MASS TRANSFER THROUGH RAREFIED GAS BETWEEN CONCENTRIC SPHERES*

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A theoretical analysis of mass transfer under a reduced pressure, especially the transition regime was made on the basis of the gas kinetic theory. Maxwell moment method utilizing the two-sided Maxwellian distribution function proposed by Lees et al. was applied to the mass transfer between two concentric spheres, and the accommodation coefficient was taken into account. The analytical solution with the evaluated accommodation coefficient 0.9 agreed well with the experimental results of sublimation of naphthalene performed by the authors.

Introduction

Mass transfer operations under reduced pressures, such as freeze-drying, molecular distillation, metallizing under vacuum and so on, are accompanied with phase change in all cases and play a very important roll in chemical engineering. Considering the mass transfer phenomena with phase change, for example, sublimation, condensation and evaporation, two kinds of resistance for mass transfer should be evaluated. One is the resistance through the surrounding space, so called the resistance of gaseous diffusion and the other is the one for phase change at the interface, which is concerned in accommodation, condensation or evaporation coefficients.

For mass transfer through the surrounding space three regimes classified by the Knudsen number, Kn , have been considered. Those are the free molecular regime under highly rarefied condition, $Kn > 10$, the continuum regime under ordinary condition, $Kn < 0.01$, and the transition regime between them, $0.01 < Kn < 10$. The mass transfer under this transition regime has given a great interest to investigators of physics and technology. Especially its applications are very important for chemical engineering but few theoretical and experimental works^{3,4,8,11} have been performed because of its complexity.

It has been generally accepted apriori by the previous investigators⁸ that Stefan's equation with the pressure-jump boundary condition which is analogous to Maxwell-Smoluchowski's temperature-jump boundary condition for heat transfer would be correct. But there has been no theoretical basis on applying Stefan's equation to the mass transfer with large degree of rarefaction, high mass flux and large pressure gradient between evaporating (or sublimating) and condensating surface.

The present work was undertaken to derive a theoretical equation of mass transfer under reduced pressures, especially transition regime, through the gas kinetic theory and compare with the experimental results.

The Maxwell moment method utilizing the two-sided Maxwellian distribution function^{2,6,7} was applied to the mass transfer between concentric spheres. The analytical

solution obtained was compared with the experimental results obtained by the sublimation of naphthalene under reduced pressures ($10^{-1} \sim 10^{-3}$ mmHg in total pressure).

1. Theoretical Works

1-1. Distribution Function and Mean Quantities

Let us consider a sphere of radius R_1 placed at the center of a hollow sphere of radius R_2 , with $R_1 < R_2$ (Fig. 1).

Sublimation (or evaporation) of molecule A occurs at the outer surface of the inner sphere and condensation at the inner surface of the outer sphere. The annular region ($R_1 < R < R_2$) is filled with diffusion gas A and inert gas B .

By applying Lee's model², the "two-sided Maxwellian" velocity distribution functions are defined at an arbitrary point in the annular space as follows (Fig. 1); all outwardly directed molecules with velocity vector ξ ,

$$\xi = (\xi_R^2 + \xi_\theta^2 + \xi_\phi^2)^{1/2} \quad (1)$$

$$\psi = \tan^{-1}\{(\xi_\theta^2 + \xi_\phi^2)^{1/2} \xi_R^{-1}\} \quad (2)$$

lying inside "the wedge of influence"² (region 1 in Fig. 1) are characterized by one Maxwellian f_1 ,

$$f = f_1 = n_1 \left(\frac{m}{2\pi kT} \right)^{3/2} \exp\left(-\frac{m\xi^2}{2kT} \right) \quad (3)$$

for

Fig. 1 Spherical polar coordinates

* Received on November 30, 1967

$$-\frac{\pi}{2} + \alpha < \psi < \frac{\pi}{2} - \alpha$$

in which

$$\alpha = \cos^{-1}\left(\frac{R_1}{R}\right) \quad (4)$$

Then, all molecules with velocity vector ξ lying outside of region 1 are characterized by f_2 ,

i. e.,

$$f = f_2 = n_2 \left(\frac{m}{2\pi kT} \right)^{3/2} \exp\left(-\frac{m\xi^2}{2kT}\right) \quad (5)$$

for

$$\frac{\pi}{2} - \alpha < \psi < \frac{3}{2}\pi - \alpha$$

where $n_1(R)$ and $n_2(R)$ are unknown functions of radial distance.

In the above definitions, we consider the case that the temperature difference between two spheres is small so the each temperature of two spheres can be approximately denoted as the same absolute temperature T [°K]. The Maxwellian velocity distribution for stationary gas is assumed applicable when the net flow of molecule A between concentric spheres is relatively small.

Knowing the distribution function f , one can evaluate all mean quantities $\langle Q \rangle$ by averaging over whole velocity space,

$$\langle Q \rangle = \int Q f d\xi = \int_1 Q f_1 d\xi + \int_2 Q f_2 d\xi \quad (6)$$

$$\int f d\xi = n = \frac{1}{2} \{n_1(1 - \sin \alpha) + n_2(1 + \sin \alpha)\} \quad (7)$$

$$\int f \xi_R d\xi = u_R = \frac{1}{4} \left(\frac{2kT}{m\pi} \right)^{1/2} (1 + \cos 2\alpha) (n_1 - n_2) \quad (8)$$

$$\begin{aligned} - \int m \xi_R^2 d\xi &= P_{RR} \\ &= -\frac{kT}{2} \{n_1(1 - \sin^3 \alpha) + n_2(1 + \sin^3 \alpha)\} \end{aligned} \quad (9)$$

$$\begin{aligned} p &= -\frac{1}{3} \{P_{RR} + P_{\theta\theta} + P_{\phi\phi}\} \\ &= \frac{kT}{2} \{n_1(1 - \sin \alpha) + n_2(1 + \sin \alpha)\} \end{aligned} \quad (10)$$

1-2. The Maxwell Integral Equation of Transfer

In spherical co-ordinates the Maxwell integral equation of transfer for the spherical symmetric case is as follows:

$$\begin{aligned} \frac{1}{R^2} \frac{\partial}{\partial R} \left(R^2 \int f \xi_R Q d\xi \right) + \frac{1}{R \tan \theta} \int f \xi_\theta Q d\xi \\ - \int f \left(\frac{\xi_\theta^2}{R} + \frac{\xi_\phi^2}{R} \right) \frac{\partial Q}{\partial \xi_R} d\xi + \int f \left(\frac{\xi_R \xi_\theta}{R} \right. \\ \left. - \frac{\xi_\phi^2}{R \tan \theta} \right) \frac{\partial Q}{\partial \xi_\theta} d\xi + \int f \left(\frac{\xi_R \xi_\phi}{R} + \frac{\xi_\theta \xi_\phi}{R \tan \theta} \right) \frac{\partial Q}{\partial \xi_\phi} d\xi \\ = \Delta Q \end{aligned} \quad (11)$$

where ΔQ is the collision integral.

There are two kinds of molecule, diffusing molecule A and non-diffusing molecule B and one should take the distribution functions for each molecule into account. Therefore there are four unknown parameters, n_{A1} , n_{A2} , n_{B1} and n_{B2} , and they could be evaluated by utilizing four Maxwell integral equations as follows.

(a) Mass Conservation for Molecule A Setting $Q = m_A$, we find $\Delta Q = 0$ because the mass is invariant during collisions, and obtain the ordinary continuity equation as follows;

$$\frac{1}{R^2} \frac{\partial}{\partial R} \left(R^2 \int f_A m_A \xi_{AR} d\xi_A \right) = 0 \quad (12)$$

From Eqs. (8) and (12), we obtain the following relation;

$$\frac{1}{4R^2} \left(\frac{2m_A k}{\pi} \right)^{1/2} \frac{\partial}{\partial R} \{R^2(1 + \cos 2\alpha) (n_{A1} - n_{A2}) T^{1/2}\} = 0 \quad (13)$$

On the other hand;

$$R^2(1 + \cos 2\alpha) = 2R_1^2 = \text{const.}$$

then the following equation is obtained,

$$n_{A1} - n_{A2} = \gamma' = \text{const.} \quad (14)$$

where γ' is an integral constant.

(b) Mass Conservation for Molecule B

Setting $Q = m_B$,

$$\frac{1}{4R^2} \left(\frac{2m_B k}{\pi} \right)^{1/2} \frac{\partial}{\partial R} \{R^2(1 + \cos 2\alpha) (n_{B1} - n_{B2}) T^{1/2}\} = 0 \quad (15)$$

Here molecule B is the non-diffusing component, then

$$n_{B1} - n_{B2} = 0 \quad (16)$$

(c) R-momentum Conservation Summing up the

momentum transfer equations for molecule A and B , the collision integral becomes zero as the R -momentum is conservative as a whole. Substituting $Q = m_A \xi_{AR}$ or $Q = m_B \xi_{BR}$ into Eq. (11),

$$\begin{aligned} \frac{\partial}{\partial R} \left(\int f_A m_A \xi_{AR}^2 d\xi \right) + \frac{3}{R} \int \left\{ -\frac{1}{3} (f_A m_A \xi_{AR}^2 + f_A m_A \xi_{A\theta}^2 \right. \\ \left. + f_A m_A \xi_{A\phi}^2) + f_A m_A \xi_{AR}^2 \right\} d\xi = \Delta Q_{AB} \end{aligned} \quad (17)$$

Then we obtain the next equation,

$$\begin{aligned} \frac{d}{dR} \{ (n_{A1} + n_{A2}) T \} - \sin^3 \alpha \frac{d}{dR} \{ (n_{A1} - n_{A2}) T \} \\ = \frac{2}{k} \Delta Q_{AB} \end{aligned} \quad (18)$$

likewise

$$\begin{aligned} \frac{d}{dR} \{ (n_{B1} + n_{B2}) T \} - \sin^3 \alpha \frac{d}{dR} \{ (n_{B1} - n_{B2}) T \} \\ = \frac{2}{k} \Delta Q_{BA} \end{aligned} \quad (19)$$

Summing up Eqs. (18) and (19),

$$\frac{d}{dR} (n_{A1} + n_{A2} + n_{B1} + n_{B2}) = 0 \quad (20)$$

$$n_{A1} + n_{A2} + n_{B1} + n_{B2} = \delta' = \text{const.} \quad (21)$$

where δ' is an integral constant.

(d) Mass Flux for Molecule A⁵⁾ Since we are primarily interested in radial mass transfer, the mass flux equation is given by taking $Q = m_A \xi_{AR}$. In the mass flux equation, the collision integral ΔQ_{AB} is evaluated with Maxwell's inverse-fifth-power force law $F = \bar{K}_{AB}/r^{5.5}$, for simplicity. Then it should be noted that mass flux is influenced only by the collision integral ΔQ_{AB} involving the collisions between molecule A and B .

Assuming that a molecule A collides with a molecule B from a direction P (see Fig. 2), the collision integral ΔQ_{AB} is expressed as follows:

$$\Delta Q_{AB} = \iiint (Q' - Q) f_A f_B V d\xi_A d\xi_B db d\varepsilon \quad (22)$$

and by Maxwell⁵⁾,

$$\begin{aligned} \int_0^{2\pi} (Q' - Q) d\varepsilon &= \int_0^{2\pi} m_A (\xi_{AR}' - \xi_{AR}) d\varepsilon \\ &= \frac{m_A m_B}{m_A + m_B} (\xi_{BR} - \xi_{AR}) 4\pi \sin^2 \theta' \end{aligned} \quad (23)$$

$$bdb = \left\{ \frac{\bar{K}_{AB} (m_A + m_B)}{V^2 m_A m_B} \right\}^{1/2} \beta d\beta \quad (24)$$

$$A_1 = \int_0^\infty 4\pi \beta d\beta \sin^2 \theta' = 2.6595 \quad (25)$$

Fig. 3
Schematic diagram of apparatus

Fig. 4 Inner sphere and mechanism of weighing

$$p_{AII} + p_{BII} - p_{AI} \cong p_i - p_{AI} \cong p_{BII} \quad (50)$$

$$\gamma \cong - \left\{ \zeta \left(1 - \frac{1}{R^*} \right) \right\}^{-1} \ln \left(\frac{p_{BII}}{p_{BII}} \right) \quad (51)$$

because the value of p_i is approximately constant. Therefore

$$\begin{aligned} N_{AI} &\cong - \left(\frac{m_A}{2\pi kT} \right)^{1/2} \left(\frac{m_A \pi}{2kT} \right)^{1/2} \frac{2p_i [D_{AB}]}{R_i (1 - 1/R^*)} \ln \left(\frac{p_{BII}}{p_{BII}} \right) \\ &\cong \frac{m_A p_i [D_{AB}]}{kTR_i (1 - 1/R^*)} \frac{p_{AI} - p_{AII}}{(p_B)_{im}} \end{aligned} \quad (52)$$

The above equation is Stefan's equation.

The distributions of the total pressure and the partial pressure of component A are expressed by

$$\begin{aligned} p_i &= \frac{1}{2} \{ (1 - \sin \alpha) (p_{A1} + p_{B2}) \\ &\quad + (1 + \sin \alpha) (p_{A2} + p_{B1}) \} \\ &= \frac{1}{2} (\delta - \gamma \sin \alpha) \end{aligned} \quad (53)$$

and

$$p_A = \frac{1}{2} \left\{ \delta + M \exp \left(-\frac{\zeta \gamma}{R} \right) - \gamma \sin \alpha \right\} \quad (54)$$

2. Experimental Apparatus and Procedures

The experimental apparatus used is schematically shown in Fig. 3.

The measurements of the sublimation rate of naphthalene coated on the inner sphere were performed under reduced pressure. The inner sphere was placed at the center of the outer sphere working as a condensating surface.

The outer sphere, the sublimation chamber (150^{mm} I. D. × 160^{mm} O. D.), was made of bronze and jacketed by the copper cooling pipe through which methanol cooled by a refrigerator, 0 ~ -40°C, was circulated.

The inner sphere (about 22 ~ 30^{mm} ϕ) was the copper spherical shell having 1.5 mm thickness in which the electric heater was provided, and was coated with naphthalene about 2 mm thickness and located concentrically in the outer sphere (see Fig. 3 and Fig. 4). The coating of naphthalene was carried out by dipping the inner sphere into a bath of molten naphthalene purified by recrystallization. The inner sphere was electrically heated by supplying D.C. current and the surface temperature of naphthalene was regulated to be constant by a variometer and determined from the temperature of the surface of inner sphere taking account of the difference through the layer.

To determine the rate of sublimation, the weight loss of naphthalene on the inner sphere was measured by the displacement meter using the differential transformer (see Fig. 3 and Fig. 4) and recorded continuously.

For pressure measurement, Atlas-MMM diaphragm vacuum gauge for relatively low pressures and oil manometer for relatively high pressures were used. The measured value of the pressure at the outer sphere by using vacuum gauge, p_D , is

$$\begin{aligned} p_D &= \frac{1}{2} \{ (1 + \cos 2\alpha) p_{AII} + (1 - \cos 2\alpha) p_{AII} \} + p_{BII} \\ &= \left(\frac{1}{R^*} \right)^2 p_{AII} + \left\{ 1 - \left(\frac{1}{R^*} \right)^2 \right\} p_{AII} + p_{BII} \end{aligned} \quad (55)$$

From Eq. (55), the value of p_{BII} can be calculated.

In the experiments, the total pressures of the apparatus were kept constant in the range 10⁻¹ ~ 10⁻⁴ mmHg by a variable leak valve.

Run No.	t_I' [°C]	Δt_I [°C]	t_I [°C]	t_{II} [°C]	$p_{AI} \times 10^3$ [mmHg]	$p_{AII} \times 10^3$ [mmHg]	$p_D \times 10^3$ [mmHg]	$p_{BII} \times 10^3$ [mmHg]	$N_{AI} \times 10^5$ [g/cm ² ·sec]	$k_g \times 10^3$ [g/cm ² ·sec·mmHg]
1	10.4	0.7	9.7	-0.2	18.0	5.90	84.1	78.1	3.62	3.00
2	10.4	0.5	9.9	-0.3	18.2	5.82	143.0	137.1	2.32	1.80
3	0.2	1.2	-1.0	-10.9	5.40	1.65	10.7	9.00	5.14	13.7
4	-0.3	0.6	-0.9	-9.3	5.48	2.01	28.7	26.7	2.96	8.54
5	-9.7	0.8	-10.5	-30.9	1.75	0.118	2.25	2.10	3.78	23.2
6	-10.4	0.9	-11.3	-30.0	1.58	0.133	3.78	3.62	3.50	24.1
7	-9.7	0.7	-10.4	-30.0	1.75	0.133	7.56	7.40	2.98	18.4
8	-10.2	0.5	-10.7	-28.3	1.70	0.169	22.0	21.8	1.51	9.84
9	-10.0	0.4	-10.4	-29.3	1.75	0.146	61.6	61.4	0.621	3.88
10	-19.3	0.2	-19.5	-26.9	0.55	0.205	2.09	1.88	0.703	20.1
11	-20.2	0.4	-20.6	-29.0	0.485	0.152	2.90	2.74	1.10	33.0
12	-19.7	0.2	-19.9	-29.3	0.530	0.146	7.25	7.10	0.700	18.3
13	-20.0	0.2	-20.2	-29.0	0.510	0.153	23.3	23.2	0.281	7.87
14	-19.3	0.2	-19.5	-28.1	0.560	0.174	24.0	23.8	0.355	9.20
15	-20.2	0.2	-20.4	-29.7	0.495	0.139	72.5	72.4	0.127	3.58
16	0.0	0.6	-0.6	-9.8	5.60	1.90	35.2	33.3	2.50	6.74
17	-10.0	0.8	-10.8	-30.5	1.67	0.124	5.14	4.97	3.69	23.8
18	-10.0	0.8	-10.8	-29.3	1.67	0.146	6.32	6.14	3.67	24.1
19	-9.9	0.6	-10.5	-29.5	1.74	0.143	13.2	13.0	2.43	15.1
20	-9.9	0.6	-10.5	-30.0	1.74	0.133	16.5	16.3	2.00	12.4
21	-10.0	0.9	-10.9	-31.0	1.65	0.115	3.26	3.10	4.32	28.1
22	-9.9	0.9	-10.8	-30.5	1.67	0.125	4.31	4.15	3.94	25.4
23	-9.9	0.5	-10.4	-30.8	1.76	0.119	24.9	24.8	1.25	7.62
24	-10.0	0.7	-10.7	-30.8	1.70	0.119	8.68	8.53	3.13	19.8
25	-9.7	0.4	-10.1	-29.8	1.83	0.136	40.5	40.4	0.965	5.71
26	-10.0	0.7	-10.7	-30.6	1.70	0.122	9.33	9.18	3.21	20.3

t_I : temperature of the surface of copper shell
 Δt_I : temperature difference through the coated layer of naphthalene
The data of vapor pressure, thermal conductivity and diffusivity of naphthalene were cited from I. C. T..

Fig. 5 Comparison between analytical and experimental results

The procedures of the experiments were as follows; after hanging the coated inner sphere from a hook below the core of differential transformer, the sublimation chamber was evacuated for several minutes to the prescribed pressure and the outer sphere was cooled to the prescribed temperature.

Then the inner sphere was electrically heated for the surface temperature to be constant. The weight loss

of the inner sphere and the temperatures of the surfaces were recorded continuously.

The measurements and regulations of the temperatures of the inner and outer sphere were carried out within $\pm 0.1^\circ\text{C}$ and $\pm 0.4^\circ\text{C}$ in errors, respectively.

The relative error on the determination of sublimation rate was estimated to be in $\pm 2\%$.

Fig. 6 Comparison between analytical and experimental results

Fig. 7 Distributions of pressure

3. Experimental Results and Discussions

The mass transfer rate based on the surface of the inner sphere, N_{AI} , was calculated from the rate of weight loss of naphthalene by using the following equation,

$$N_{AI} = -\frac{1}{4\pi R_I^2} \frac{dW}{dt} \quad (56)$$

The value of $p_{B1\infty}$ was calculated from the value of p_D measured by the vacuum gauge by using Eqs.(55), (31) and (37).

$$p_D = \gamma + p_{B1\infty} + \frac{1}{2} \left\{ -\gamma + \delta + M \exp\left(-\frac{\zeta r}{R^*}\right) \right\} \quad (57)$$

From Eqs.(45) and (57), we obtain the following equation;

$$p_{B1\infty} = p_D - p_{A\infty} - \frac{1}{a} \left(\frac{1}{R^*} \right)^2 \gamma \quad (58)$$

The experimental results are shown in **Table 1**. Further the values of $p_{B1\infty}$ in Table 1 were calculated from Eq.(58) assuming that the value of a was equal to unity, because the error produced by this approximation was negligibly small in the final value.

In **Fig. 5**, the observed values of the sublimation rate are compared with the calculated values from Eq.(43). The observed values fairly agree with the calculated ones from the continuum to the transition region. It can be observed that in the free molecular region the observed values are a little smaller than the calculated ones as shown in **Fig. 5**.

The observed values of k_g are also compared with the calculated values from Eq.(44) in **Fig. 6**. From **Fig. 6** it may be said that the accommodation coefficient a for naphthalene is equal about to 0.9. It has been reported by Sherwood et al.⁹⁾ that this coefficient was equal to 0.88 about at -60°C .

The distributions of the total pressure and the partial pressures of component A and B are calculated by using Eqs.(53) and (54). An example is shown in **Fig. 7**. The pressure-jump is observed distinctly on the surface of inner sphere and the total pressure changes steeply near the inner sphere.

Conclusion

The equation of mass transfer between concentric spheres under reduced pressures, especially under the transition regime, was derived through the gas kinetic theory using the two-sided Maxwellian distribution function. The observed values of the sublimation rate of naphthalene were compared with the values calculated from the equation derived in the present work and both were in good agreement.

Acknowledgements

We wish to thank Mr. Yoshihiro Konaka and Mr. Masashi Asaeda for technical assistance. This work was supported in part

by the Kawakami Memorial Foundation, to which our thanks are due.

Nomenclature

A_1	= constant defined by Eq. (25)	[—]
a	= accommodation coefficient for mass transfer	[—]
b	= impact parameter, or perpendicular distance from molecule A to initial trajectory of molecule B (see Fig. 2)	[cm]
$[D_{AB}]$	= diffusion coefficient of molecule A through molecule B	[cm ² /sec]
F	= intermolecular force for the Maxwell molecule	
$F(a)$	= the function of accommodation coefficient: $\frac{1}{a} + \frac{1-a}{a} \left(\frac{1}{\bar{R}^*} \right)^2$	[—]
f	= velocity distribution function of molecule	[—]
$\bar{K}_{AB}$	= constant in expression for central force field, $F = \frac{\bar{K}_{AB}}{r^5}$	
k	= Boltzmann constant	[mmHg·cm ³ /°K]
k_g	= mass transfer coefficient	[g/cm ² ·sec·mmHg]
M	= integral constant defined in Eqs. (36)~(38)	[mmHg]
m	= mass of a molecule	[g]
N_{AI}	= mass transfer rate based on the inner sphere	[g/cm ² ·sec]
n	= number density of molecule	[1/cm ³]
P	= component of pressure tensor	[mmHg]
p	= pressure defined by $p=nkT$	[mmHg]
Q	= physical quantity	
ΔQ	= change in Q produced by collision; collision integral	
R	= radial distance from the center of the inner sphere	[cm]
$\bar{R}$	= normalized distance from the center of the inner sphere; $\bar{R}=R/R_I$	[—]
$\bar{R}^*$	= R_I/R_I	[—]
r	= distance between two molecules colliding each other	[cm]
T	= absolute temperature	[°K]
t	= time	[sec]
V	= relative velocity between two interacting molecules	[cm/sec]
W	= weight of the inner sphere	[g]

Greek letters

α	= angle defined by Eq. (4)	[—]
β	= variable concerned in variable b (cf. literature 5)	

γ	= integral constant defined by Eq. (31)	[mmHg]
γ'	= integral constant defined by Eq. (14)	[1/cm ³]
δ	= integral constant defined by Eq. (21)	[1/cm ³]
ϵ	= angle between plane of the orbit and plane containing the original relative velocity and the X -axis in a binary collision (see Fig. 2)	[—]
θ'	= scattering angle, or angle between relative velocity before and after collision	[—]
ξ	= velocity of molecule	[cm/sec]
ξ	= velocity vector of molecule	[cm/sec]
ϕ	= angle defined in Fig. 1	[—]
ζ	= constant defined by Eq. (35)	[1/mmHg]

Suffix

A	= diffusing component
B	= stagnant component
R	= radial direction on polar co-ordinate
D	= pressure directed outwardly at the inner surface of the outer sphere
t	= total pressure
θ	= θ -direction on polar co-ordinate
ϕ	= ϕ -direction on polar co-ordinate
1	= inside of the wedge of "influence" (see Fig. 1)
2	= outside of the wedge of "influence" (see Fig. 1)
I	= the inner sphere
II	= the outer sphere
< >	= averaged value
'	= after collision

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