to be ellipsoidical. The calculation is based on an approximation which in the case of spherical particles proves to give results identical with those obtained by Böttcher's method. The effect of the shape of the ellipsoides is discussed. Considerations are given concerning the nature of the approximations used in this paper and in other theories on this subject.

1. *Introduction.* For several purposes it is of importance to know how the dielectric or magnetic permeability of a mixture of substances, which show mutually different permeabilities in pure state, depends on the composition of the mixture. The problem arises for instance with the calculation of the dielectric constant of a homogeneous mixture of a number of different liquids. The theory of this case has been extensively investigated by Onsager [1]) and Böttcher [2]). In this paper we shall deal with the case where the different substances are powders of solids, suspended or packed together in a medium like oil or wax or air. Evidently the case of a solid which contains holes filled with some liquid or with air is automatically included in this treatment. We shall always suppose that the mixtures are as homogeneous as possible. In practice we find a magnetic example in the cores of iron dust as used in chokes. The problem of the calculation of the mean dielectric constant of a single crystal from measurements on the effective dielectric constant of the crystal powder also shows the importance of a reliable theory on this subject.

Several authors [3]) have given formulae for the effective permeability of mixtures, some of them being pure interpolation formulae, while others are more or less theoretically based. Here the difficulty arises that it is impossible to derive a theoretically exact expression, so that simplifications or approximations must be introduced. This is also the case in the derivation of the formula, that Böttcher [3]) gives for the effective dielectric constant ε' of a powder of a solid material (with $\varepsilon = \varepsilon_0$) in air. The formula of Böttcher is

$$\frac{\varepsilon' - 1}{3\varepsilon'} = f \, \frac{\varepsilon_0 - 1}{\varepsilon_0 + 2\varepsilon'}, \tag{1}$$

where f is the filling factor, i.e. the volume percentage of the solid material in the powder. Fair agreement with the experimental data on KCl was found. The formula should hold in the whole region $0 \leqslant f \leqslant 1$, independent of the shape of the individual solid particles, respectively of the shape of the holes enclosed in the solid material.

We believe however, that the last statement cannot be correct. Firstly we have a pure theoretical argument in support of our opinion. Suppose that a small amount ($f \ll 1$) of a solid material with a high value of ε_0 ($\varepsilon_0 = 100$ for instance) is suspended in a medium with $\varepsilon = 1$ and that each particle has the shape of a sphere. In this case it is well known that the polarization per cm^3 induced in one sphere is

$$\frac{3}{4\pi} \, \frac{\varepsilon_0 - 1}{\varepsilon_0 + 2} E$$

and from this it is easily seen that for a high value of ε_0, always $\varepsilon' = 1 + 3f$ approximately. The reason of this behaviour is the occurrence of the depolarizing field in each sphere, which prevents the occurrence of a high polarization. Suppose now that the same amount of the material is suspended as particles of the shape of very long and thin needles, directed in the direction of the field. In this case the depolarization is very small and the polarization induced in the needles is $\varepsilon_0 E/4\pi$, or $\varepsilon' = 1 + f\varepsilon_0$ and if the needles are orientated at random $\varepsilon' = 1 + \frac{1}{3} f\varepsilon_0$. This value of ε' is for high values of ε_0 much larger than that obtained for spheres.

The second argument has an experimental origin. In the investigations of sintered dielectric materials with high dielectric constant, it was found [4]) that, comparing two samples containing equal volume percentages of solid material (TiO_2), the sample, consisting of highly compressed powder has a lower dielectric constant than the sample, which had been obtained by a sintering process. We believe that this effect of sintering can be explained in the following way. In the case of the compressed powder the air is dispersed as wedge-shaped holes between the crystals, which are limited by their natural cleaving faces. In the case of the intensively sintered material the holes may be supposed to have assumed a more or less spherical shape. Now it can be made plausible by arguments similar to those used in the preceding paragraph, that the wedge shaped holes cause a much lower dielectric constant of the mixture than an equal volume percentage of spherical holes would do.

It is the aim of this paper to investigate in more detail the effect of the shape of the particles on the effective permeability. In order to simplify the problem we assume the individual particles or holes to be ellipsoides, which are orientated at random in the medium. With the use of one approximation, which we shall discuss in section 2, a formula for the effective permeability can be derived, in dependence of the shape of the ellipsoides. In the case of spherical particles the formula runs as follows:

$$\frac{\varepsilon' - 1}{3\varepsilon'} = \sum_p f_p \frac{\varepsilon_p - 1}{\varepsilon_p + 2\varepsilon'}, \tag{2}$$

in which f_i and ε_i are the volume fractions and the permeabilities of the individual substances and of the medium. The formula of Böttcher appears to be a special case of our general formula for spherical particles and from this it may be concluded that analogous approximations are used in our derivation and in Böttcher's theory.

A further result of the theory is that the relative deviations from (2) in consequence of the non spherical shape of the particles or holes are the larger the higher the permeability of the solid. For a powder of a solid with $\varepsilon = 4$, the deviations of ε from the value given by (1) may be about 5%. This explains the fair agreement between the theoretical values of ε' and the experimental data on crystal powder with $\varepsilon \sim 4$ as was found by Böttcher. In a solid with $\varepsilon_0 = 100$ however, 20% of empty holes with the shape of very flat ellipsoides may cause a decrease of the dielectric constant to $\varepsilon' = 20$, while 20% of empty spheres would reduce ε to $\varepsilon' = 70$. From this it can easily be understood why the effect of sintering on the effective dielectric constant is only found in materials of high dielectric constant.

It may be emphasized once more that the formulae which will be derived in the next section are not exact. As the consequences of the approximation involved are not precisely known, doubt about the validity is quite justified. Only in the region of small amounts of particles or holes the formulae may be fairly correct. Nevertheless we should like to propose formula (2) as a useful approximation for materials in which the individual particles and holes may be assumed to have a more or less spherical shape, for instance in intensively sintered materials. Further experimental data are wanted to decide whether (2) is applicable indeed.

It must be mentioned that the importance of the shape of the particles has been recognised also by Ollendorf[5]) in a theoretical paper on the effective magnetic permeability of cores of iron dust. We believe, however, that the approximation used in his paper is less adequate. Therefore we shall discuss the discrepancies between Ollendorf's theory and ours in a separate section.

Finally we remark that in the theories given by Böttcher, by Ollendorf and in this paper, there is found no effect of the size of the particles on the effective permeability of the mixtures.

2. *Theory.* We want to carry out the calculations for the dielectric case. The results can be easily translated into the magnetic language.

We assume the material to be placed between the plates of a large condenser, by means of which an electric field is established in the dielectric. The distance d between the plates is small compared with the dimensions of the plates. The electric displacement $\mathbf{D}$, the electric field $\mathbf{E}$ and the dielectric constants ε_p can be defined in the usual way in each particle and in the medium. For the sake of simplicity the individual substances are supposed to be isotropic, so that $\mathbf{D} = \varepsilon_p \mathbf{E}$ holds in each substance. We now consider an element of volume situated somewhere in the middle of the condenser, with dimensions being small in comparison with the distance d between the plates, but large as compared with the size of the individual particles or holes. The effective electric displacement and the effective electric field will be defined as the values of $\mathbf{D}$ and $\mathbf{E}$, averaged over the volume just mentioned. As a result of the experimental arrangement, the effective displacement $\bar{\mathbf{D}}$ and the effective electric field $\bar{\mathbf{E}}$ will be independent of the special choice of the element of volume V and will point in a direction perpendicular to the plates of the condenser. The effective dielectric constant ε' is now defined by

$$\bar{\mathbf{D}} = \varepsilon \bar{\mathbf{E}}. \tag{3}$$

For the effective polarization obviously holds:

$$4\pi \bar{\mathbf{P}} = \bar{\mathbf{D}} - \bar{\mathbf{E}} = (\varepsilon' - 1)\, \bar{\mathbf{E}}. \tag{4}$$

The calculation runs as follows:

$$V\bar{\mathbf{D}} = \int_{V_e} \varepsilon_e \mathbf{E} dv + \sum_j \int_{V_j} \varepsilon_j \mathbf{E} dv = V \varepsilon_e \bar{\mathbf{E}} + \sum_j \int_{V_j} (\varepsilon_j - \varepsilon_e)\, \mathbf{E}\, dv. \tag{5}$$

Here V_j means that part of the volume element, occupied by particles or holes with dielectric constant ε_j, while V_e indicates the remaining part, occupied by the medium with $\varepsilon = \varepsilon_e$. Formula (5) can be written as:

$$\bar{\mathbf{D}} = \varepsilon_e\, \bar{\mathbf{E}} + \sum_j f_j\, (\varepsilon_j - \varepsilon_e)\, \frac{1}{V_j} \int_{V_j} \mathbf{E}\, dv, \tag{6}$$

in which f_j is the volume fraction, occupied by the particles or holes with dielectric constant ε_j. From (6) it can be seen that the problem is to find the mean value of the electric field in the interior of all the particles with dielectric constant ε_j.

An exact solution of this problem does not exist. Therefore we approximate the situation in the following way. We fix our attention to one special particle and assume that this particle with dielectric constant ε_j is surrounded by a medium with dielectric constant ε' (the effective ε of our mixture), in which a homogeneous field $\bar{\mathbf{E}}$ is maintained at large distances from the particle. Now our hypothesis is that *the maen value of the field in the interior of the particle, under these simplified conditions, provides a good approximation for the actual mean field in this special particle.* Even in our simplified representation, an exact calculation of the mean field $\mathbf{E}_m$ in the interior of the special particle is not possible for a particle of an arbitrary shape. It can only be stated that $\mathbf{E}_m$ will be linearly related with $\bar{\mathbf{E}}$ by means of a tensor α:

$$E_m^{(k)} = \sum_l \alpha_{kl}^{(j)}\, (\varepsilon')\, \bar{E}^{(l)}, \quad (k, l = 1, 2, 3). \tag{7}$$

Here the tensor α depends on ε_j, ε' and the shape and the orientation of the particle. If we suppose that the shape of all particles is the same and the orientation is at random, then, according to our hypothesis, the mean value of the field in the interior of all particles with $\varepsilon = \varepsilon_j$ will be fairly approximated by

$$\frac{1}{V_j}\int_{V_j} \mathbf{E}\, dv = \tfrac{1}{3}(\alpha_{11}^{(j)} + \alpha_{22}^{(j)} + \alpha_{33}^{(j)})\, \overline{\mathbf{E}} = \dot{\alpha}^{(j)}(\varepsilon')\, \overline{\mathbf{E}}. \tag{8}$$

In accordance with a well known property of tensors, only the diagonal sum ($3a^{(j)}$) of the tensor $\alpha^{(j)}$ occurs in (8), in consequence of the orientation at random. With (3) and (8) we find the following implicite equation for ε':

$$\varepsilon' = \varepsilon_e + \Sigma f_j(\varepsilon_j - \varepsilon_e)\, a^{(j)}(\varepsilon'). \tag{9}$$

So far we have used only one hypothesis, but it is rather unsatisfactory that it cannot be easily seen what are the consequences of our approximative treatment for the validity of (9). Some aspects of the character of the approximation are discussed in the last section in connection with the theory of Ollendorf. Here we shall prove that for small values of f_j (9) is correct, or more exactly: in a series expansion of ε' into powers of f_j, (9) gives the correct terms of the first power. The proof is the following: In the case of small f_j, a way to find the mean field in a special particle in first approximation, is to assume that exclusively this particle is present in the medium with $\varepsilon = \varepsilon_e$, while the charges on the plates of the condenser remain unchanged. The effect of the other particles on the field in this special one gives a contribution to ε', which is of a higher order in f. The charges on the plates give rise now to a field $\overline{D}/\varepsilon_e$ in the medium at large distances from the particle. In this approximation (7) becomes:

$$E_m^{(k)} = \Sigma\, \alpha_{kl}^{(j)}(\varepsilon_e)\, \frac{\overline{D}^{(l)}}{\varepsilon_e}. \tag{7'}$$

The difference between (7') and (7) is the use of ε_e instead of ε' and gives rise to a deviation in E_m proportional to f. In the series expansion of ε' this deviation does not effect the terms of the first power in f, however, as may easily be seen, from (6) or (9).

3. *Ellipsoidical particles*. With (9) we have reduced the problem to a more simple one, namely the determination of $a^{(j)}$. In order to obtain more insight in the effect of the shape of the particles, we now assume that the particles are ellipsoides. This is the only case in which a closed expression can be given for the tensor α. In an external

field $\bar{E}$ in the direction of the a-axis, the ellipsoide will be homogeneously polarized and (7) runs as follows:

$$E_m^{(a)} = \frac{\bar{E}^{(a)}}{1 + \left(\frac{\varepsilon_j}{\varepsilon'} - 1\right) A_a}. \tag{10}$$

Here A_a is the depolarization factor of the ellipsoide in the direction of the a-axis. The value of A_a of an ellipsoide with semi axes a, b and c is given by the elliptic integral [6])

$$A_a = \frac{abc}{2} \int_0^\infty \frac{du}{(a^2 + u)\sqrt{(a^2 + u)(b^2 + u)(c^2 + u)}}. \tag{11}$$

Further is valid:

$$A_a + A_b + A_c = 1 \tag{12}$$

For a needle shaped ellipsoide with $a = b \ll c$, A_a, A_b and A_c tend to $\frac{1}{2}$, $\frac{1}{2}$ and 0 respectively. For a sphere with $a = b = c$, the three factors are $\frac{1}{3}$, for a disc shaped ellipsoide with $a = b \gg c$, A_a, A_b and A_c tend to 0, 0 and 1 respectively.

With the aid of (10) we can rewrite (9) into the form:

$$\varepsilon' = \varepsilon_e \left[1 - \frac{1}{3} \sum_j \left\{ f_j (\varepsilon_j - \varepsilon_e) \sum_{i=1}^{3} \frac{1}{\varepsilon' + (\varepsilon_j - \varepsilon') A_i} \right\} \right]^{-1}. \tag{13}$$

The formula should be applicable on media with dielectric constant ε_e, containing ellipsoidical particles or holes with dielectric constant ε_j.

4. *Discussion. a.* Spheres. In the case of spherical particles or holes, $A_i = \frac{1}{3}$ and it is not difficult to write (13) as

$$\frac{\varepsilon' - 1}{3\varepsilon'} = \sum_p f_p \cdot \frac{\varepsilon_p - 1}{\varepsilon_p + 2\varepsilon'}. \tag{14}$$

Here p indicates the different substances in the spheres, as well as the medium, thus p includes the index e together with all indices j. It is remarkable that according to (14), the effective dielectric constant should depend only on the volume percentages of the different substances and should be independent of the choice of the substance which plays the role of the medium between the spheres. If we have to deal with the mixture of one solid substance with air, we find

Böttcher's formula (2), in which f is always the volume percentage of the solid substance. According to our theory, the formula should be valid for a mixture of solid spherical particles with air as well as for the solid substance, containing spherical holes.

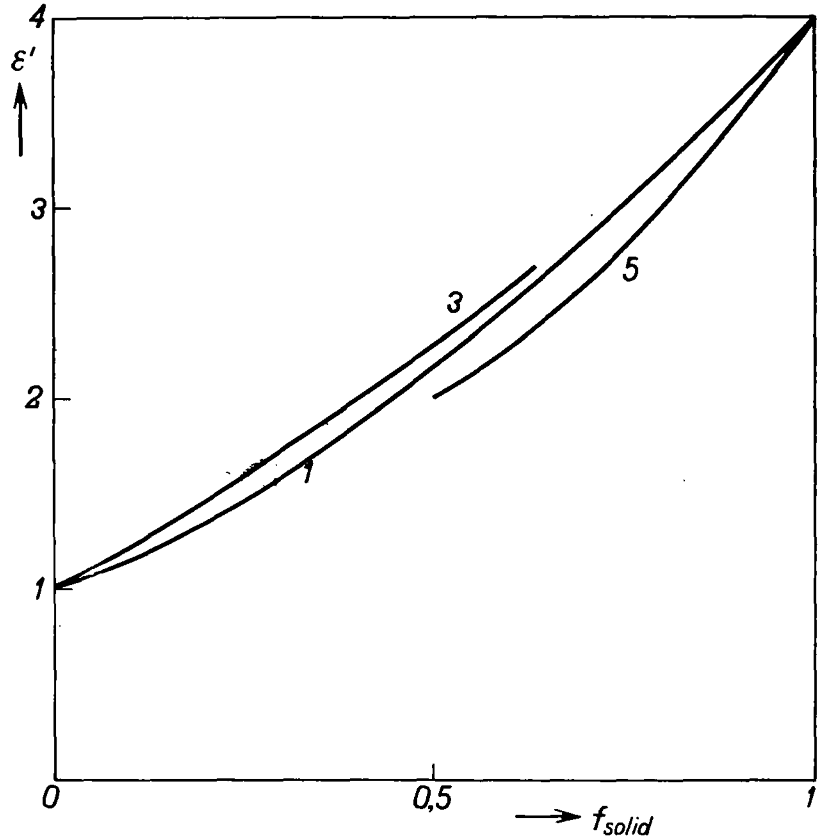

Fig. 1. Effective dielectric constant of a mixture of vacuum and a solid substance with $\varepsilon = 4$ as a function of the composition.
1 spherical particles in vacuum or spherical holes in the solid substance;
3 disc shaped particles in vacuum;
5 disc shaped holes in solid substance.
In the cases 3 and 5, $\delta \ll 1/\varepsilon_{solid}$. The curves for needle shaped particles in vacuum and needle shaped holes in solid substance are not given in this figure, as they are practically identical with curve 1.

In figures 1 and 2, we have plotted the values of ε', computed with (2), versus f for mixtures of air and a solid substance with $\varepsilon = 4$, and $\varepsilon = 100$ respectively. In the case of a mixture of two substances with $\varepsilon_2/\varepsilon_1 \to \infty$, we find for spherical particles:

$$\frac{\varepsilon'}{\varepsilon_1} = (1 - 3f_2)^{-1} \text{ for } f_2 < \tfrac{1}{3}$$
$$\frac{\varepsilon'}{\varepsilon_2} = \tfrac{1}{2}(3f_2 - 1) \text{ for } f_2 > \tfrac{1}{3}. \tag{15}$$

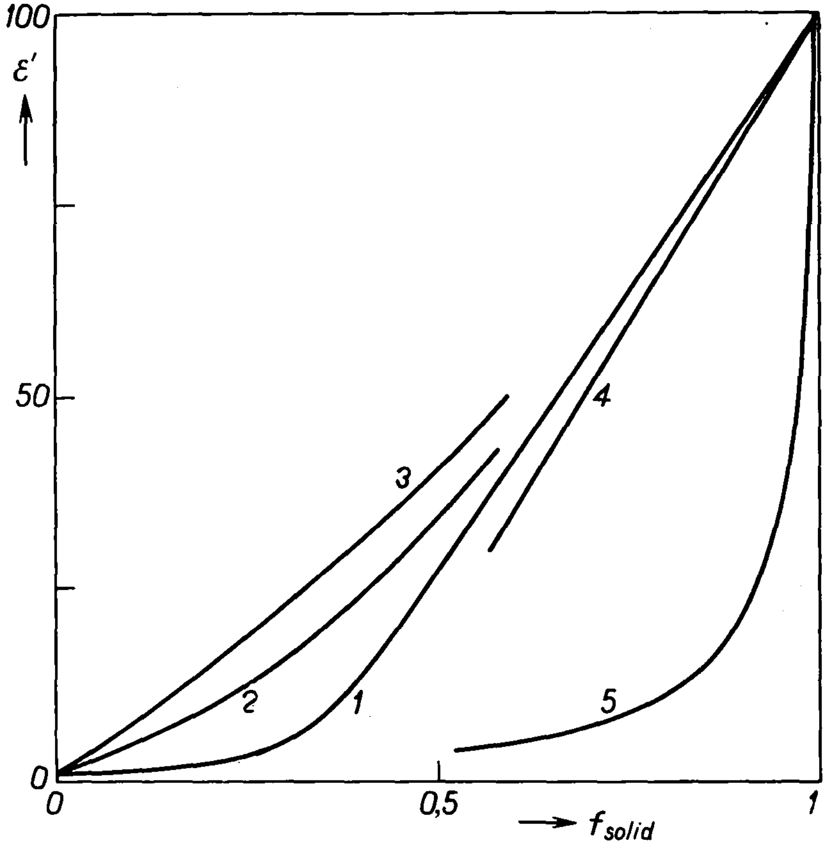

Fig. 2. Effective dielectric constant of a mixture of vacuum and a solid substance with $\varepsilon = 100$ as a function of the composition.

1 spherical particles in vacuum or spherical holes in the solid substance;
2 needle shaped particles in vacuum;
3 disc shaped particles in vacuum;
4 needle shaped holes in solid substance;
5 disc shaped holes in solid substance.
For the cases 2, 3, 4 and 5, $\delta \ll 1/\varepsilon_{solid}$.

In the following part of this section we shall confine ourselves to cases where only two substances are present, a medium with $\varepsilon = \varepsilon_e$ and particles or holes with $\varepsilon = \varepsilon_j$, while f will indicate now always the volume percentage of the particles or holes. First we shall give a formula, valid for spheres, if f is small:

$$\varepsilon' = \varepsilon_e \left[1 + 3f \, \frac{\varepsilon_j - \varepsilon_e}{\varepsilon_j + 2\varepsilon_e} \right]. \tag{16}$$

The formula is exact, as long as higher powers of f may be neglected.

Further we shall discuss the effect of deviations from the spherical shape. It can be demonstrated without difficulty with the aid of (12) and (13) that the deformation of the spheres into ellipsoides of any shape, always causes an increase of ε', if $\varepsilon_e < \varepsilon_j$ and a decrease of ε', if $\varepsilon_e > \varepsilon_j$; provided that f remains constant. This is correct for all values of f. In connection with this extremum property of the sphere, it may be understood that not too large deviations from the spherical shape have only a slight effect on the effective dielectric constant. As soon as the shape of the ellipsoides is more like that of a needle or a disc, large effects may be expected. This will be treated in the next sub-sections.

b) Needles. We consider ellipsoides with axes $a = b \ll c$, so that the depolarization factors can be assumed to be $\frac{1}{2} - \delta$, $\frac{1}{2} - \delta$ and 2δ respectively ($\delta \ll 1$). For the sake of simplicity we discuss only the limiting case of small values of f. For small values of δ and f (13) becomes approximately:

$$\varepsilon' = \varepsilon_e \left| 1 + \frac{f}{3} (\varepsilon_j - \varepsilon_e) \left\{ \frac{4}{\varepsilon_j + \varepsilon_e} + \frac{1}{\varepsilon_e + (\varepsilon_j - \varepsilon_e) \, 2\delta} \right\} \right]. \tag{17}$$

In the case $\varepsilon_e \gg \varepsilon_j$ (holes in solid substance), ε' becomes approximately

$$\varepsilon' = \varepsilon_e \left(1 - \tfrac{5}{3} f\right). \tag{18}$$

Here the relative decrease of ε is proportional to the part of the volume occupied by the holes.

In the case $\varepsilon_j \gg \varepsilon_e$, δ may be so small that even $2\delta < \varepsilon_e/\varepsilon_j$ and we find the approximation:

$$\varepsilon' = \varepsilon_e + \frac{f}{3} \varepsilon_j. \tag{19}$$

If, however, for a fixed small value of δ, $\varepsilon_e/\varepsilon_j$ becomes much smaller than 2δ, we find approximately

$$\varepsilon' = \varepsilon_e\left[1 + \frac{f}{3}\left(4 + \frac{1}{2\delta}\right)\right]. \tag{20}$$

Evidently, the relative increase of ε can be greatly enlarged by the decrease of δ, as long as $2\delta > \varepsilon_e/\varepsilon_j$, but it will never surpass the value indicated by (19).

c. Discs. We consider ellipsoides with axes $a = b \gg c$, so that the depolarization factors can be assumed to be δ, δ and $(1 - 2\delta)$ respectively. Again we discuss only the limiting cases of small values of f. In the case $\varepsilon_j \gg \varepsilon_e$, the ε' can be enlarged by the decrease of δ, as long as $\delta \gg \varepsilon_e/\varepsilon_j$, according to the approximate formula:

$$\varepsilon' = \varepsilon_e\left[1 + \frac{f}{3}\left(1 + \frac{2}{\delta}\right)\right], \tag{21}$$

but ε' will not surpass the value

$$\varepsilon' = \varepsilon_e + \tfrac{2}{3} f\, \varepsilon_j, \tag{22}$$

which is valid for $\delta \ll \varepsilon_e/\varepsilon_j \ll 1$.

In the case $\varepsilon_e \gg \varepsilon_j$ the relative decrease of ε can be enlarged by the decrease of δ, as long as $2\delta \gg \varepsilon_j/\varepsilon_e$, according to the formula:

$$\varepsilon' = \varepsilon_e\left[1 - \frac{f}{3}\left(2 + \frac{1}{2\delta}\right)\right], \tag{23}$$

but ε' will not be smaller than:

$$\varepsilon' = \varepsilon_e\left[1 - \frac{f}{3}\left(2 + \frac{\varepsilon_e}{\varepsilon_j}\right)\right], \tag{24}$$

which formula holds for $2\delta \ll \varepsilon_j/\varepsilon_e \ll 1$.

A special feature of the last expression is that the relative decrease of ε is the larger, the higher the dielectric constant of the solid medium, provided that $2\delta < \varepsilon_j/\varepsilon_e$. This makes understandable why exactly in materials of high dielectric constant the decreasing effect of flat, wedge like holes on ε, is so important.

In figures 1 and 2 we have given curves for ε' in the case of needles and discs, computed with the aid of formula (13).

5. *Comparison with other theories.* In this section we want to investigate the differences between the theory given in this paper and

the theory of Ollendorf [5]). The discussion will give rise to some considerations of more general character, in connection with the theories of Onsager and Böttcher. We discuss only the case of identical spherical particles in air, as our arguments are essentially the same for the case of ellipsoidical particles, which is also treated in Ollendorf's paper. The approximation used by Ollendorf is based on the following. Each particle is assumed to be polarized in consequence of a "polarizing" field $\mathbf{E}_p$. In order to define $\mathbf{E}_p$ for one special particle, we imagine the particle to be removed out of the mixture, while the charges on the plates of the condenser and the polarization of the other particles and the medium remain fixed. The field which now appears in the spherical hole, is the polarizing field for this special particle. The mean polarization per cm^3 $\mathbf{P}_m$ in the particle will be exclusively determined by the space-average of $\mathbf{E}_p$ over the hole, denoted by $\mathbf{E}_{pm}$. The well known formula:

$$4\pi\, \mathbf{P}_m = \mathbf{E}_{pm} \frac{3\,(\varepsilon_i - 1)}{\varepsilon_i + 2} \tag{25}$$

is valid here.

The effective polarization of the mixture $\overline{\mathbf{P}}$ is now given by:

$$4\pi\, \overline{\mathbf{P}} = 4\pi\, f\, \overline{\mathbf{P}}_m\,(Av) = f\, \mathbf{E}_{pm}\,(Av)\, \frac{3\,(\varepsilon_i - 1)}{\varepsilon_i + 2}\,, \tag{26}$$

where (Av) means the averaged value over all particles. When $E_{pm}(Av)$ can be expressed in terms of the mean field $\overline{E}$ and the effective dielectric constant ε', we find an implicite equation for ε' and the problem is solved. It will be seen that the relation between $E_{pm}(Av)$ and $\overline{E}$, used by Ollendorf, is different from the relation that must be assumed in order to obtain the results of our theory.

In the case of a complete disordered arrangement of the particles, it is correct to replace the average of E_{pm} over all particles by the E_{pm} of one particle, averaged over all possible configurations of the other particles in the medium around this special one. This process of averaging is of a typical statistical character and the problem has been investigated by Kirkwood [7]). In consequence of the mutual interaction of the polarized particles (or the medium) there exists a statistical correlation between the polarization of the particles (or the medium) and their place with respect to the special particle we were considering. Even for a somewhat simplified model, such as used in Kirkwood's paper, it is impossible to handle this correlation in such a way that practical conclusions can be drawn for the effective dielectric constant of mixtures. Moreover, there is another feature in the problem, which is a serious obstacle for the exact treatment of the problem. Consider a point in the dielectric, situated at a distance from the special particle, being large as compared with the dimensions of the particles. The probability to find a substance with a definite ε_j at this point, is given by the corresponding filling factor f_j. In the immediate surrounding of our special particle, however, there will be a relatively larger probability for the presence of the medium. This is a direct result of the "steric hindrance" of the particles with finite dimensions. For these reasons one must take recourse for some approximation in order to calculate the value of ε'.

The most crude approximation that can be made is to neglect the statistical correlation mentioned above. For the averaged polarizing field we find then the field caused by the charges on the plates of the condenser, to which is added the field caused by an averaged polarization $\overline{\mathbf{P}}$, homogeneously distributed over the whole space of the condenser around our special spherical particle. In this case $\mathbf{E}_{pm}(Av)$ is simply the homogeneous field given by the well known expression of Lorentz:

$$\mathbf{E}_{pm}(Av) = \frac{\varepsilon' + 2}{3} \overline{\mathbf{E}}. \tag{27}$$

It can easily be seen that the effect of steric hindrance is of no importance here. It results in a change $\Delta\mathbf{P}$ of the assumed averaged polarization $\overline{\mathbf{P}}$ in the neighbourhood of the special particle and as $\Delta\mathbf{P}$ only depends on the radial distance from the particle, it does not effect the polarizing field. The approximation in which all statistical correlations are neglected was used by Ollendorf for spherical particles as well as for ellipoidical particles. For the resulting formulae for ε' we refer to the original paper.

As the statistical correlation is the more important, the higher the dielectric constant and the filling factors, we have to look for a method which accounts for it in some way. It can be demonstrated in connection with Kirkwood's theory that the procedures used by Onsager[1]) or Böttcher, fulfill this requirement to a certain extent [8]).

In the procedure of Böttcher the dielectric around the special spherical particle is schematized by a substance with a homogeneous dielectric constant ε' and thus the effects of steric hindrance are neglected at once. For the polarizing field is written the sum of two homogeneous fields, the field E_h, which is established in an empty spherical hole in a medium with dielectric constant ε':

$$\mathbf{E}_h = \frac{3\varepsilon'}{2\varepsilon' + 1} \overline{\mathbf{E}}. \tag{28}$$

and the field E_r, caused in this hole by the extra polarization of the medium due to the introduction of a sphere with polarization $\mathbf{P}_m(Av)$, fitting exactly in the hole:

$$\mathbf{E}_r = \frac{4\pi}{3} \mathbf{P}_m(Av) \frac{2(\varepsilon' - 1)}{2\varepsilon' + 1}. \tag{29}$$

Thus:

$$\mathbf{E}_{pm}(Av) = \mathbf{E}_h + \mathbf{E}_r. \tag{30}$$

The electrostatic problem described with the formulae (30), (29), (28) and (26) is equivalent to the problem we met in section (2), in which a particle (in this case a sphere) with $\varepsilon = \varepsilon_i$, was surrounded by medium with $\varepsilon = \varepsilon'$, in which a field $\overline{\mathbf{E}}$ was maintained at large a distance from the particle. It can easily be shown that formulae (30), (29), (28) and (26) lead to formula (1). Obviously, the procedure of Böttcher and our fundamental hypothesis, put forward in section 2, are equivalent for the case of identical spherical particles. From this it will be clear that the differences between Ollendorf's theory and ours may be formally ascribed to the difference in the equations (27) and (30) in the case of spherical particles, or generally speaking to the the different assumption concerning the relation between the polarizing field $E_{pm}(Av)$ and the effective field $\overline{E}$.

The considerations in this section may have shown some aspects of our fundamental hypothesis, in connection with other theories, which could not be put forward in section 2.

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