Iron Droplet Formation Due to Bubbles Passing through Molten Iron/Slag Interface

Saburo KOBAYASHI

Institute for Advanced Materials Processing, Tohoku University, Katahira, Aoba-ku, Sendai, Miyagi-ken, 980 Japan.

(Received on December 7, 1992; accepted in final form on February 25, 1993)

Iron suspension is formed in slag when gas bubbles pass through molten iron/slag interface. This paper proposes a model to explain iron droplet formation and to estimate thereby the behavior of the droplets in slag.

When a gas bubble passes through molten iron/slag interface, an iron film is considered to be formed on the bubble surface. It was predicted that the film reached a constant thickness within 0.5 s at the longest. The probable range of the mass of the iron droplet formed from the film was evaluated with the thickness and the surface area of the film. The experimental values of the mass of droplet fell within the predicted range. The movement of the iron droplets in the slag phase was estimated taking the surface energies of bubble and iron film as energy source. The penetration distance and the residence time of droplets in the slag phase were evaluated from 1 to 6 cm and from 1 to 0.1 s, respectively. These small values suggest possible high speed recirculation of iron droplets between the iron and slag phases through the interface.

KEY WORDS: gas bubble; smelting reduction; iron droplet; iron emulsion; droplet formation; droplet size; iron film; surface energy; movement of droplet.

1. Introduction

It has been reported that the iron suspension in slag increased with increasing flow rate of bottom blowing gas in in-bath smelting reduction.¹⁾ Since the increase in the iron suspension results in the decrease in the post combustion ratio, it is intended to depress the iron suspension. So far, it has been well known that iron suspension in slag took place when gas bubbles passed through an iron/slag interface,^{2,3)} but this phenomenon is still one of practically important problems in developing recent new iron smelting processes.

Using the mercury/water–glycerin and mercury/ethyl alcohol systems, Davenport *et al.* observed the behavior of argon bubbles when they rose through mercury phase and then passed through the interfaces between the two phases.^{4,5)} According to their observation, the bubble with a mercury film on its surface moves upward to the upper phase, in which the film ruptures to form mercury droplets. The droplets remain in the upper phase for some time, and then return to the mercury phase. Davenport *et al.* further performed a series of argon bubbling tests for lead/fused salt and copper/slag systems, and found that the amount of metal droplets formed in the upper phase increased with increasing flow rate of gas.

Recently, in order to investigate the gas injection effect on the desulfurization by CaO–Al₂O₃–SiO₂ slag, Deng injected argon gas from the bottom of a crucible filled with molten iron/slag and found many iron droplets in the slag samples.⁶⁾ According to the tests, the size of iron droplets ranged from several micrometers to several

millimeters, but calculation revealed that the mass-mean size deviated toward several millimeters.

The iron emulsion forms additional interface in slag phase which may enhance the metal/slag interfacial reaction rate. The actual extent of the enhancement depends on their residence time in slag phase as well. Therefore it is necessary to estimate kinetic behavior of droplets in slag.

No theory have been proposed yet to explain the size of iron droplets formed in slag and their behavior. This paper proposes a model to explain the iron droplet formation for molten iron/slag system and estimates thereby the behavior of the droplets in slag.

2. Iron Droplet Formation Mechanism

From the analogy of the behavior of a bubble passing through the mercury/water–glycerin interface,⁴⁾ when a bubble floating up through the stationary molten iron passes through the horizontal iron/slag interface, an iron film is formed on the bubble above the interface as shown on the left hand side in Fig. 1. If the film is thermodynamically unstable, it may be followed by rupture and may subsequently split into droplets by some stimulation such as displacement of the bubble by crossing the interface.

The parameters X , Y and Z which Conochie *et al.* have already proposed are available to predict whether the iron film is stable or unstable; the film is unstable when $Z < 0.5$.⁷⁾ These parameters are defined in Appendix 1. As described later, the surface tensions of molten iron

Fig. 1. Formation of a film on bubble.

and slag under consideration are 1.2–1.7 and 0.2–0.7 N/m respectively, and the interfacial tension between iron and slag is 1–1.5 N/m. Using these values, $Z=0.06$ –0.24, that is, the iron film is unstable in the present system.

For the other parameters, $X=0.35$ –0.43, $Y=0.4$ –0.5. These values suggest that floatation or dispersion of iron droplets is thermodynamically stable. In fact, the kinetics of rupture of the film is, however, considered to play a definitive role in subsequent behavior of the droplets, since an instantaneous release of the large amount of surface energy may cause a violent motion of iron as described below.

According to Pruppacher *et al.*, on the other hand, the droplets formed when the bubble floating up through water is released into the air consist of the two kinds; the fine “film” ones generated by the rupture of the water film formed in the aerial part of the bubble and the comparatively large “jet” ones generated by surface wave.⁸⁾ These droplets are released upward. A part of the mass of water film forms the “film” droplets. Most part of it drops downward along the surface of a cave formed on the surface of liquid, and then turns at the bottom of the cave, moving upward in the form of a column. The column then splits into the several “jet” droplets. The energy source of the formation and movement of each droplet is the surface energy of the film. When a metal film collapses in the slag phase, iron droplets may be released into the slag phase by the same mechanism as mentioned above.

So far, the size of a film under consideration has been studied with respect to binary phase systems; liquid/liquid and gas/liquid. The size is evaluated in the dynamic equilibrium state between the phases I and II as shown on the right hand side in Fig. 1. When the phases I and II are gas and liquid, respectively, the bubble in the liquid forms the bulge of liquid film in the upper gas phase. If the bubble is so small that its spherical shape may be maintained and its film thickness is so thin that the gravitational effect of the film may be negligible, then the area of the liquid film A_{1e} can be written as follows⁹⁾

$$A_{1e} = (2\pi/3) c_1 r_b^4 \quad \dots\dots\dots (1)$$

$$c_1 = (\rho_{II} - \rho_I) g / \sigma \quad \dots\dots\dots (2)$$

where r_b is radius of bubble, ρ_i density of phase i , g gravitational acceleration, and σ interfacial tension between phases I and II. These equations were derived from the balance between the buoyancy force of the bubble and the vertical component of the surface tension of the liquid film.

Fig. 2. Schematic diagram of droplet formation by bubble.

3. Iron Droplet Formation Model

The above equations are now applied to the bubble in the molten iron/slag interface as shown on the left hand side in Fig. 1. These equations must be modified to fit the three phase system under consideration. The term of bubble's buoyancy force $(\rho_{II} - \rho_I)g$ is approximated with buoyancy force for gas in iron, and the term of the interfacial tension with a mean value of iron/gas and iron/slag interfacial tensions. In this case the following equation is used in place of Eq. (2).

$$c_1 = (\rho_m - \rho_g) g / \sigma \quad \dots\dots\dots (3)$$

$$\sigma = (\sigma_{m-g} + \sigma_{m-s}) / 2 \quad \dots\dots\dots (4)$$

where ρ_m and ρ_g are densities of metal and gas, and σ_{m-g} and σ_{m-s} interfacial tensions of metal–gas and metal–slag, respectively.

It is extremely difficult to evaluate accurately the stability of iron film, and the size and number of the “film” or “jet” droplets. Here, it is attempted to evaluate the formation of iron droplets and kinetic characteristics by using a simple model.

The assumptions for the evaluation are as follows:

- (1) The bubble is spherical and its volume is constant.
- (2) The collapse of the iron film takes place in the equilibrium state mentioned above.
- (3) All of the surface energy of iron film is converted to surface energy required for the formation of iron droplets in the slag and to kinetic energy of iron droplets.
- (4) One iron film produces one droplet with the same mass as that of the film.
- (5) The bubble in the iron has a terminal rising velocity.

On these assumptions, the droplet formation by bubble may be depicted as in Fig. 2.

The physical properties of molten iron and slag are given in Table 1. For comparison of the present simulation with the experimental result obtained by Deng,⁶⁾ the effects of [S] and [O] on surface tension of iron and interfacial tension of iron/slag were taken into account. Although the effect of composition on surface and interfacial tensions is very complicated, they may be summarized as $\sigma_{m-g} = 1.2$ –1.7, $\sigma_{s-g} = 0.2$ –0.7 and $\sigma_{m-s} = 1$ –1.5 N/m for the chemical composition shown in Table 1.

The interfacial tension seems to be smaller than the surface tension of iron by about 0.4 N/m. This difference

Table 1. Physical properties of iron and slag (1600°C).

	Surface tension (N/m)	Density ⁽¹⁰⁾ (g/m ³)	Viscosity ⁽¹⁰⁾ (Pa·s)
Iron		7	$6 \cdot 10^{-3}$
[O] = 0.02–0.04	1.6–1.4 ⁽¹²⁾		
[S] = 0.001–0.1	1.7–1.2 ⁽¹¹⁾		
Slag	0.2–0.7 ⁽¹²⁾	2.7	0.1–1
Iron/Slag (CaO–Al ₂ O ₃ –SiO ₂)			
$a_o = 10^{-2}$ – 10^{-3}	1.1–1.5 ^{(11)*}		
$a_s = 10^{-1}$ – 10^{-3}	0.9–1.3 ^{(11)*}		

a_o and a_s are activities of oxygen and sulfur in iron, respectively.

* means interfacial tension.

is, however, too small to cause significant variation in the simulation result. For example, the value of σ in Eq. (4) is almost same regardless of the difference. Hence, it is assumed that $\sigma_m = \sigma_{m-g} = \sigma_{m-s}$.

The surface tension of slag is smaller than the interfacial tension. The contribution of the surface tension of slag to the formation and the movement of the droplet is a little because of its small value. There was no significant difference in the simulation results between $\sigma_{s-g} = 0.2$ and 0.5 N/m.

In the present work, the simulation results with the following parameter values are shown.

$$\sigma_{m-g} = \sigma_{m-s} = 1 \quad \text{and} \quad 1.5 \text{ N/m}$$

$$\sigma_{s-g} = 0.2 \text{ N/m}$$

$$\rho_m = 7 \text{ g/cm}^3$$

$$\rho_g = 2.7 \text{ g/cm}^3$$

$$\eta_s = 0.5 \quad \text{and} \quad 1 \text{ Pa} \cdot \text{s}$$

where η_s is the viscosity of slag.

4. Formation of Iron Film and Droplets

The first step of the process is the formation of the equilibrium iron film. The energy required for the process is considered below. First of all, it is considered whether the kinetic energy accompanied by the bubble floating up with the terminal velocity is enough to form the equilibrium film. If the kinetic energy is completely used for the film formation, in other words, if the bubble stops on the interface, then the energy balance can be expressed by the following equation

$$(1/2)k\rho_m V_b u_{bt}^2 = (A_1 - A_2)\sigma_{m-s} \quad \dots\dots\dots (5)$$

where V_b is the volume of bubble, u_{bt} the terminal velocity of bubble,⁽¹³⁾ and A_1 and A_2 are the area of the film and the base area of the spherical cap respectively. The term $k\rho_m V_b$ is the virtual mass associated with the bubble, and $k=1/2$. The kinetic energy of the mass of the gas contained in the bubble was neglected because $\rho_m \gg \rho_g$.

Figure 3 shows the area ratios A_{1e}/A_0 and A_1/A_{1e} obtained by using Eqs. (1) and (5). d_b and A_0 stand for the diameter and the whole surface area of bubble, respectively. To compare with the results of experiments later, 0.5 to 2.0 cm are taken as d_b . In Fig. 3, the solid line (A_1/A_{1e}) almost lies below 0.8, suggesting that the kinetic energy of bubble is not enough to form an

Fig. 3. Bubble size dependence of surface area ratios.

Fig. 4. Relation between work ratio W_s/W_b and θ .

equilibrium film.

The next problem is the energy due to the buoyancy force of the bubble. The film formation results in the net increment of iron/slag interface area $\pi r_b^2 (1 - \cos \theta)^2$ when the bubble has been located as shown on the left hand side in Fig. 2. Therefore, the surface energy required for the film formation can be written as the following equation

$$W_s = \pi r_b^2 (1 - \cos \theta)^2 \sigma_{m-s} \quad \dots\dots\dots (6)$$

On the other hand, while a bubble passes through the bulk interface, the bubble is located across two phases. Because the buoyancy force of the bubble depends on the volumes occupying in both the slag and the iron, the work W_b done by the floating up bubble can be expressed by the following equation, taking into account the above two factors.

$$W_b = (\pi r_b^4 g / 3) \{ (\rho_s - \rho_g) [-(\cos \theta)^4 / 4 - 3(\sin \theta)^2 / 2 - 2 \cos \theta + 9/4] + (\rho_m - \rho_g) [(\cos \theta)^4 / 4 + 3(\sin \theta)^2 / 2 - 2 \cos \theta + 7/4] \} \quad \dots\dots\dots (7)$$

The derivation of this equation is given in Appendix 2. The first and second terms on the right hand side of the equation imply the contributions of slag and metal, respectively.

The energy ratio W_s/W_b is shown in **Fig. 4**. The right hand end of each curve is equal to the angle θ at the time of the equilibrium film formation, and the ratio is less than 0.3. This fact suggests that the work done by the buoyancy force is adequate to form the equilibrium film. Then, the following assumption is made.

(6) The energy source for the metal film formation is the buoyancy work of bubble.

Upon deriving Eq. (1), it is assumed that the mass of film is negligible, but no limitation is made to the thickness of the film. This means that the thickness of the equilibrium film is still variable within the range in which the effect of mass is negligible. The change in the thickness of film can be approximated by the Reynolds equation⁹⁾

$$-dy/dt = \{2\pi F / (3\eta A^2)\} y^3 \quad (8)$$

representing the thinning rate of the gap thickness y due to the drain away of liquid from the gap between two parallel solid disks to which forces F 's are applied from the both sides. In this equation, η and A are viscosity of liquid and surface area of disk, respectively. In the present case, F is the buoyancy force by bubble.

$$F = (\pi/6)(\rho_m - \rho_g)gd_b^3 \quad (9)$$

Taking into consideration the fact that $\rho_m \gg \rho_g$, the thinning rate of film u_f can be represented by the following equation

$$-dy_f/dt = u_f = ay_f^3 \quad (10)$$

$$a = N\sigma/(\eta_m c_1 (r_b)^5) \quad (11)$$

where y_f is film thickness, η_m viscosity of molten iron and $N=2$ for spherical bubble.

To use the Reynolds equation, one must define the term "film". Figure 5 shows u_f given by Eq. (10) and the terminal velocity of bubble u_{bt} . The rising velocity of bubble is equal to u_{bt} on the right hand side of the intersection of both curves, and follows u_f given by Eq. (10) on its left side. It is reasonable, therefore, to define the left side of the intersection as film, i.e., coordinate y_{f0} as the initial thickness of film. The plot of y_{f0} vs. d_b is shown in Fig. 8. Although y_{f0} increases with d_b , the effect of surface tension is small.

Integration of Eq. (10) with the initial condition, $y_f = y_{f0}$ and $u_f = u_{bt}$ at $t = 0$ yields thickness y_f and mass m_d of film

$$Y_f = y_f/y_{f0} = 1/(1 + \tau)^{1/2} \quad (12)$$

$$m_d = A_{1e} y_f \rho_m \quad (13)$$

where

$$\tau = \alpha t \quad (14)$$

Fig. 5. Definition of "film" and initial film thickness y_{f0} .

$$\alpha = 2(au_{bt}^2)^{1/3} \quad (15)$$

Dimensionless quantities M_d and U_f are defined as follows.

$$M_d = m_d/m_{d0} \quad (16)$$

$$U_f = u_f/u_{bt} \quad (17)$$

where m_{d0} is the initial mass of film.

Figure 6 shows that α is in the range from 200 to 600 (s^{-1}). Figure 7 shows the variations of Y_f , M_d and U_f with time. Y_f decreases rapidly when $\tau < 20$, but extremely slowly when $\tau > 100$. Accordingly, the actually obtainable values of Y_f and M_d may range from 1 to 0.1, because the magnitude of U_f reduces to 1/100 of the initial velocity already at $\tau = 20$. As described above, the minimum value of α is about 200. When $\alpha = 200$, $\tau = 100$ corresponds to the real time $t = 0.5$ s, so film thickness will in fact reach a certain value after 0.5 s at the longest. These considerations reveal that the range of the mass of film is actually $0.1 < M_d < 1$, and that the quantity is achieved shorter than 0.5 s. According to assumption (2), this is the mass of the film to collapse.

According to assumption (4), M_d is the mass of one iron droplet. Figure 8 shows the plots of the droplet mass m_{d0} and m_{d01} at $M_d = 1.0$ and 0.1 respectively. The bubble size dependence of the droplet mass is greater than that of film thickness because of the stronger bubble size dependence of film surface area. According to the present model, the mass of iron droplet lies between the curves for m_{d0} and m_{d01} ; for example, when $\sigma_m = 1.5$ N/m, the mass of the droplet which one bubble with 1.5 cm

Fig. 6. Relation between α and bubble size.

Fig. 7. Changes of Y_f , M_d and U_f with time.

Fig. 8. Comparison of observed mass of iron droplet (●) with calculated.

Fig. 9. Dependence of sizes of iron droplet and bubble (1600°C) on argon flow rate in the desulfurization experiment of iron.⁶⁾ Slag: ca. 50%CaO, ca. 3%SiO₂, ca. 35%Al₂O₃ and others. [S]=0.12–0.001 mass%, [O]=0.01–0.03 mass% (estimated).

diameter may form is predicted to be 0.1 to 1 g.

Figure 9 depicts the data Deng obtained in the argon bubbling experiments for the molten iron/slag system.⁶⁾ d_b is the values at 1600°C, and d_d is the mean diameter of the iron particles contained in the sampled slag. The mean mass of the particle is shown already in Fig. 8. The observed mass lies between the two predicted curves, proving that the present model well approximates the actual formation process of iron droplet. The bubble size dependence of the observed values seems to be weaker than that of predicted curves. The increase in bubble size may make deviation from spherical approximation greater, or may result in further ruptures of droplet, thereby reducing possibly the adequacy of the assumptions in the present model. The latter phenomenon clearly deviates the mass of droplet toward a smaller value.

5. Movement of Iron Droplet in Slag Phase

On the basis of assumption (3), the formation and the movement of droplet are determined from the balance with surface energy. In other words, since the bubble floats up not only through the molten iron phase but also through the slag, the kinetic energy of the bubble itself may be considered to be maintained over all processes and to have almost nothing to do with the formation and the movement of iron droplet.

Fig. 10. Residence time and penetration distance of iron droplet in slag. $\sigma_m = 1.5$ N/m; l_1 for $\eta_s = 0.5$ Pa·s, l_2 for $\eta_s = 1$ Pa·s; $d_b = 1.0$ cm for $0.01 < m_d < 0.1$ g; $d_b = 1.5$ cm for $0.1 < m_d < 1$ g; $d_b = 2.0$ cm for $1 < m_d < 10$ g.

The following equation represents the energy balance before and after the droplet formation.

$$\pi d_b^2 \sigma_{m-g} + A_{1e} \sigma_{m-s} = N_d \pi d_d^2 \sigma_{m-s} + \pi d_b^2 \sigma_{s-g} + A_{2e} \sigma_{m-s} + N_d (1/2) \rho_m V_d u_{d0}^2 \quad (18)$$

where N_d is number of droplets formed from one bubble, V_d volume of droplet, and u_{d0} initial velocity of it. $N_d = 1$ from assumption (4). The first and second terms on the left hand side of the equation are the surface energies of the bubble in molten iron and of the metal/slag interface of the film, respectively. The first and second terms on the right hand side represent the surface energies of the droplet and the bubble in slag phase, respectively, whereas the third and fourth terms are the surface energy of the iron/slag interface formed newly after the annihilation of the film and the kinetic energy of the droplet, respectively.

The droplets may move in various direction with the initial kinetic energy expressed by the last term in Eq. (18). Here, discussion is confined only to the movement in the vertical direction. Provided the drag force acting on the droplet in the slag is assumed to follow the Stokes law, the equation of motion for the droplet in the stationary slag can be written as follows

$$-du_d/dt = b_0 + b_1 u_d \quad (19)$$

$$b_0 = (\rho_m - \rho_s)g/\rho_m \quad (20)$$

$$b_1 = 3\pi\eta_s d_d/m_d \quad (21)$$

where u_d is vertical velocity of droplet and its initial velocity u_{d0} is obtained from Eq. (18).

From Fig. 8, in the case of $\sigma_m = 1.5$ N/m, m_d has 0.01 to 0.1, 0.1 to 1, and 1 to 10 g as significant values for bubble sizes 1.0, 1.5 and 2.0 cm, respectively. Conversely, there are innumerable bubble sizes that can give these values of m_d . The three values above of bubble size are taken as typical ones, which will be used in the following calculation when m_d is a variable. Now, for simplification, the position of the droplet formation is taken on the interface of the bulk phases.

Figure 10 shows the vertical penetration distance l and the residence time t_r of droplet in the slag phase, both of which were obtained from Eq. (19). l_1 and l_2 are the

penetration distances for $\eta_s = 0.5$ and $1 \text{ Pa} \cdot \text{s}$, respectively. The logarithm of u_{d0} decreases linearly with the increasing droplet mass. Since bubble size was changed discretely, the curves become stepwise. u_{d0} is independent of viscosity of slag. The t_r curves for $\eta_s = 0.5$ and $1 \text{ Pa} \cdot \text{s}$ are almost superposed upon each other in Fig. 10, which means the extremely small effect of slag viscosity on the residence time. The slopes of the u_{d0} and t_r curves are almost same. As m_d increases, l_1 and l_2 increase for $d_b = 1.0 \text{ cm}$, almost constant for 1.5 cm , and decrease, coming close to each other for 2.0 cm . In Fig. 10, u_{d0} ranges from 10 to 0.6 m/s , l_1 from 1 to 6 cm and t_r from 1 to 0.1 s . The values of the last two quantities are very small. This suggests possible high speed recirculation of droplets between the two phases through the interface. The upward convection of the slag associated with the bubble rising may retard the recirculation by increase in the penetration distance and the residence time.

6. Conclusions

(1) It was confirmed that the work done by the buoyancy force acting on the bubble provided the surface energy required for the iron film to be formed on the bubble surface when the bubble rising in the molten iron passed through iron/slag interface.

(2) The probable range of thickness of iron film was evaluated by the thinning rate equation of the film. It was predicted that the film reached a constant thickness within 0.5 s at the longest.

(3) The probable range of the mass of the iron droplet was evaluated with the thickness and the surface area of the film. The experimental values of the mass of the droplet fell within the predicted range.

(4) The movement of the iron droplets in the slag phase was estimated taking the surface energies of bubble and iron film as energy source. The penetration distance and the residence time of droplets in the slag phase were evaluated from 1 to 6 cm and from 1 to 0.1 s , respectively. These small values suggest possible high speed recirculation of iron droplets between the iron and slag phases through the interface.

Acknowledgment

A part of the present study was performed in Berlin Technical University, Germany in 1990. The author wishes to thank Professor F. Oeters for support and is indebted to Dr. L. Zhang for assistance.

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Appendix 1

Conochie *et al.* considered a situation in which one gas bubble was formed in a bulk liquid phase and then small amount of another immisible liquid was put on the surface of the bubble.⁷⁾ They used parameters X , Y and Z to predict thermodynamically stable states of the second liquid in the situation. The parameters are written as

$$X = \sigma_{m-s}/\sigma_t \quad \text{.....(A1-1)}$$

$$Y = \sigma_{m-g}/\sigma_t \quad \text{.....(A1-2)}$$

$$Z = \sigma_{s-g}/\sigma_t \quad \text{.....(A1-3)}$$

$$\sigma_t = \sigma_{m-s} + \sigma_{m-g} + \sigma_{s-g} \quad \text{.....(A1-4)}$$

for the present work.

The stable states are predicted as follows:

- (1) film for $Z > 0.5$,
- (2) dispersion for $Y > 0.5$,
- (3) floatation for $Z < 0.5$, $Y < 0.5$ and $X < Y$.

Appendix 2

For the bubble shown on the left side in Fig. 2, the partial volumes of bubble above and below the iron/slag interface are given respectively by

$$V_s = (\pi r_b^3/3) \{ (\cos \theta)^3 - 3 \cos \theta + 2 \} \quad \text{.....(A2-1)}$$

and

$$V_m = (\pi r_b^3/3) \{ -(\cos \theta)^3 + 3 \cos \theta + 2 \} \quad \text{.....(A2-2)}$$

The buoyancy force of the bubble F_b is given by

$$F_b = (\rho_s - \rho_g)gV_s + (\rho_m - \rho_g)gV_m \quad \text{.....(A2-3)}$$

The work done by F_b is given by

$$\begin{aligned} W_b &= r_b \int_0^\theta F_b \sin \theta d\theta \\ &= (\pi r_b^4 g/3) \{ (\rho_s - \rho_g) [-(\cos \theta)^4/4 - 3(\sin \theta)^2/2 \\ &\quad - 2 \cos \theta + 9/4] + (\rho_m - \rho_g) [(\cos \theta)^4/4 \\ &\quad + 3(\sin \theta)^2/2 - 2 \cos \theta + 7/4] \} \quad \text{.....(A2-4)} \end{aligned}$$