Conductivity of random sphere packings: Effects of a size distribution

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Experimental results for the formation factor of packings of spheres with two well defined sizes, in a conducting fluid, are compared with an equation of Berryman [Phys. Rev. B **27**, 7789 (1983)]. When Berryman's formula is used twice, in a repeated-mixing fashion, we obtain good agreement with the experimental data. The procedure can be generalized to wide grain size distributions and then constitutes a justification for Archie's law.

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I. INTRODUCTION

The electrical properties of porous solids, impregnated with a liquid, are of interest in many fields of research. For brine-impregnated sedimentary rocks, Archie [1] found the following empirical relation:

$$\sigma_{\rm app}=\sigma_f a\varphi^m \ . \quad (1)$$

Here, $\sigma_{\rm app}$ is the apparent conductivity of the medium, σ_f the conductivity of the pore fluid, φ the porosity, and a and m constants. This expression has accordingly been named "Archie's law." It was originally assumed that the prefactor a equals one, but it was later found that this is not always the case. The ratio $\sigma_f/\sigma_{\rm app}$ is called the formation factor F (assuming negligible effects from surface conduction).

In several cases, model systems of so-called artificial rocks have been studied. Nettelblad *et al.* [2] found for artificial "rocks" made of sand grains glued with small amounts of epoxy that $m\approx 1$ and that a is dependent on the grain size distribution; the wider the distribution, the higher the value of a. Wong, Koplik, and Tomanic [3], and Holwech and Nøst [4] found for sintered monosize glass spheres that $m\approx 2.3$ and $a\approx 0.3$. Their values of m are similar to those of Nettelblad [5], who measured on sintered, multisize, nonspherical polypropylene grains. However, he obtained a higher value of a, which appeared to increase with widening grain size distribution.

Calculations of electrical conductivity for spherical inclusions in regular arrays have been performed [6–8]. For the case of a porous material consisting of random arrays of equal spheres, Berryman [9] used the concept of tortuosity to obtain the approximate formula

$$\sigma_{\rm app}=\sigma_f\frac{\varphi(1+\varphi)}{2} \ . \quad (2)$$

This formula gives results that are close to the results from calculations for spheres in cubic arrays [7,8]. Effective-medium theories are often used to characterize disordered materials. As an example, the theory of Maxwell Garnett [10] yields for spherical insulator inclusions in a host medium

$$\sigma_{\rm app}=\sigma_f\frac{2\varphi}{3-\varphi} \ . \quad (3)$$

Note that for $\varphi\approx 1$, this expression gives asymptotically the same result as Eq. (2). For low φ values, such as for a close-packed fcc lattice, Eq. (2) is a better approximation to the exact value [8]. Another important model is the Bruggeman asymmetric theory. In this theory, one starts with the pure host material, and adds an infinitesimal amount of guest material in several stages. For each addition, one uses the Maxwell Garnett formula to calculate $\sigma_{\rm app}$, however, with σ_f determined by a similar calculation at the previous stage. Integration then yields for spherical insulating inclusions in a conducting medium

$$\sigma_{\rm app}=\sigma_f\varphi^{1.5} \ . \quad (4)$$

Other exponents are obtained if, e.g., spheroids are used [11,12]. Note that Jackson, Smith, and Stanford [13], when measuring unconsolidated sands of different grain shapes, found that the exponent in Archie's law increased as the grains became less spherical. Lemaître *et al.* [14] performed measurements on packs of spheres of two different sizes. The porosity was varied by using different proportions of "small" and "large" spheres. Two different sphere diameter ratios were used; the large spheres having 11 times larger diameter than the small ones and a second set with the diameter ratio 4. The results on the conductivity did not show good agreement with the Berryman formula [Eq. (2)], except for the highest porosities (i.e., for single-size sphere packings). In this paper, we show how a repeated-mixing procedure, using the Berryman formula, can give much better agreement with experimental results. We then extend the argument to wide distributions of grain sizes in order to give an argument for Archie's law.

II. PROPOSED THEORY AND COMPARISON WITH EXPERIMENT

Lemaître *et al.* [14] used different volume fractions of small spheres x (volume of small spheres divided by total volume of spheres). For each of the volume fractions used, they obtained different porosities φ_{tot} and different formation factors F. In order to model this situation, we used a repeated-mixing argument with two stages. We first consider the conductivity of a medium consisting of the small spheres and the pore liquid. The porosity of this "medium" φ_{med} is

$$\varphi_{\text{med}} = \frac{\varphi_{\text{tot}}}{\varphi_{\text{tot}} + x(1-\varphi_{\text{tot}})} . \tag{5}$$

The conductivity of this medium σ_{med} is obtained from Eq. (2). We now assume that the medium is approximately homogeneous on the length scale of the large spheres, which are considered to be inclusions in this medium. At this second stage we can apply Eq. (2) a second time. Now, we take σ_{med} to be the "host" conductivity and the porosity is replaced with the volume fraction of the small-sphere–pore-liquid medium, namely,

$$\varphi_{\text{eff}} = \varphi_{\text{tot}} + x(1-\varphi_{\text{tot}}) . \tag{6}$$

Finally, we obtain for the conductivity:

$$\sigma_{\text{app}} = \frac{\sigma_f}{4} \frac{\varphi_{\text{tot}}[2\varphi_{\text{tot}} + x(1-\varphi_{\text{tot}})][1+\varphi_{\text{tot}} + x(1-\varphi_{\text{tot}})]}{\varphi_{\text{tot}} + x(1-\varphi_{\text{tot}})} . \tag{7}$$

It should be noted that φ_{tot} and x are not independent variables in the case we consider, but related because of the procedure for sample preparation. Thus, Eq. (7) can be interpreted as giving a unique relation between σ_{app} and x. Yet, the variable φ_{tot} as a function of x displays a minimum [14], and a given φ_{tot} thus corresponds to two different values of x. One could thus surmise that a σ_{app} versus φ_{tot} curve would show two different branches. Still, the experimental data [14] do not display any clear division into two branches, and, as is shown in our Fig. 1, nor do the data computed from Eq. (7).

In Fig. 1, we show the formation factor data of Lemaître *et al.* for the sphere diameter ratio 11 as a function of porosity. We include the predictions of the Berryman formula as well as the predictions using the "repeated Berryman formula." The agreement between the predictions using "repeated Berryman" and experiments is good, considering the crudity of the model. The agreement is especially good at high and low porosities, and less good in the intermediate range. The good agreement at high porosities is not surprising, since in this case only one sphere size is present and also the Berryman Eq. (2) yields good agreement. Also our equation describes the behavior at low porosities much better than the simple Berryman relation. Still, the ratio 11 between the two sphere sizes may be insufficient to render the "medium" consisting of small spheres and pore fluid homogeneous on the length scale of the large spheres. Especially at the interstices between several touching large spheres, the packing of small spheres is severally influenced by the configuration of the large spheres. This possible deviation from homogeneity may explain the discrepancies between theory and experiment seen in Fig. 1.

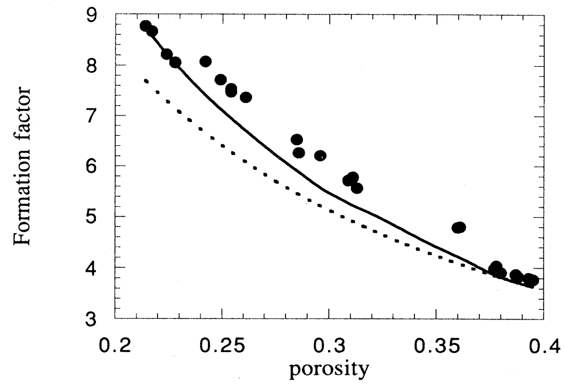

FIG. 1. The formation factor as a function of porosity for experimental data (circles), the Berryman formula, Eq. (2) (dotted line), and the repeated Berryman formula suggested in the article (solid line).

III. IMPLICATIONS FOR ARCHIE'S LAW

We now generalize the above repeated-mixing procedure in order to explain Archie's law. Let us start with the assumption that a particular sedimentary rock has a very wide range of grain sizes, and that the grain shape does not depend on the grain size. We then assume that the formation factor depends on the porosity solely according to some function:

$$F = f(\varphi) . \tag{8}$$

Consider starting with a finite, but very wide grain size distribution, with porosity φ_{initial}, and then mixing this material with another grain size distribution, also wide, but with all grains much smaller than the grains in the initial distribution. Inside the original pore space, we now have a porous material with porosity φ_{med}. The resulting porous material then has the porosity $\varphi_{\text{initial}}\varphi_{\text{med}}$, and it should thus have the formation factor $f(\varphi_{\text{initial}}\,\varphi_{\text{med}})$ (as we assumed that the formation factor only depended on the porosity). On the other hand, the medium consisting of smaller grains and pore liquid has the formation factor $f(\varphi_{\text{med}})$, and, as the original formation factor was $f(\varphi_{\text{initial}})$, the repeated-mixing argument yields the following relation for the function f:

$$f(\varphi_{\text{initial}})f(\varphi_{\text{med}}) = f(\varphi_{\text{initial}}\varphi_{\text{med}}) . \tag{9}$$

It is easily seen that, except for the physically uninteresting solution $f(\varphi)\equiv 0$, the only possible solution to this equation is

$$f(\varphi) = \varphi^{-m} . \tag{10}$$

For physical reasons, we can discard the solutions $m \le 0$ [the notation $-m$ is used to conform with Eq. (1)]. It should be noted that Bruggeman's asymmetric theory can be derived from a repeated-mixing argument [15]. Hence, Eq. (4) is a special case of Eq. (10). The main difference between the methods of derivation is that the derivation of the Bruggeman theory assumes that the effect of the addition of a small amount of solid material can be described by effective-medium theory, also for high volume fractions of the solid. We are less restrictive about the possible choices for $f(\varphi)$. Still, our argument thus yields the Archie equation as the only possible solution, for a wide enough grain size distribution, if the formation factor solely depends on the porosity. It is possible, though, that the wide size distribution is a sufficient, rather than necessary, condition for Archie's law. This assertion is supported by the fact that the special case, Eq. (4), can also be derived from a randon unit cell argument, as shown elsewhere [16].

We also note that our argument for Archie's law demands that the prefactor a be one. However, the exponent m is not determined from this argument, and it is probably dependent on the grain shape (as discussed in Sec. I). Atkins and Smith [17] showed that a mixture of two grain sizes (having a large difference in size), where samples of each grain obey Archie's law [Eq. (1)] with a equal to unity but having different values of the exponent m, still could obey Archie's law, but with a prefactor different from unity. A value of the prefactor different from one may thus be an indication that the grain shape is dependent on the grain size.

IV. CONCLUSIONS

We have shown that the Berryman expression for the electrical formation factor, $\sigma_{\mathrm{app}}=\sigma_f\varphi(1+\varphi)/2$, gives good agreement with experimental results for binary mixtures of spheres, if it is used in a repeated-mixing procedure. If such a procedure is generalized to very wide grain size distributions, under the assumption that the formation factor is only dependent on the porosity, Archie's law (with prefactor one) is the inevitable result.

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