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## A NEW BOUND ON THE ABSORPTION COEFFICIENT OF A TWO-PHASE MEDIUM

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The Doi bound on the effective absorption coefficient of a random two-phase medium is revisited in this brief note. Defects are created in one of the constituents, being absorbed by the other one, which thus act as a perfect sink. Use is made of a variational principle due to Rubinstein and Torquato. The trial fields generalize the ones, originally proposed by Doi, and hence the new bound is more restrictive than the original Doi's one for an arbitrary medium. In the particular case of an array of nonoverlapping array of spherical sinks, the new bound however coincides with Doi's and with the one, derived by Talbot and Willis. In passing, besides the known "particle-particle" bound, a curious new "surface-surface" bound is extracted. Though a bit weaker than the Doi's, this bound relies only upon the two-point "surface" statistics. In the dilute case it reproduces the classical Smoluchowski result.

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Consider a two-phase random medium, consisting of a phase '1', immersed into an unbounded matrix (phase '2'). The medium is assumed statistically homogeneous and isotropic. Let a species (defects) be generated at the rate  $K$  within the phase '2' (matrix) occupying the region  $\mathcal{K}_2$ . It is absorbed by the "sink" phase '1' in the region  $\mathcal{K}_1 = \mathbb{R}^3 \setminus \mathcal{K}_2$ . In the steady-state limit the concentration of the defects  $c(x)$  is governed by the well-known equations

$$\Delta c(x) + K = 0, \quad x \in \mathcal{K}_2, \quad c(x) \Big|_{\partial \mathcal{K}_2} = 0. \quad (1)$$

The creation of defects is *exactly* compensated by their removal from the sinks

$$k^* \langle c(x) \rangle = K(1 - \eta_1). \quad (2)$$

(The brackets  $\langle \cdot \rangle$  denote ensemble averaging.) The rate constant  $k^{*2}$  is the so-called *effective absorption coefficient* (or *the sink strength*) of the medium. Its evaluation and bounding for special kinds of random constitution and, above all, for random dispersion of spheres, have been the subject of numerous works, starting with the classical studies of Smoluchowski [1], see, e.g., [2–7], the survey [8], and the references therein. (Note that we have added the factor  $1 - \eta_1$  in (2), due to the fact that in the case under study defects are created *only* within the phase ‘2’, see [9] for a discussion.)

We shall confine the analysis to variational bounding of the sink strength  $k^{*2}$ , taking into account the two-point statistical information concerning medium constitution. The basic tool to be employed to this end is the variational principle of Rubinstein and Torquato (R-T) [6]. The principle states that in the class of statistically homogeneous trial fields such that

$$\mathcal{A} = \left\{ u(x) \mid \Delta u(x) + K = 0, x \in \mathcal{K}_2 \right\}, \quad (3)$$

the following inequality holds:

$$k^{*2} \geq \frac{K^2(1 - \eta_1)}{\langle I_2(x) |\nabla u(x)|^2 \rangle}. \quad (4)$$

Moreover, the equality sign in (4) is achieved if  $u(x) = c(x)$  is the actual field that solves the problem (1).

Since  $\langle I_2(x) |\nabla u(x)|^2 \rangle \leq \langle |\nabla u(x)|^2 \rangle$ , another bound follows from (4):

$$k^{*2} \geq \frac{K^2(1 - \eta_1)}{\langle |\nabla u(x)|^2 \rangle}, \quad (5)$$

see [6]. Though weaker than (4), the evaluation of the bound (5) is simpler, because it obviously employs a smaller amount of statistical information.

Consider the trial fields

$$u(x) = -\frac{K}{\eta_1} \int G(x - y) \left( \lambda I_1(y) - \eta_1 + \frac{\mu \eta_1}{S} |\nabla I_1(y)| \right) dy, \quad (6)$$

where  $G(x) = 1/(4\pi|x|)$  and the (nondimensional) constants  $\lambda, \mu$  are adjustable. Since  $I_1(x)$  is the characteristic function of  $\mathcal{K}_1$ ,  $|\nabla I_1(x)|$  is  $\delta$ -function, concentrated on the interphase boundary. In turn, the quantity  $S$  in (6) is the so-called mean surface, defined as  $S = \langle |\nabla I_1(x)| \rangle$ .

Since  $\Delta G(x) + \delta(x) = 0$ , one has  $\Delta u(x) + K = 0$  if  $x \in \mathcal{K}_2$ . This means that the fields  $u(x)$  in (6) are indeed admissible,  $u(x) \in \mathcal{A}$ .

Consider now the quantity of central importance

$$\mathcal{U} = \langle |\nabla u(x)|^2 \rangle / K^2 \quad (7)$$

that enters the estimate (5). For the latter to be finite, and hence to produce a nontrivial lower bound (5), it is necessary that the integrand in (6) have a zero mean value. This implies

$$\lambda + \mu = 1, \quad (8)$$

since  $\langle I_1(y) \rangle = \eta_1$ .

Note that the class of trial fields (6) generalizes the one, proposed by Doi himself [2]:

$$u(x) = K \int G(x-y) \left( I_2(y) + \xi |\nabla I_1(x)| \right) dy. \quad (9)$$

The condition that mean value of the integrand in (9) vanishes reads

$$\eta_2 + \xi S = 0, \quad (10)$$

so that, as pointed out in [6, 7], there is no place for optimization with respect to  $\xi$ , as envisaged originally by Doi [2].

A simple check shows that our fields (6) reproduce the Doi's one (9) for a special choice of  $\lambda$ , namely, for

$$\lambda = \eta_1, \quad \mu = 1 - \eta_1, \quad \xi = \mu/S, \quad (11)$$

cf. Eq. (8).

With Eq. (8) taken into account, the class (6) is recast as

$$\begin{aligned} u(x) &= -\frac{K}{\eta_1} \int G(x-y) \left\{ \lambda I_1'(y) + \frac{\mu \eta_1}{S} \left( |\nabla I_1(x)| - S \right) \right\} dy, \\ \langle I_1'(y) \rangle &= 0, \quad \langle |\nabla I_1(x)| - S \rangle = 0, \\ I_1'(y) &= I_1(y) - \eta_1, \end{aligned} \quad (12)$$

so that both random variables in the right-hand side of (11)<sub>1</sub> are fluctuations. Then the needed quantity  $\mathcal{U}$ , cf. (7), becomes

$$\mathcal{U} = \frac{a^2}{\eta_1^2} \left( \lambda^2 \theta_1^{\text{pp}} + 2\lambda\mu\eta_1 \theta_1^{\text{ps}} + \mu^2 \eta_1^2 \theta_1^{\text{ss}} \right) \quad (13)$$

after an appropriate integration by parts. Here

$$\begin{aligned} \theta_1^{\text{pp}} &= \int_0^\infty \rho F^{\text{pp}}(\rho) d\rho, \\ \theta_1^{\text{ps}} &= \int_0^\infty \rho F^{\text{ps}}(\rho) d\rho, \\ \theta_1^{\text{ss}} &= \int_0^\infty \rho F^{\text{ss}}(\rho) d\rho \end{aligned} \quad (14)$$

are the first moments on the semiaxis  $(0, \infty)$  of the “particle-particle,” “particle-surface,” and “surface-surface” (two-point) correlation functions, respectively, defined here as follows:

$$\begin{aligned} F^{\text{pp}}(\rho) &= \langle I_1'(x) I_1'(0) \rangle, \\ F^{\text{ps}}(\rho) &= \frac{1}{S} \langle I_1'(x) (|\nabla I_1(0)| - S) \rangle, \\ F^{\text{ss}}(\rho) &= \frac{1}{S^2} \langle (|\nabla I_1(x)| - S) (|\nabla I_1(0)| - S) \rangle. \end{aligned} \quad (15)$$

The multipliers  $1/S$  and  $1/S^2$  have been added in the definitions (14) in order to make the respective correlations dimensionless. In Eq. (14)  $\rho = r/a$ ,  $r = |x|$ , where  $a$  is a certain characteristic length for the phase ‘1’, for example, the mean size of the sinks. If the sinks are identical spheres — a case to be specially discussed below — then obviously  $a$  is to be identified with their radius.

In virtue of (8) and (13) we have

$$\mathcal{U} = \mathcal{U}(\lambda) = \frac{a^2}{\eta_1^2} \left\{ \lambda^2 (\theta_1^{\text{PP}} - 2\eta_1\theta_1^{\text{PS}} + \eta_1^2\theta_1^{\text{SS}}) + 2\lambda\eta_1 (\theta_1^{\text{PS}} - \eta_1\theta_1^{\text{SS}}) + \eta_1^2\theta_1^{\text{SS}} \right\}. \quad (16)$$

Optimizing (15) with respect to  $\lambda$  gives the estimate

$$k^{*2}a^2 \geq k_{\text{N}}^{*2}a^2, \quad k_{\text{N}}^{*2}a^2 = (1 - \eta_1) \frac{\theta_1^{\text{PP}} - 2\eta_1\theta_1^{\text{PS}} + \eta_1^2\theta_1^{\text{SS}}}{\theta_1^{\text{PP}}\theta_1^{\text{SS}} - (\theta_1^{\text{PS}})^2}. \quad (17)$$

This is our generalization of Doi’s bound. The latter shows up if  $\lambda = \eta_1$  in  $\mathcal{U}$ , as given in (16):

$$k^{*2} \geq k_{\text{D}}^{*2}, \quad k_{\text{D}}^{*2}a^2 = \frac{1 - \eta_1}{\theta_1^{\text{PP}} + 2\eta_2\theta_1^{\text{PS}} + \eta_2^2\theta_1^{\text{SS}}}. \quad (18)$$

It is clear that (17) always improves upon Doi’s bound (18), since we have allowed for  $\lambda$  to be adjustable. The bounds (17) and (18) will coincide only if the optimal  $\lambda$ , that minimizes  $\mathcal{U}(\lambda)$  in (16), is *exactly*  $\eta_1$ . The latter is the case if the moments (14) for a given random constitution are, by chance, interconnected as follows:

$$\theta_1^{\text{PP}} + (1 - 2\eta_1)\theta_1^{\text{PS}} - \eta_1\eta_2\theta_1^{\text{SS}} = 0. \quad (19)$$

It is to be noted that, besides “Doi’s choice”  $\lambda = \eta_1$ , two more particular values of  $\lambda$  deserve a special attention.

The first choice is  $\lambda = 1$ . Then  $\mathcal{U} = a^2\theta_1^{\text{PP}}$  and Eqs. (5) and (7) yield

$$k^{*2} \geq k_{\text{PP}}^{*2}, \quad k_{\text{PP}}^{*2}a^2 = \frac{\eta_1^2(1 - \eta_1)}{\theta_1^{\text{PP}}}. \quad (20)$$

This is a known bound, called by Torquato and Rubinstein [5] “particle-particle.” The reason behind this term is clear — the evaluation of  $k_{\text{PP}}^{*2}$  requires only the statistical information incorporated into the “particle” correlation function  $F^{\text{PP}}(\rho)$ .

The second choice of interest is  $\lambda = 0$ . Then  $\mathcal{U} = a^2\theta_1^{\text{SS}}$ , see (15), and Eqs. (5) and (7) yield

$$k^{*2} \geq k_{\text{SS}}^{*2}, \quad k_{\text{SS}}^{*2}a^2 = \frac{1 - \eta_1}{\theta_1^{\text{SS}}}. \quad (21)$$

This is a new bound, which is natural to be called “surface-surface.” The reason for this term is again clear. The evaluation of  $k_{\text{SS}}^{*2}$  this time requires only the statistical information incorporated into the “surface” correlation function  $F^{\text{PP}}(\rho)$ .

Consider two classical examples that concern dispersions of identical spherical sinks of radius  $a$  with number density  $n$ .

In the first example the sinks are forbidden to overlap. Their centers are distributed randomly with the two-point distribution function

$$f_2(r) = n^2 g(r). \quad (22)$$

The interpretation is that  $f_2(r) dV_A dV_B$  provides the probability of finding spheres centers in the vicinities  $dV_A$  and  $dV_B$  of the points  $A$  and  $B$ , respectively, such that the distance between the latter is  $r$ ; the function  $g(r)$  in (21) is the familiar radial distribution function.

The moments (14) have been evaluated in [10], among other statistical characteristics of dispersions of nonoverlapping spheres, making use of the function  $g(r)$ . The needed, for our purposes, expressions read

$$\begin{aligned} \theta_1^{\text{pp}} &= \eta_1^2 \left( \frac{2 - 9\eta_1}{5\eta_1} + m_1 \right), \\ \theta_1^{\text{ps}} &= \eta_1 \left( \frac{5 - 26\eta_1}{15\eta_1} + m_1 \right), \\ \theta_1^{\text{ss}} &= \frac{1 - 5\eta_1}{3\eta_1} + m_1, \end{aligned} \quad (23)$$

where  $\eta_1 = \frac{4}{3}\pi n a^3$  is the volume fraction of the sinks (the phase ‘1’) and

$$m_1 = \int_2^\infty \rho \nu_2(\rho) d\rho, \quad \nu_2(\rho) = g(\rho) - 1, \quad (24)$$

so that  $\nu_2(r)$  is the so-called total correlation function. A simple check shows that the condition (19) is satisfied, whatever the total correlation. Hence our bound coincides in this case with Doi’s one, yielding

$$k^{*2} \geq k_{\text{D}}^{*2}, \quad k_{\text{D}}^{*2} a^2 \geq \frac{3\eta_1(1 - \eta_1)}{1 - 5\eta_1 - \eta_1^2/5 + 3\eta_1 m_1} = 3\eta_1 + o(\eta_1). \quad (25)$$

As indicated, the bound (25) is *exact* in the dilute limit  $\eta_1 \ll 1$ , since it reproduces in this case the well-known Smoluchowski’s result [1].

The bound (25) first appeared in Willis’ lecture [4]; a more precise derivation, together with some generalizations, is due to Talbot and Willis [5], see also [11] for a discussion and an alternative derivation, based on the Rubinstein-Torquato variational principle (4). The fact that the original Doi’s bound for a dispersion of nonoverlapping spheres can be recast in the elegant Talbot-Willis’ form (24) was noticed also by Beasley and Torquato [12].

It is noted that the “particle-particle” and “surface-surface” bounds for the dispersion under study have, respectively, the form

$$\begin{aligned} k_{\text{pp}}^{*2} a^2 &= \frac{5\eta_1(1 - \eta_1)}{2 - 9\eta_1 + 5\eta_1 m_1} = \frac{5}{2}\eta_1 + o(\eta_1), \\ k_{\text{ss}}^{*2} a^2 &= \frac{3\eta_1(1 - \eta_1)}{1 - 5\eta_1 + 3\eta_1 m_1} = 3\eta_1 + o(\eta_1), \end{aligned} \quad (26)$$

see Eqs. (20) – (22).

Clearly, the “surface” bound  $k_{\text{ss}}^{*2}$  is superior to the “particle” one, which is natural — the absorption phenomenon under study, Eq. (1), is governed by the absorption, taking place on the interphase boundary. Moreover,  $k_{\text{ss}}^{*2}$  reproduces the correct Smoluchowski’s value in the dilute limit, unlike  $k_{\text{pp}}^{*2}$ . Also,  $k_{\text{ss}}^{*2}$  is very close to Doi’s bound  $k_{\text{D}}^{*2}$ , the sole difference being the term  $-\eta_1^2/5$  in the denominator, cf. Eqs. (25) and (26). However, one cannot claim that  $k_{\text{ss}}^{*2}$  will be superior to  $k_{\text{pp}}^{*2}$  for all random constitutions and for all values of  $\eta_1$ . An example, when this is not so, will be supplied below, when discussing the appropriate bounds for the Boolean model of overlapping spheres.

In the simplest “well-stirred” dispersion  $g(r) - 1 = h_{2a}(r)$ , i.e. the spheres are only forbidden to overlap ( $h_{2a}(r)$  is the characteristic function for a sphere of radius  $2a$  located at the origin). Then  $\nu_2(\rho) = 0$ , if  $\rho \geq 2$ ,  $m_1 = 0$  and the bounds (24), (25) become

$$\begin{aligned} k_{\text{pp}}^{*2} a^2 &= \frac{5\eta_1(1 - \eta_1)}{2 - 9\eta_1}, & k_{\text{ss}}^{*2} a^2 &= \frac{3\eta_1(1 - \eta_1)}{1 - 5\eta_1}, \\ k_{\text{D}}^{*2} a^2 &= \frac{3\eta_1(1 - \eta_1)}{1 - 5\eta_1 - \eta_1^2/5}. \end{aligned} \quad (27)$$

Obviously, the “particle” bound  $k_{\text{pp}}^{*2}$  fails in this case if  $\eta_1 \geq 2/9$ . Similarly, the “surface” one,  $k_{\text{ss}}^{*2}$ , fails at  $\eta_1 \geq 0.2$ , and Doi-Talbot-Willis  $k_{\text{D}}^{*2}$  — at  $\eta_1 \geq \eta_1^0$ ,  $\eta_1^0 \approx 0.1984$  — a fact, explicitly underlined in [4, 5]. This means that the “well-stirred” approximation is unrealistic beyond the value  $\eta_1^0$  of sphere fraction. This fact, however, is of little interest due to the more recent result of Markov and Willis [13, 14], stating that “well-stirred” approximation is already unrealistic if  $\eta_1 \geq 1/8$ .

More realistic than the well-stirred one is the Percus-Yevick (PY) approximation for a dispersion of nonoverlapping spheres [15], widely used in the liquid state theory. The Laplace transform of the function  $\nu_2$  is analytically known due to Wertheim [16]. An appropriate asymptotic analysis of the Wertheim’s formula allows one to obtain, in turn, a number of statistical characteristics of a PY dispersion, see [16, 6]. In particular, it turns out that the parameter (24), needed in the bounds under consideration, is simply

$$m_1^{\text{PY}} = \frac{\eta_1(22 - \eta_1)}{5(1 + 2\eta_1)}. \quad (28)$$

(Note that an equivalent, but much more complicated formula for  $m_1^{\text{PY}}$  is given by Talbot and Willis [5, Eqs. (8.14) and (8.15)].)

The formula (28), when inserted into (25), gives the Doi-Talbot-Willis bound for a Percus-Yevick dispersion in an extremely simple form:

$$k_{\text{D}}^{*2} a^2 = \frac{3\eta_1(1 + 2\eta_1)}{(1 - \eta_1)^2}. \quad (29)$$

The values of  $k_{\text{D}}^{*2}$  obviously remain finite for all sphere fractions  $\eta_1 \in (0, 1)$ . This fact makes the application of the PY approximation suspicious for higher volume

fractions. The reason is that *any* realistic model of dispersions, in which the spheres are forbidden to overlap, should fail for volume fractions higher than 0.64 — the value corresponding approximately to the close packing of the inclusions.

The second case is the well-known randomly imbedded model of spheres [13, 14], called also Boolean [15]. Here an infinite family of points are placed “fully” randomly throughout the space — more precisely, forming a Poissonian system of number density (intensity)  $n$ . Identical spheres of the radius  $a$  are centered then at these points, with overlapping permitted. The phase ‘2’ (the “sink-free” part) is then defined as the region, empty of spheres. The “sink” phase ‘1’ comprises either the single spheres or the aggregates, formed by families of overlapping spheres.

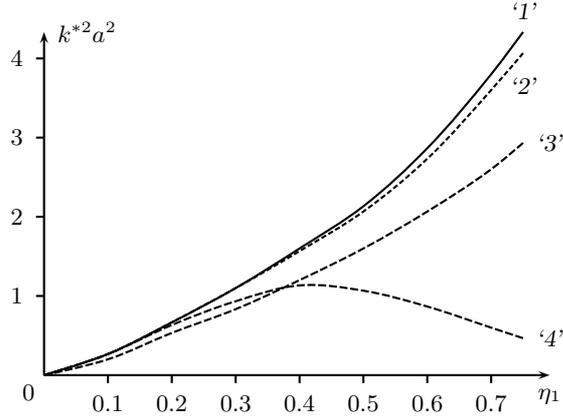


Fig. 1. The various bounds on the dimensionless effective absorption coefficient for the Boolean model. ‘1’ — our new bound  $k_N^{*2}a^2$ , see (17); ‘2’ — the Doi bound  $k_D^{*2}a^2$ , see (18); ‘3’ — the “surface-surface” bound  $k_{ss}^{*2}a^2$ , see (21); ‘4’ — the “particle-particle” bound  $k_{pp}^{*2}a^2$ , see (20)

Here the needed two-point correlations, as evaluated by Doi [2], read

$$\begin{aligned}
 \eta_2 &= \exp\left(\frac{4}{3}\pi na^3\right), \quad D(\rho) = 1 + \frac{3}{4}\rho - \frac{3}{16}\rho^3, \\
 F^{\text{pp}}(\rho) &= \left(\eta_2^{D(\rho)} - \eta_2^2\right) (1 - H(\rho - 2)), \\
 \frac{1}{S}F^{\text{ps}}(\rho) &= \left(\eta_2 - \frac{(2 + \rho)(\eta_2^2 + F^{\text{pp}}(\rho))}{4\eta_2}\right) (1 - H(\rho - 2)), \\
 \frac{1}{S^2}F^{\text{ss}}(\rho) &= \left\{ \left(\frac{1}{6\eta_1\eta_2^2\rho} + \frac{(2 + \rho)^2}{16\eta_2^2}\right) (\eta_2^2 + F^{\text{pp}}(\rho)) - 1 \right\} (1 - H(\rho - 2)),
 \end{aligned} \tag{30}$$

here  $\rho = r/a$ ,  $H(r)$  is the Heaviside function.

The moments (14) can be evaluated in this case only numerically. The appropriate bounds on the dimensionless effective absorption coefficient  $k^{*2}a^2$  are

shown in Fig. 1 as functions on the volume fraction  $\eta_1$  of the sink constituent. It is seen that our bound (17) does improve upon the Doi one, given in (17), but the improvement is small and shows up only at very high values of the fraction  $\eta_1$ . The behaviour of the “surface-surface” bound  $k_{ss}^* a^2$ , see (21), deserves some more attention. For dilute fractions  $\eta_1 \ll 1$ , it is undoubtedly better than the “particle-particle” one, see (26), since the Boolean and “nonoverlapping” dispersions share the same effective properties in the dilute limit. However, in the region  $\eta_1 \in (0.1, 0.3)$ , a bit unexpectedly,  $k_{ss}^* a^2$  falls below  $k_{pp}^* a^2$ . Only at  $\eta_1 > 0.3$  the “surface-surface” bound becomes superior as compared to the “particle-particle” one. Moreover, it becomes much more sensible when  $\eta_1$  increases. At the same time the bound  $k_{pp}^* a^2$  deteriorates badly with increasing  $\eta_1$ . The reason is clear: in the Boolean model, when the sink fraction increases, the overlapping becomes more and more frequent, the shape of the aggregates formed by the particles becomes more and more complicated and the specific surface increases considerably as a result.

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