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ORIGINAL CONTRIBUTIONS

The average stress tensor in systems with interacting particles*)

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Abstract: The derivation of an expression of the macroscopic stress tensor in terms of microscopic variables in systems of finite interacting particles is discussed from different points of view.
It is shown that in volume averaging the introduction of a fictitious "interaction stress field" $\boldsymbol{T}^I$ with special boundary conditions on the boundary of the averaging volume is needed. In ensemble averaging similar results are obtained by using a multipole expansion of the local stress and force fields. In the appropriate limiting cases, the obtained results are shown to be consistent with the results of kinetic theories of polymer solutions.

Key words: Average stress tensor, interacting particles, ensemble averaging, multiple expansion

List of Symbols

$\boldsymbol{A}$ arbitrary field
$\boldsymbol{D}$ rate-of-strain tensor
$\boldsymbol{f}$ force density (per unit volume)
$\boldsymbol{f}_i$ force acting on particle i
$\boldsymbol{f}_{ij}$ force acting on particle i due to particle j
$\boldsymbol{n}$ unit normal
$\boldsymbol{r}$ radius vector with respect to an arbitrary origin
$\boldsymbol{r}_i$ position of particle i
$\boldsymbol{r}_{ij}$ $\boldsymbol{r}_j - \boldsymbol{r}_i$
$\boldsymbol{T}$ stress tensor
$\boldsymbol{t}$ traction
$\boldsymbol{v}$ velocity field
V averaging volume
∂V boundary of V
V_i volume of particle i
v_i arbitrary volume enclosing particle i

Superscripts

I interaction
H hydrodynamic
C contact

Subscripts

P particle
F fluid

Special symbols

$\overline{(\ldots)}$ volume average
$\langle \ldots \rangle$ ensemble average
$\tilde{(\ldots)}$ macroscopic quantity
$\bar{\nabla}$ or $\bar{\bar{\nabla}}$ spatial differential operator (arrow denotes direction of the quantity to be operated on)

1. Introduction

In any rheological model relating macroscopic properties of a system to its microstructure, an expression is required that relates the macroscopic stress tensor to microscopic stresses and forces in the system. For polymeric systems that can be modelled by bead-spring or bead-rod models such relations are well established now by the fundamental work of Curtiss, Bird, and Hassager [1] on the kinetic theory of macromulecular solutions. This work is based upon methods of statistical mechanics, originally developed by Irving and Kirkwood [2] for monoatomic liquids. The simplest form of the expression of the particle contribution to the average stress tensor in dilute solutions according to this approach reads – in our notation (see Appendix A) –

$$\tilde{\boldsymbol{T}}_p = \sum_i \langle \boldsymbol{f}_i^H \boldsymbol{r}_i \rangle \,. \tag{1}$$

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Here and elsewhere in this paper a tilde is used to denote macroscopical quantities and brackets to denote ensemble averages; f_i^H is the hydrodynamical force on the bead i located at the point with radius vector r_i.

From (1) other expressions for the particle contribution to the macroscopic stress tensor in polymeric systems, such as the so-called "Giesekus form" and the "Kramers form", may be obtained. For an extensive discussion on this subject we refer to Bird et al. [3]. Expressions for concentrated systems of interacting point particles can be found in [4] and [5].

All results mentioned so far are based upon statistical-mechanical theories in which particles are treated as mass points. Such theories are suitable for homogeneous polymeric systems, but not for heterogeneous systems, such as filled polymers, dispersions, and so on, in which the finite size of the particles is essential. For such systems a general statistical-mechanical framework is not yet available and other averaging techniques have to be employed.

In the case of dilute dispersions expressions for the macroscopic stress tensor can be obtained by means of volume averaging [6, 7]. The basic result for the particle contribution to the stress tensor in a system of non-interacting perticles obtained in this way is:

$$\tilde{\boldsymbol{T}}_p = \frac{1}{V} \sum_i \int_{\partial V_i} \boldsymbol{T} \cdot \boldsymbol{n} \boldsymbol{r} dS . \tag{2}$$

Here V is the spatial volume in which the volume averaging is performed, $\boldsymbol{T}$ is the local stress tensor, $\boldsymbol{n}$ the external unit normal on the particle surface ∂V_i and $\boldsymbol{r}$ a radius vector with respect to an arbitrary origin.

In the present paper some features of the volume- and ensemble-averaging techniques in the case of concentrated systems of finite interacting particles will be discussed. It will be shown in which way the contributions of type (2), as a result of the finite size of the particles and the contributions of the interaction forces, appear in the expressions for the macroscopic stress tensor of such systems and how these results are related to expressions of type (1) and to allied results in the limiting case of point particles. In order to concentrate the attention upon the contributions mentioned above, some effects that may be important in certain special cases will be neglected in the present paper; these are: inertial, Brownian, and interfacial forces.

Our treatment of volume averaging techniques is based upon related work on semi-dilute and concentrated dispersions [8 – 11]. For a general discussion of the technique of ensemble averaging in dense suspension, we refer to [12]. In that paper, interaction forces, however, are not taken into account.

2. Example: Two interacting particles

In order to discuss first a basic problem in concern with the volume averaging of local stresses, we now discuss a simple example, namely a system of two particles, dispersed in a fluid, on which hydrodynamical forces

$$\boldsymbol{f}_i^H = \int_{\partial V_i} \boldsymbol{T} \cdot \boldsymbol{n} dS \quad (i = 1,2) \tag{3}$$

are acting; these forces – since Brownian and inertial forces are neglected – are balanced by the interaction forces

$$\boldsymbol{f}_i^I = \int_{V_i} \boldsymbol{f}^I dV . \tag{4}$$

In these expressions V_i is the region in space occupied by particle i, $\boldsymbol{T}$ the local stress tensor, $\boldsymbol{n}$ an external unit normal on the particle surface ∂V_i, and $\boldsymbol{f}^I$ a local force density in V_i, describing the mutual interaction force between the particles.

In each point of the fluid as well as the particles the equation of motion (without inertia),

$$\operatorname{div} \boldsymbol{T} + \boldsymbol{f} = \boldsymbol{0} , \tag{5}$$

with $\boldsymbol{f} = 0$ in the fluid and $\boldsymbol{f} = \boldsymbol{f}^I$ in the particles, is valid. From eqs. (3) and (4) we obtain the balance of forces

$$\boldsymbol{f}_i^H + \boldsymbol{f}_i^I = \boldsymbol{0} \quad (i = 1,2) . \tag{6}$$

As a model of the present system we first consider a dumbbell (fig. 1) and calculate the average stress in a volume V around this particle:

$$\boldsymbol{T} = \frac{1}{V} \int_V \boldsymbol{T} dV = \bar{\boldsymbol{T}}_F + \bar{\boldsymbol{T}}_P , \tag{7}$$

with

$$\bar{\boldsymbol{T}}_F = \frac{1}{V} \int_{V_F} \boldsymbol{T} dV \tag{8}$$

the fluid contribution and

$$\bar{\boldsymbol{T}}_P = \frac{1}{V} \int_{V_P} \boldsymbol{T} dV = \frac{1}{V} \sum_i \int_{V_i} \boldsymbol{T} dV \tag{9}$$

the particle contribution.

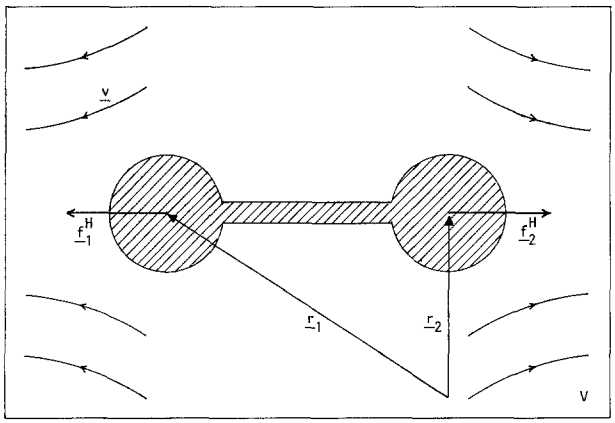

Fig. 1. Two interacting particles, modelled by a dumbbell on which the hydrodynamical forces f_i^H are acting

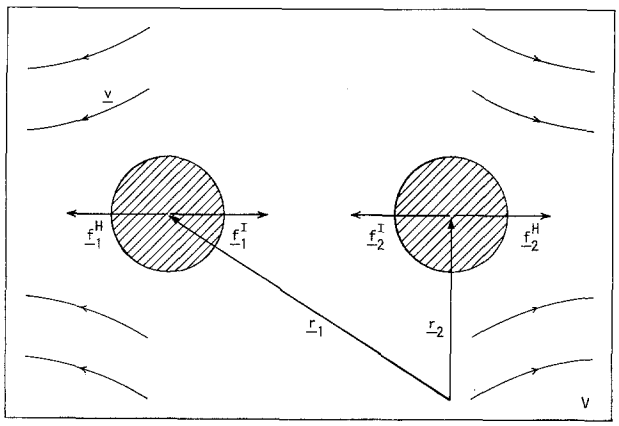

Fig. 2. Two interacting particles, modelled by spheres on which hydrodynamical forces f_i^H and interaction forces f_i^I are acting

Here and elsewhere in this paper we use the convention that a subscript F denotes the fluid and a subscript P the particle contribution in the sense that for any field $A(r)$

$$\begin{aligned} A_P &= A \text{ and } A_F = \mathbf{0} \quad \text{for} \quad r \in V_P \\ A_F &= A \text{ and } A_P = \mathbf{0} \quad \text{for} \quad r \in V_F . \end{aligned} \tag{10}$$

Expressions (8) and (9) may be considered as spatial averages over the volume V (indicated by the overbar) of the fields T_F and T_P.

In the dumbbell model the interaction force is represented by the connection rod and the dumbbell itself may be considered as a force-free particle; so by eq. (5) we have $\operatorname{div} T = \mathbf{0}$ in V_P, from which the following identity may be derived [6]:

$$\int_{V_P} T dV = \int_{\partial V_P} T \cdot n r dS . \tag{11}$$

From eqs. (9) and (11) we obtain:

$$\bar{T}_P = \frac{1}{V} \int_{\partial V_P} T \cdot n r dS . \tag{12}$$

This is a special case of the general expression (2), mentioned before. For an ideal dumbbell with small beads with negligible hydrodynamic forces on the connection rod (12) reduces to

$$\bar{T}_P = \frac{1}{V} \sum_{i=1}^{2} f_i^H r_i , \tag{13}$$

which is of the same shape as the general result (1) of kinetic theory. So in this case we conclude that the particle contribution to the stress tensor may be obtained by volume averaging: $\tilde{T}_P = \bar{T}_P$.

We now consider the same system not modelled, however, by a dumbbell, but by two interacting particles and an interaction force field f^I as described in connection with eq. (4) (fig. 2). Then instead of eq. (11) we have

$$\int_{V_P} T dV = \int_{\partial V_P} T \cdot n r dS + \int_{V_P} f^I r dV . \tag{14}$$

Since in the particles eq. (5) applies, with $f = f^I \neq \mathbf{0}$, instead of eq. (12) we obtain

$$\bar{T}_P = \frac{1}{V} \int_{\partial V_P} T \cdot n r dS + \frac{1}{V} \int_{V_P} f^I r dV . \tag{15}$$

Taking again the limit of point particles, we get

$$\bar{T}_P = \frac{1}{V} \sum_i f_i^H r_i + \frac{1}{V} \sum_i f_i^I r_i , \tag{16}$$

from which by eq. (6) we obtain the strange result

$$\bar{T}_P = \mathbf{0} . \tag{17}$$

So we see that in this model the volume average of T_P cannot be interpreted as the particle contribution to the macroscopic stress:

$$\bar{T}_P \neq \tilde{T}_P . \tag{18}$$

The reason why we do not get the right result in this case is that in volume averaging all local stresses are

added but the effect of the interaction between distant points, which is the significant contribution to $\tilde{T}_P$, in this case is disregarded.

It is still possible to use the method of volume averaging in this case, namely by employing the concept of a fictitious stress field T^I describing the interaction forces. This field, which has been used in related problems by Russel [9] and by Batchelor [10], is defined by the equation

$$\operatorname{div} T^I - f = 0 \tag{19}$$

($f = f^I$ in V_P, $f = 0$ in V_F). The field T^I is not fully determined by eq. (19); it is still possible, however, to determine volume averages of it by imposing suitable boundary conditions. These boundary conditions will be discussed in general in the next section; for the example discussed here we assume T^I to be localized in the neighbourhood of the two particles and therefore impose the boundary condition

$$T^I \cdot n = 0 \quad \text{on} \quad \partial V. \tag{20}$$

A direct physical interpretation of T^I follows by integrating eq. (19) over any volume $v_i \subset V$ enclosing one of the two particles. Then it follows that

$$f_i^I = \int_{\partial v_i} T^I \cdot n dS. \tag{21}$$

In this way T^I describes the interaction forces acting on the particles. Further we note that by eqs. (5) and (19) the balance equation for $T + T^I$ becomes

$$\operatorname{div}(T + T^I) = 0. \tag{22}$$

This equation is of the same form as the macroscopic equation of motion $\operatorname{div} \tilde{T} = 0$. In fact eqs. (21) and (22) form the basis for the identification of T and the volume average of $(T + T^I)$, as will be discussed in general in the next section.

We first consider again the example of two interacting particles (fig. 3). In the same way as eq. (15) has been derived from eq. (5) we obtain from eq. (19):

$$\overline{T^I} = \frac{1}{V} \int_{\partial V} T^I \cdot n r dS - \frac{1}{V} \int_V f r dV. \tag{23}$$

Using the boundary condition (20) and the fact that $f = 0$ in V_F and $f = f^I$ in V_P, we get from eq. (23):

$$\overline{T^I} = -\frac{1}{V} \int_{V_P} f^I r dV. \tag{24}$$

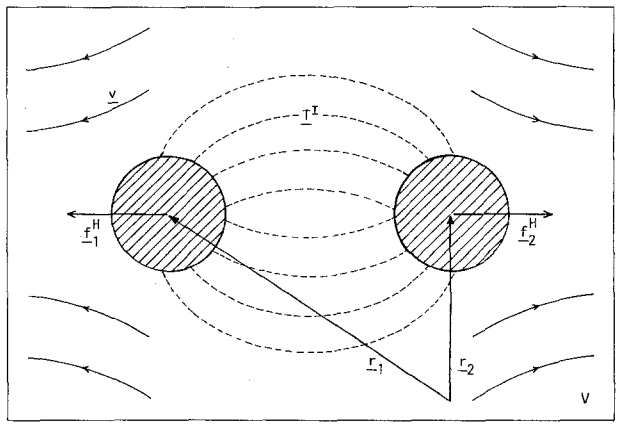

Fig. 3. Figure 3. Two interacting particles, modelled by spheres on which hydrodynamical forces f_i^H and interaction forces, described by the interaction stress tensor T^I, are acting

From eqs. (7), (15), and (24) we now obtain

$$\overline{T + T^I} = \bar{T}_F + \frac{1}{V} \int_{\partial V_P} T \cdot n r dS. \tag{25}$$

Comparing this expression with the previous results (7), (8), and (12) of the dumbbell model, we see that the macroscopic stress tensor in this example is given by the volume average of $T + T^I$:

$$\tilde{T} = \overline{T + T^I}. \tag{26}$$

The results obtained so far are in accordance with the approach of Russel [9] and Batchelor [10] for dilute dispersions with interacting particles. In the next section the general case of more concentrated systems will be discussed.

3. Concentrated systems

For concentrated systems of interacting particles the boundary condition eq. (20) is no longer applicable since through any part of the boundary ∂V of a control volume V interactions between neighbouring particles take place. In order to obtain the correct boundary conditions for this case we note first that similar to eq. (21) we have

$$\sum_{i,\alpha} f_{i\alpha}^I = \int_{\partial V} T^I \cdot n dS, \tag{27}$$

where $f^I_{i\alpha}$ is the interaction force on the particle i in V due to the particle α outside V. Here we assume pair interactions between the particles, i.e.:

$$f^I_i = \sum_{j\in V} f^I_{ij} + \sum_{\alpha\notin V} f^I_{i\alpha}. \tag{28}$$

We further assume that the interactions have a range which is short in comparison with the dimensions of the control volume V. This means that an equation of the form (27) holds for arbitrary parts of the surface V and also for any arbitrary surface element $d\tilde{S}$ which is infinitesimal on a macroscopic scale but large compared with the ranges of the interaction forces. So we have:

$$\sum_{i,\alpha} f^I_{i\alpha} = \int_{d\tilde{S}} \boldsymbol{T}^I \cdot \boldsymbol{n} dS, \tag{29}$$

where the particles i and α are located on both sides of $d\tilde{S}$. Eq. (28) will be used as a boundary condition for $\boldsymbol{T}^I$ in concentrated systems.

For later use we further note that because of the assumption of shortrange interactions it follows from eq. (28) that

$$\sum_i \sum_\alpha f^I_{i\alpha} r_i = \int_{d\tilde{S}} \boldsymbol{T}^I \cdot \boldsymbol{n} r dS \tag{30}$$

in cases where $|\boldsymbol{r} - \boldsymbol{r}_i| \ll |\boldsymbol{r}|$.

We now prove that the volume average $\overline{\boldsymbol{T} + \boldsymbol{T}^I}$ over a volume V that is infinitesimal on a macroscopic scale, but large with respect to microscopic dimensions, may be interpreted as the macroscopic stress tensor $\tilde{\boldsymbol{T}}$. To this end we first note that under suitable conditions of continuity the operations of volume averaging and spatial differentiation are interchangeable (see Appendix B) and further that eq. (22) is still applicable, so we have:

$$\overline{\operatorname{div}(\boldsymbol{T} + \boldsymbol{T}^I)} = \operatorname{div}\overline{(\boldsymbol{T} + \boldsymbol{T}^I)} = \boldsymbol{0}, \tag{31}$$

which is of the same form as the macroscopical balance equation for $\tilde{\boldsymbol{T}}$:

$$\operatorname{div}\tilde{\boldsymbol{T}} = \boldsymbol{0}. \tag{32}$$

We further note that the total contact forces transmitted through a surface element $d\tilde{S}$ is given by

$$\tilde{f}^c(d\tilde{S}) = \tilde{\boldsymbol{T}} \cdot \tilde{\boldsymbol{n}} d\tilde{S}, \tag{33}$$

whereas from a microscopic point of view we have:

$$f^c(d\tilde{S}) = \int_{d\tilde{S}} \boldsymbol{T} \cdot \boldsymbol{n} dS + \sum_{i,\alpha} f^I_{i\alpha}, \tag{34}$$

where the forces $f^I_{i,\alpha}$ are the same as in eq. (29).

Substitution of eq. (29) gives:

$$\tilde{f}^c(d\tilde{S}) = \int_{d\tilde{S}} (\boldsymbol{T} + \boldsymbol{T}^I) \cdot \boldsymbol{n} dS. \tag{35}$$

Since the (local) volume averages of $\boldsymbol{T}$ and $\boldsymbol{T}^I$ are smooth functions of place and approximately constant on $d\tilde{S}$, it follows from eqs. (35) and (B-4) of Appendix B that

$$\tilde{f}^c(d\tilde{S}) = \overline{(\boldsymbol{T} + \boldsymbol{T}^I)} \cdot \tilde{\boldsymbol{n}} d\tilde{S}, \tag{36}$$

where the macroscopic unit normal $\tilde{\boldsymbol{n}}$ is defined by

$$\tilde{\boldsymbol{n}} d\tilde{S} = \int_{d\tilde{S}} \boldsymbol{n} dS. \tag{37}$$

From eqs. (33) and (36) it follows that $\tilde{\boldsymbol{T}}$ and $\overline{\boldsymbol{T} + \boldsymbol{T}^I}$ obey the same boundary conditions on $d\tilde{S}$. Since, as we have seen from eqs. (31) and (32), also the balance equation for both fields is the same, we conclude that eq. (26) applies in the general case of concentrated systems.

We now proceed to derive an expression for $\tilde{\boldsymbol{T}}$ in terms of microscopic stress- and force-fields. In the same way as eqs. (8), (15), and (23) were derived in the prevous example we obtain from eq. (26) in the general case:

$$\tilde{\boldsymbol{T}} = \bar{\boldsymbol{T}}_F + \frac{1}{V}\int_{\partial V_P} \boldsymbol{T} \cdot \boldsymbol{n} r dS + \frac{1}{V}\int_{\partial V} \boldsymbol{T}^I \cdot \boldsymbol{n} r dS. \tag{38}$$

In the second term of the right-hand side we write, using eq. (3):

$$\int_{\partial V_P} \boldsymbol{T} \cdot \boldsymbol{n} r dS = \sum_i \int_{\partial V_i} \boldsymbol{T} \cdot \boldsymbol{n}(\boldsymbol{r} - \boldsymbol{r}_i) dS + \sum_i \int_{\partial V_i} \boldsymbol{f}^H_i \boldsymbol{r}_i, \tag{39}$$

where $\boldsymbol{r}_i$ is the radius vector of any specific point (for instance the centre of mass) of particle i with respect to a fixed origin in space. By using the balance equation of forces eq. (6) and the decomposition eq. (28) of the interaction forces f^I_i the last term in eq. (39) may be writen as

$$\sum_i \boldsymbol{f}^H_i \boldsymbol{r}_i = -\sum_{i,j} \boldsymbol{f}^I_{ij} \boldsymbol{r}_i - \sum_{i,\alpha} \boldsymbol{f}^I_{i\alpha} \boldsymbol{r}_i, \tag{40}$$

where, again, the particles $i, j \in V$ and the particles $\alpha \notin V$. From eqs. (39–41), using the fact that

$$f_{ij}^I = -f_{ji}^I \tag{41}$$

and the boundary condition (30), we finally obtain the following expression for $\tilde{T}$:

$$\tilde{T} = \bar{T}_F + \frac{1}{V} \sum_i \int_{\partial V_i} T \cdot n(r - r_i) dS + \frac{1}{2V} \sum_{i,j} f_{ij}^I r_{ij}, \tag{42}$$

where $r_{ij} = r_j - r_i$.

We now have three expressions for $\tilde{T}$, based upon the concept of volume averaging: (26), (38), and (42). The first and the second one of these expressions contain the fictitious stress field T^I, which is locally undetermined on a microscopic scale. These expressions are therefore not directly applicable in general (only in dilute systems, where the boundary condition (20) holds, eq. (38) can be used directly). Eq. (42) containing only real physical quantities, therefore seems to be the most useful for practical purposes.

In the case of the fluid phase (F) consisting of a Newtonian fluid we have:

$$T = -p\mathbf{1} + 2\eta D \quad (\text{in } V_F), \tag{43}$$

where p is the pressure, η the viscosity, and D the rate-of-strain tensor. The fluid contribution $\bar{T}_F$ in eq. (42) then becomes:

$$\bar{T}_F = -\bar{p}_F\mathbf{1} + 2\eta\bar{D}_F, \tag{44}$$

where the averages $\bar{p}_F$ and $\bar{D}_F$ are defined similarly to eq. (8). The term $\bar{D}_F$ may be expressed also in terms of the total volume average $\bar{D}$ of the rate-of-strain tensor by means of identity [7]

$$\bar{D} = \bar{D}_F + \frac{1}{2V} \int_{\partial V_P} (vn + nv) dV, \tag{45}$$

in which v is the velocity field and n the external unit normal at the surface of the particles. In the case of rigid particles the last term in eq. (45) vanishes.

The second term of eq. (42), which is similar to eq. (2) of dilute dispersions, reflects the effect of the finite size of the particles. The last term in eq. (42) represents the effect of the interaction forces. This term is similar to the so-called "Kramers expression" for the stress tensor in polymeric systems modelled by bead-spring models [3].

It is important to note that in concentrated systems of finite interacting particles both the term due to the finite size of the particles and the interaction term occur simultaneously.

4. Ensemble averaging

The derivation of expression (42) of the macroscopic stress tensor was based upon the interaction stress tensor T^I. The introduction of this rather artificial concept was necessary since we were using the technique of volume averaging. It will be shown now that an equation similar to (42) may be obtained without using the introduction of T^I, by employing the method of ensemble averaging.

We start with the local equation of motion, which in any point inside the particle (P) or the fluid phase (F) reads:

$$\operatorname{div} T + f = \mathbf{0}. \tag{46}$$

In this expression using the notation (10), we write:

$$T = T_P + T_F; \quad f = f_P + f_F. \tag{47}$$

So we have:

$$f_P = f^I, \quad f_F = \mathbf{0}. \tag{48}$$

The fields T_P and f_P are expanded now in terms of multipoles with respect to the centres of the particles. For any field A_P this can be achieved as follows:

$$\begin{aligned} A_P(r) &= \int_{V_P} A(s)\delta(r - s) d^3s \\ &= \sum_i \int_{V_i} A(s)\delta(r - s) d^3s \\ &= \sum_i \sum_{m=0}^{\infty} \frac{(-1)^m}{m!} \int_{V_i} A(s)(s - r_i)^m \\ &\quad \cdot d^3s\delta(r - r_i) \odot \overleftarrow{\nabla}_r^m. \end{aligned} \tag{49}$$

In these expressions $\odot$ denotes an m-fold contraction and $\overleftarrow{\nabla}_r^m$ an m-fold dyadic product of the spatial differentiation operator $\overleftarrow{\nabla}_r \equiv \frac{\partial}{\partial r}$ acting on the δ-functions in eq. (49). Assuming a smooth dependence of the probability densities of the coordinates r_i to be used in the ensemble averaging, we may neglect higher-order terms in expansions like eq. (49). In particular, we write for the fields T_P and f_P:

$$\boldsymbol{T}_P = \sum_i \int_{V_i} \boldsymbol{T} dV \delta(\boldsymbol{r} - \boldsymbol{r}_i) \tag{50}$$

$$\boldsymbol{f}_P = \sum_i \int_{V_i} \boldsymbol{f} dV \delta(\boldsymbol{r} - \boldsymbol{r}_i) - \operatorname{div}\left(\sum_i \int_{V_i} \boldsymbol{f}(\boldsymbol{r})(\boldsymbol{r} - \boldsymbol{r}_i) dV \delta(\boldsymbol{r} - \boldsymbol{r}_i)\right). \tag{51}$$

Using in eq. (50) the same integral transformation (14) as has been used in connection with the volume averaging, we obtain from eqs. (46), (47), (48), (50), and (51) the following reduced form of the equation of motion:

$$\operatorname{div}\left(\boldsymbol{T}_F + \sum_i \int_{\partial V_i} \boldsymbol{T} \cdot \boldsymbol{n}(\boldsymbol{r} - \boldsymbol{r}_i) dS \delta(\boldsymbol{r} - \boldsymbol{r}_i)\right) + \sum_i \boldsymbol{f}_i^I \delta(\boldsymbol{r} - \boldsymbol{r}_i) = \boldsymbol{0}. \tag{52}$$

Note that the second moments of the force field $\boldsymbol{f}^I(\boldsymbol{r})$ inside the particles in the last term of eq. (51) do no longer appear in expression (52). We now take the ensemble average of eq. (52) with respect to the probability density of the coordinates $\boldsymbol{r}_i$ and the internal coordinates of the particles. Since this probability density (of the whole system) is not an explicite function of the spatial coordinate $\boldsymbol{r}$, the ensemble averaging commutes with the differentiation in the first term of eq. (52). The ensemble average of the second term of eq. (52) may be treated in the same way as in the case of point particles [2]:

$$\begin{aligned} \langle \boldsymbol{f}_i^I \delta(\boldsymbol{r} - \boldsymbol{r}_i) \rangle &= \sum_{i,j} \langle \boldsymbol{f}_{ij}^I \delta(\boldsymbol{r} - \boldsymbol{r}_i) \rangle \\ &= \frac{1}{2} \sum_{i,j} \langle \boldsymbol{f}_{ij}^I (\delta(\boldsymbol{r} - \boldsymbol{r}_i) - \delta(\boldsymbol{r} - \boldsymbol{r}_j)) \rangle \\ &= \frac{1}{2} \operatorname{div} \sum_{i,j} \langle \boldsymbol{f}_{ij}^I \boldsymbol{r}_{ij} \delta(\boldsymbol{r} - \boldsymbol{r}_i) \rangle . \end{aligned} \tag{53}$$

Here we used eq. (41) and a Taylor-series expansion of the δ-functions, in which higher-order terms have been neglected.

From eq. (52) we obtain in this way an expression of the form of the macroscopic of motion (32), with a macroscopic stress tensor given by:

$$\tilde{\boldsymbol{T}} = \langle \boldsymbol{T}_F \rangle + \left\langle \sum_i \int_{\partial V_i} \boldsymbol{T} \cdot \boldsymbol{n}(\boldsymbol{r} - \boldsymbol{r}_i) dS \delta(\boldsymbol{r} - \boldsymbol{r}_i) \right\rangle + \left\langle \tfrac{1}{2} \sum_{i,j} \boldsymbol{f}_{ij}^I \boldsymbol{r}_{ij} \delta(\boldsymbol{r} - \boldsymbol{r}_i) \right\rangle . \tag{54}$$

The form of this expression is indeed in accordance with our previous result (42), which was based upon volume averaging.

5. Discussion

In this paper the effect of the finite size of the particles and of interaction forces on the macroscopic stress tensor of concentrated systems has been discussed from different points of view. We have seen that in the case of volume averaging being employed, the effect of local forces should be described by the global concept of an interaction stress field $\boldsymbol{T}^I$, whereas in the case of ensemble averaging the microscopic stress- and force-fields are localized as singularities in the centres of the particles. In both cases we obtain similar expressions for the contribution to the macroscopic stress tensor as a result of the effects mentioned above. These expressions showed to be consistent with the special forms known from the statistical-mechanical theories of point particles and of the theories of dilute dispersions.

Several effects that may be important in certain special cases have not been discussed in this paper. In dilute systems for instance the effect of Brownian forces may be significant. The effect of such forces associated with the special distribution of the particles can be described with a tensor field $\boldsymbol{T}^B$ of the same type of the interaction tensor field $\boldsymbol{T}^I$. This is discussed in [10] for a dilute suspension of spherical particles. The general incorporation of Brownian effects in concentrated dispersions of particles of an arbitrary shape, however, requires a more detailed specification of the statistical distribution functions. For systems consisting of very large particles or in porous media a relatively large fraction of the particles will lie in the boundary ∂V of the control volume V of the volume averaging. In that case, a refinement of the boundary condition (27) may be required. Further modifications of our expressions are needed of course in cases where the assumptions of negligible inertial and interfacial forces are not valid.

Appendix A

In literature expressions may be found that are similar to our results, but in some transposed tensorial form or even with opposite signs. Such differences are caused by different conventions in the definition of stress and related differences in the equation of motion. In this paper we use the convention that

$$\boldsymbol{t} = \boldsymbol{T} \cdot \boldsymbol{n}, \tag{A-1}$$

where t is the traction on a surface element with external unit normal n and T the stress tensor.

The corresponding term in the equation of motion may then be written symbolically as

$$\mathrm{div}\, T = T \cdot \overleftarrow{\nabla} \tag{A-2}$$

or in component form:

$$(\mathrm{div}\, T)_i = T_{ij} \overleftarrow{\nabla}_j = T_{ij,j} . \tag{A-3}$$

Appendix B

In order to define spatial derivatives and integrals on a macroscopic scale of volume averages, we use the concept of local volume averaging, introduced by Slattery [13], in connection with the theory of flow through porous media. In that approach with any point in the phases F or P a volume V with a particular shape is associated (for instance a sphere the centre of which coincides with the point considered). The local volume average $\bar{A}(r)$ of a field A in the point r is then defined as the average over the volume associated with the point r. For the volume averages of A_P and A_F, defined by eq. (10), it can be proved then [13] that

$$\mathrm{grad}\bar{A}_P = \frac{1}{V} \int_{V_P} \mathrm{grad} A\, dV + \frac{1}{V} \int_{\partial V_P'} A \cdot n\, dS , \tag{B-1}$$

where $\partial V_P'$ is the interface between the phases F and P inside V and n the unit normal, external to particles. In the case of $A \cdot n$ being continuous at $\partial V_P'$ we obtain from an addition of eq. (B-1) and the corresponding equation for $\bar{A}_F$ the simple result that

$$\mathrm{grad}\bar{A} = \overline{\mathrm{grad} A} . \tag{B-2}$$

This result has been used in the derivation of the balance eq. (31) of $(T + T^I)$. The condition of continuity at $\partial V_P'$ requires in this case that interfacial stresses should be negligible.

The size of the averaging volume V should be small compared with macroscopical dimensions, i.e. the length scale over which significant variation of the macroscopic fields take place. In that case it can be proved [14] that volume and surface integrals are approximately equal to the corresponding integrals of the microscopical fields:

$$\int_{\tilde{V}} \bar{A}\, dV \simeq \int_{\tilde{V}} A\, dV , \tag{B-3}$$

$$\int_{\tilde{S}} \bar{A}\, dS \simeq \int_{\tilde{S}} A\, dS . \tag{B-4}$$

In these equations the volume $\tilde{V}$ and the surface $\tilde{S}$ are of macroscopic dimensions.

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