MECHANISTIC ANALYSIS AND COMPUTER SIMULATION OF IMPACT BREAKAGE OF AGGLOMERATES: EFFECT OF SURFACE ENERGY

R. Moreno-Atanasio and M. Ghadiri

Institute of Particle Science and Engineering, University of Leeds, Leeds, LS2 9JT UK.

ABSTRACT

Agglomerates are ubiquitous as intermediate or manufactured products in chemical, pharmaceutical and food industries. During handling and processing they may suffer breakage if they are weak. On the other hand, if they are too strong, their dispersion and disintegration could be difficult. The control of their mechanical strength is therefore highly desirable. However, the analysis of agglomerate strength is complex due to the large number of parameters that influence agglomerate behaviour, such as the primary particle size, density and elastic modulus, and the interparticle bond strength.

A simple mechanistic model is presented here which relates the number of broken contacts in agglomerate due to impact velocity, interparticle adhesion energy and the particle properties of the particles forming the agglomerate. The model is based on the hypothesis that the energy used to break contacts during impact is proportional to the incident kinetic energy of the agglomerate. The damage ratio defined as the ratio of broken contacts to the initial number of bonds is shown to depend on the dimensionless group, Δ, in the form $(\rho V^2 D^{5/3} E^{2/3})/\Gamma^{5/3}$, where V is the impact velocity, E the elastic modulus, D the particle diameter, ρ the particle density and Γ the interface energy. This dimensionless group, Δ, incorporates the Weber number, ($\rho DV^2/\Gamma$), which was previously shown to be influential in agglomerate breakage, and may be presented in the form, $\Delta = We I_e^{2/3}$, where $I_e = ED/\Gamma$.

The predicted dependency of the damage ratio on the surface energy has been tested using Distinct Element Method (DEM). Four different agglomerates have been formed and impacted against a target for three different values of the surface energy of the primary particles. The simulation results show that the effect of surface energy is better described by the above mechanistic model than by the Weber number alone, as previously used to characterise the impact strength of agglomerates.

Keywords: Agglomeration; Granulation; Impact; Modelling; Simulation; Surface Energy.

1. Introduction

There are many factors that influence agglomerate strength but one of the most important one is the interparticle bond strength. Depending on the method of agglomeration, the bond strength is brought about by the presence of a binder or interparticle cohesion.

There are two widely cited models regarding this topic that have been developed by Rumpf (1962) and Kendall (1988). Rumpf (1962) defined the strength of agglomerates as the force that is required to break all contacts simultaneously on a prescribed failure plane. Kendall (1988) defined agglomerate strength as the resistance of the agglomerate to crack propagation based on the linear elastic fracture mechanics. These two models apply to the cases in which agglomerates show a fragmentation plane. However, agglomerates can also suffer size reduction in the

form of detachment of small debris and not necessarily by fragmentation. Furthermore, the application of the models of Kendall and Rumpf to the case of impact is not straightforward, since agglomerates can develop different patterns of breakage depending on the impact velocity and agglomerate properties (Moreno *et al.* 2004, Moreno *et al.* 2003, Subero, 2001) .

For agglomerates which can store elastic strain energy, Kendal's model is consistent with the Griffith criterion for crack propagation, whilst Rumpf's model may be more applicable to cases involving plastic deformation as the model is based on a simple force balance. Experimental data reported in the literature show consistency with either or both models. There is no detailed analysis of the applicability of the above models in the literature. However, Subero (2001) has shown that in so far as the dependency of the packing fraction is concerned both models predict numerically similar values, albeit from very different functional relationships as given by these models.

The analysis of agglomerate strength by continuum mechanics has not been successful so far due to the degree of freedom arising from the structure of agglomerates. Recently, significant progress has been made by the use of DEM (Kafui and Thornton, 1993). The main advantage of this method is its versatility, allowing an easy variation of any material property without affecting the others. A further advantage of DEM is the possibility of quantifying the effect of impact on the number of interparticle contacts which break within the agglomerate, a feature that cannot be easily diagnosed experimentally. This paper is focussed on the effect of

interparticle cohesion on the impact breakage of agglomerates using the above method.

The first work analysing the effect of bond strength on agglomerate breakage using DEM was carried out by Kafui and Thornton (1993). They analysed the effect of the surface energy on the strength of regularly-packed agglomerates having a face centred cubic and a body centred cubic structures, and related the breakage of interparticle contacts to the Weber number, *We,* as defined by

$$We = \frac{\rho D V^2}{\Gamma} \tag{1}$$

where ρ is the particle density, D the primary particle diameter, V the impact velocity and Γ is the interface energy which is defined by the Dupré equation (Israelachvili, 1985) as:

$$\Gamma = \gamma_A + \gamma_B - \gamma_{AB} \tag{2}$$

where γ_A and γ_B are the surface energies of two particles made of different materials, A and B, in contact with each other and γ_{AB} is the interaction energy between them. For surfaces of the same material γ_{AB} is zero and therefore $\Gamma=2\gamma$.

Kafui and Thornton (1993) carried out simulations of impact breakage of agglomerates with a two-dimensional (2-D) particle motion. They defined the

damage ratio as the ratio of broken contacts to the initial number of bonds and expressed it as a function of the Weber number for a range of impact velocities and surface energies. They found that the curves corresponding to the values of surface energy between 0.1 J/m^2 and 1.0 J/m^2 were reasonably unified by the use of Weber number. However, in a later work, Thornton *et al.* (1996) obtained a better unification by modifying the Weber number and defining a lower limit of impact velocity, V_0, below which no contact is broken, as given by Eq. 3.

$$We' = \frac{\rho D(V - V_0)^2}{\Gamma} \tag{3}$$

The above analysis was also carried out for a 2-D ordered packing with the surface energy in the range between 0.3 and 3.0 J/m^2. Later, Subero *et al.* (1999) carried out simulations using three-dimensional (3-D) motion of the particles and analysed the effect of the surface energy in the range 0.5 to 5.0 J/m^2 in randomly packed agglomerates using Eq. 3. They plotted the damage ratio as a function of We' and found the results in agreement with the work of Thornton *et al.* (1996), *i.e.* a good unification of data for different surface energies was obtained. However, the range of surface energies used in the above simulations covered only one order of magnitude variations. In a later work, Moreno (2003) varied the surface energy by two orders of magnitude and found that the use of the modified Weber number no longer unified the data adequately. Therefore an alternative analysis is proposed here based on the idea that the damage suffered by agglomerates during impact is related to the incident

kinetic energy and also to the physical and mechanical properties of the agglomerates. The basis of the model is described in the following section.

2. Mechanistic Model of Agglomerate Breakage

The model development is based on the consideration that the work required to break interparticle contacts varies linearly with the incident kinetic energy. Associated with each contact breakage is an amount of work to furnish the required surface energy and therefore the higher the incident energy the larger will be the number of broken contacts. The incident kinetic energy, E_K, before impact is given by

$$E_K = N\frac{1}{2}mV^2 \tag{4}$$

where N is the number of particles in the agglomerate, m is the mass of a primary particle and V is the impact velocity of the agglomerate. The case of normal impact is considered here. If the total number of broken contacts after impact is N_B, the work for breaking these contacts, W, assuming that all contacts have the same contact area, A, is:

$$W = N_B \Gamma A \tag{5}$$

Let us consider that the total work spent in breaking N_B bonds is proportional to the incident kinetic energy of the agglomerate with k being the proportionality factor.

$$N_B \Gamma A = kN \frac{1}{2} m V^2 \quad (6)$$

Rewriting Eq. 6 and expressing the particle mass as a function of particle density, ρ, and particle diameter D, N_B,, can be expressed as:

$$N_B = k \frac{1}{12} \pi N \frac{\rho D V^2}{\Gamma} \frac{D^2}{A} \quad (7)$$

The contact area, A, depends on the interface energy and mechanical properties of the contact. Therefore, the dependency of the number of broken contacts on the interface energy is not fully determined by the exponent of –1 of the interface energy in Eq. 7. In order to analyse the full dependency on the surface energy, the model of Johnson *et al.* (1971) is used to describe the relationship between the contact area and surface energy. The model provides an expression for the contact area radius, a, which is similar to the expression provided by Hertz analysis, but substituting the actual external load, P, by an effective load, P_{EFF}, which depends on the absolute value of the pull-off force, P_{OFF}:

$$a^3 = \left(\frac{3R^*}{4E^*} \right) P_{EFF} \quad (8)$$

$$P_{EFF} = P + 2P_{OFF} + (4PP_{OFF} + 4P_{OFF}^2)^{1/2} \quad (9)$$

$$P_{OFF} = \frac{3}{2} \pi \Gamma R^* \quad (10)$$

where, R^* and E^* are the reduced radius and reduced elastic modulus of the two particles in contact as defined by the Eqs. 11 and 12, respectively.

$$\frac{1}{R^*} = \frac{1}{R_1} + \frac{1}{R_2} \tag{11}$$

$$\frac{1}{E^*} = \frac{1-\nu_1^2}{E_1} + \frac{1-\nu_2^2}{E_2} \tag{12}$$

Considering that before impact the external forces acting on the contacts are small as compared to P_{OFF}, the value of the effective force, P_{EFF}, is approximately equal to $4P_{OFF}$ and Eq. 9 can be rewritten as:

$$P_{EFF} = 4P_{OFF} \tag{13}$$

and substituting Eq. 10 into Eq. 13, the following equation is obtained:

$$P_{EFF} = 6\pi\Gamma R^* \tag{14}$$

If the agglomerate is made of monodispersed particles $R^* = R/2$ and the particles are all of the same material $E^* = E/2(1-\nu^2)$, where ν is Poisson's ratio, Eqs 8 and 14 are reduced to:

$$a^3 = \left(\frac{3R(1-\nu^2)}{4E} \right) P_{EFF} \tag{15}$$

$$P_{EFF} = 3\pi\Gamma R \tag{16}$$

Substituting Eq. 16 into Eq. 15, the radius of the contact area is expressed as a function of the primary particle properties and the interface energy from which the contact area, A, is calculated:

$$A = \pi a^2 = \left(\frac{3}{4} \right)^{4/3} \pi^{5/3} (1-\nu^2)^{2/3} \left(\frac{D^2}{E} \Gamma \right)^{2/3} \tag{17}$$

Finally, if the value of the contact area A is introduced in Eq. 7 and the terms are rearranged, the number of broken contacts after impact can be expressed as:

$$N_B = k \frac{4^{1/3}}{3^{7/3} \pi^{2/3}} N \frac{1}{(1-\nu^2)^{2/3}} \frac{\rho D^{5/3} V^2}{\Gamma^{5/3}} E^{2/3} \tag{18}$$

Equation 18 relates the number of broken contacts in the agglomerate with the primary particle properties (particle density, ρ, particle diameter, D, and particle elastic modulus, E), the interparticle interaction, the agglomerate properties (the number of particles in the agglomerate, N) and the characteristics of the test (*i.e.* impact velocity, V).

Now, considering the definition of damage ratio, it is necessary to relate the initial number of bonds in the assembly, N_0, to the number of particles in the agglomerate. This can be carried out using the coordination number of particles in the assembly, Z. The relationship between N, N_0 and Z is given by:

$$N = \frac{2N_0}{Z} \tag{19}$$

Therefore, Eq. 18 can be rewritten in the form,

$$N_B = k\frac{2^{2/3}}{3^{7/3}\pi^{2/3}}\frac{2N_0}{Z}\frac{1}{(1-\nu^2)^{2/3}}\frac{\rho D^{5/3}V^2}{\Gamma^{5/3}}E^{2/3} \tag{20}$$

and damage ratio can be given in the form

$$D_R = k\frac{2^{5/3}}{3^{7/3}\pi^{2/3}}\frac{1}{Z}\frac{1}{(1-\nu^2)^{2/3}}\frac{\rho D^{5/3}V^2}{\Gamma^{5/3}}E^{2/3} \tag{21}$$

Now considering the terms in Eq. 2.1 it is possible to define a new dimensionless number, Δ, as given by Eq. 22, incorporating particle density, particle diameter, elastic modulus and interface energy.

$$\Delta = \frac{\rho D^{5/3}E^{2/3}V^2}{\Gamma^{5/3}} \tag{22}$$

Following Thornton's (2001) suggestion this number can be written as a product of two primary dimensionless groups, the Weber number, *We*, and the elastic adhesion index, I_e, which is defined by Eq. 23:

$$I_e = \left(\frac{ED}{\Gamma} \right) \tag{23}$$

Therefore the damage ratio, D_R, is given by:

$$D_R \alpha We\, I_e^{\,2/3} \tag{24}$$

It will now be of interest to explore if such a relationship prevails in the numerical simulation of agglomerate breakage and this is described below.

3 Simulations of Agglomerate Breakage

3.1 Simulation details

In order to analyse the effect of bond strength on the agglomerate breakage behaviour, four agglomerates of 3000 primary particles were formed in exactly the same way, but with different initial configurations of the primary particle positions. The primary particle properties are given in Table 1 and the size and properties of the four agglomerates are given in Table 2. The coordination number, *Z*, is given within each

bracket next to the value of the contact number. The surface energy was varied within a range of two orders of magnitude.

The simulations were carried out by using DEM whereby the primary particles accelerations, positions and velocities were calculated using Newton's laws of motion and updated cyclically, details of which are described by Moreno (2003). The particles deformed elastically according to the Hertz model for normal contact interactions (see Johnson, 1985) and Mindlin and Deresiewicz (1953) and Thornton and Randall (1988) for tangential contact interactions. When the surface energy was assigned to the particles in contact with each other the models of Johnson *et al.* (1971) and Savkoor and Briggs (1977) were used to determine the normal and tangential contact forces respectively.

The agglomerates were formed by randomly positioning 3000 particles within a spherical space. A centripetal field was applied and the particles were brought together in order to form a dense spherical agglomerate. During this first stage the particles were frictionless and non-cohesive, which allowed an easy agglomeration of the particles. Once the assemblies reached a stable number of interparticle contacts, friction and surface energy were introduced slowly in small steps in order to avoid the accumulation of residual stresses. The four agglomerates A, B, C and D have therefore nearly the same characteristics, but have been generated with different random seed for positioning the primary particles.

For each impact velocity the mean and standard deviation of the number of broken contacts were determined for the four agglomerates. Each agglomerate was impacted in a range of impact velocities from full elastic rebound with no breakage of contacts to the disintegration of the agglomerate into small fragments. The number of broken contacts in the agglomerates was analysed by using the modified Weber number, We' and the new dimensionless group, Δ.

3. 2 Analysis of Damage Ratio

Figure 1 shows the average damage ratio as a function of the impact velocity for the impact of the agglomerates reported in Table 2. At low impact velocities the damage ratio is not sensitive to the impact velocity, but once a threshold velocity is exceeded the damage ratio quickly rises and eventually approaches unity in an asymptotic way. An increase in the impact velocity implies an increase in the incident kinetic energy and consequently more energy is available in the system for breaking contacts. However, at high impact velocities the process of breakage of contacts slows down and the damage ratio becomes again insensitive to the impact velocity. In general, the dependency of the damage ratio with the impact velocity is in agreement with the previous results of Thornton *et al*. (1996) and Subero *et al*. (1999).

For a given impact velocity, the damage ratio decreases as the surface energy is increased (Fig. 1). This is due to linear dependency between the force required to break a contact and the surface energy.

In Fig. 2, the damage ratio has been plotted as a function of the modified Weber Number as given by Eq. 3. There is some normalisation of the data with different surface energies although it is still possible to distinguish one curve from another and furthermore each curve is out of the limits of the error bars of the other curves for intermediate values of the modified Weber number. Thornton *et al.* (1996) and Subero *et al.* (1999) showed a good unification of the data when the modified Weber number was used. The difference with the previous work is thought to be mainly due to the wider range of surface energies tested here. The simulations carried out here cover two orders of magnitude of the surface energy, in contrast to previous work, where the values of surface energy were within approximately one order of magnitude. Furthermore, differences in the agglomeration method could produce slightly different structures and packings.

Figure 3 shows the damage ratio as a function of the new dimensionless group, Δ. As it can be seen a much better unification of data is obtained as compared to Fig. 2. At low values of the dimensionless group, Δ, each curve is now within the error bars of the other curves. However, at high values of the abscissa, the unification is not as good as for the rest of the curves for the case of lower surface energy (0.35 J/m^2) although the curves for larger values of surface energy are reasonably well unified. The lack of good unification of the smallest value of surface energy (0.35 J/m^2) at large value of Δ is unclear and need a further investigation.

4. Discussion and Conclusions

The main assumption made in the model presented in section 2 is the consideration that the work for breaking contacts is proportional to the incident kinetic energy. This leads to Eq. 18, in which the number of broken contacts is related to the primary particles properties (elastic modulus, Poisson's ratio, particle density, particle diameter and bond strength), agglomerate properties (number of particles in the agglomerate and coordination number) and external parameters (impact velocity).

The analysis of the simulation results clearly shows that the dimensionless group, Δ, describes the effect of the surface energy on the agglomerate strength much better than the Weber number, We, and its modified form, We'. Other factors such as the agglomeration process, the level of residual stresses in the agglomerate, the range of the surface energy are undoubtly influencial in the results. Therefore, an accurate comparison with the previous work is not strictly feasible. However, the new dimensionless group is based on an energy balance and should in principle have more predictability.

Another point that needs to be highlighted is related to the minimum velocity under which no breakage of contacts is observed, V_0, and to the modified Weber number, $We^{'}$. In the simulations reported here it has been observed that the subtraction of V_0, from the impact velocity does not significantly improve the fit for the curves of damage ratio versus Δ. Furthermore, the subtraction will only affect the data at low impact velocities where V and V_0 are comparable. However, in this range Δ already describes the behaviour of the system very well. Moreover, the physical meaning of

subtracting V_0 from V is unclear and we have therefore opted for not including such a modification in the dimensionless group, Δ.

In summary, a new mechanistic model of the impact breakage of agglomerates has been presented relating the number of broken interparticle contacts to the interparticle bond strength as represented by surface energy. The model is based on the assumption that the work required for breaking contacts is proportional to the incident kinetic energy. A new dimensionless group incorporating the Weber number and the elastic adhesion index has been obtained. According to this new group the number of broken contacts varies with the surface energy to the power index of -5/3. This is in contrast with the previous work reported in the literature that uses the Weber number as a dimensionless group in the analysis of the effect of bond strength on agglomerate breakage due to impact.

The results of the computer simulations have corroborated the relationship between the surface energy and number of broken contacts as predicted by the new model.

Acknowledgments

The authors gratefully acknowledge ICI plc and the Engineering and Physical Sciences Research Council for the sponsorship of this work. Special thanks are also due to Prof. R. Oliver and Dr D. Sutton for coordination of the project at ICI. We are also grateful to Dr C. Thornton for his helpful comments on the mechanistic model presented here.

Nomenclature

A	Area of a contact	m^2
a	Contact area radius	m
D	Particle diameter	m
D_R	Damage ratio	-
E_K	Incident kinetic energy of an agglomerate	J
E	Elastic modulus	Pa
k	Proportionality factor	-
I_e	Elastic adhesion index	-
m	Particle mass	kg
N	Number of particles in an agglomerate	-
N_B	Number of broken contacts	-
N_0	Initial number of bonds in an agglomerate	-
P	External load in a contact	N
P_{EFF}	Effective force in JKR model	N
P_{OFF}	Force to break a contact	N
R	Particle radius	M
T	Tangential force	N
V	Particle velocity	m/s
V_0	Velocity under which no contacts are broken in agglomerates	m/s
W	Work for breaking contacts	J
We	Weber Number	-
We'	Modified Weber number	-

Z	Coordination number	-

Greek characters

Δ	New dimensionless group	-
Γ	Interface energy	J/m^2
γ	Surface energy	J/m^2
ε	Porosity	-
μ	Friction coefficient	-
ν	Poisson's ratio	-
ρ	Particle density	kg/m^3
Φ	Packing fraction	-

References

Israelachvili, (1985). Intermolecular & Surface Forces. London Academic Press.

Johnson, K. L. (1985). Contact Mechanics. Cambridge University Press, Cambridge.

Johnson, K. L., Kendall, K., and Roberts, A. D.(1971). Surface energy and the contact of elastic solids. Proceedings of the Royal Society of London A., **324**, 301-313.

Kafui, K. D. and Thornton, C. (1993). Computer simulated impact of agglomerate. Powders & Grains 93, the proceedings of the 2nd International Conference on Micromechanics of Granular Media. C. Thornton (Ed.), A. A, Balkema, Rotterdam, 401-406.

Kendall, K. J. (1988). Agglomerate strength. Powder Metallurgy, **31**, 28-31.

Mindlin, R. D. and Deresiewicz, H. (1953). Elastic spheres in contact under varying oblique forces. Journal of Applied Mechanics, **20**, 327-344.

Moreno, R. (2003). Computer simulation of impact of spherical agglomerates using Distinct Element Method. PhD thesis. University of Surrey, Guildford, UK.

Moreno, R. and Ghadiri, M., (2004). Computer simulation analysis of the effect of bond strength on the breakage pattern of agglomerates. PARTEC. Nuremberg, Paper 3.5.

Moreno, R., Ghadiri, M. and Antony, S. J., (2003). Effect of the impact angle on the breakage of agglomerates. Powder Technology, **130**, pp. 132-137.

Savkoor, A. R. and Briggs, G. A. D (1977). The effect of the tangential force on the contact of elastic solids in adhesion. Proceedings of the Royal Society of London, A., **356**, 103-114.

Subero, J. (2001). PhD Dissertation. Impact breakage of agglomerates. Surrey University, Guildford, U.K.

Subero, J., Ning. Z., Ghadiri, M. and Thornton, C. (1999). Effect of interface energy on the impact strength of agglomerates. Powder Technology, **105,** 66-73.

Thornton, C. (2001). "Comments on the mechanistic model". Personal communication.

Thornton C., Yin, K. K. and Adams M. J. (1996). Numerical simulation of the impact fracture and fragmentation of agglomerates. J. Phys. D: Appl. Phys. **29,** 424-435**.**

Thornton, C. and Randall, C. W. (1988). Applications of theoretical contact mechanics to solid particle system simulation. Micromechanics of Granular Materials. Elsevier Science Publishers B. V. Amsterdam, 133-142

Table 1 Single particle properties

Young's modulus (GPa)	31
Poisson's ratio	0.3
Density (kg/m^3)	2000
Friction coefficient	0.35
Particle radius (μm)	50

Table 2 Agglomerate properties. The coordination number, *Z*, is given within each bracket next to the value of the contact number.

Agglomerate	**A**	**B**	**C**	**D**
Aggl. radius (mm)	0.902	0.922	0.921	0.912
Packing fraction	0.555	0.520	0.522	0.537
Contact No. for 35 J/m^2	9151 (6.10)	9061 (6.04)	9102 (6.07)	9093 (6.06)
Contact No. for 3.5 J/m^2	8932 (5.96)	8796 (5.86)	8836 (5.89)	8854(5.90)
Contact No. for 0.35 J/m^2	8718 (5.81)	8513 (5.68)	8560 (5.71)	8621(5.75)

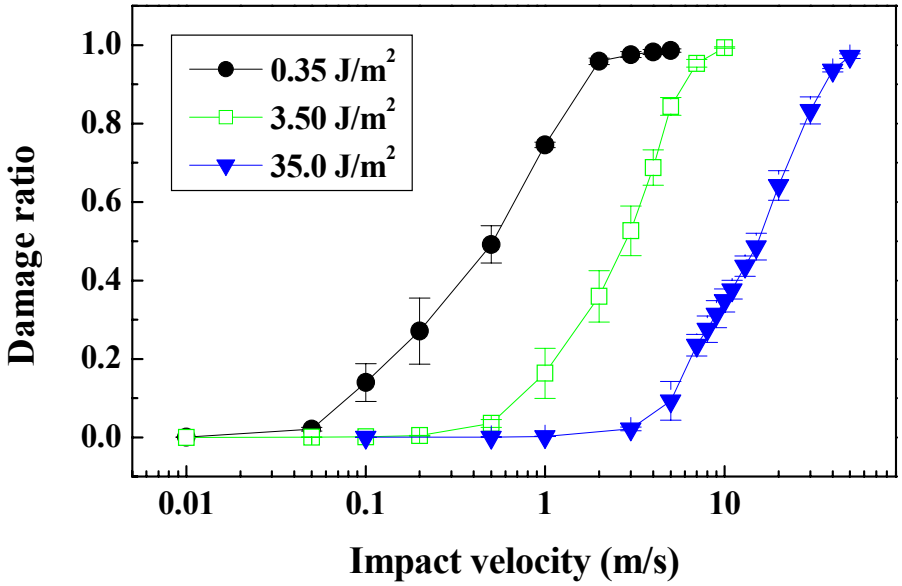

Fig. 1. Relationship between damage ratio and impact velocity for different values of surface energy. The data points correspond to the average values of damage ratio for the impact of Agglomerates A-D reported in Table 2 and the error bars to the standard deviation.

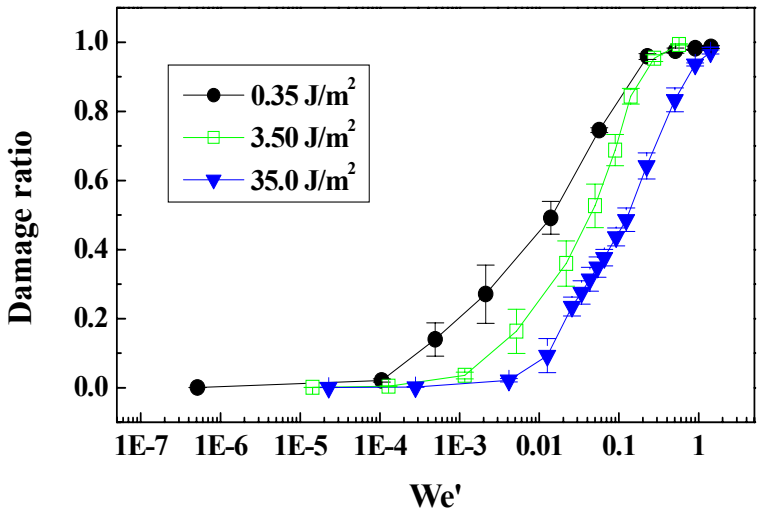

Fig. 2. Relationship between damage ratio and modified Weber Number, *We'*. The data points correspond to the average damage ratio for the impact of Agglomerates A-D reported in Table 2.

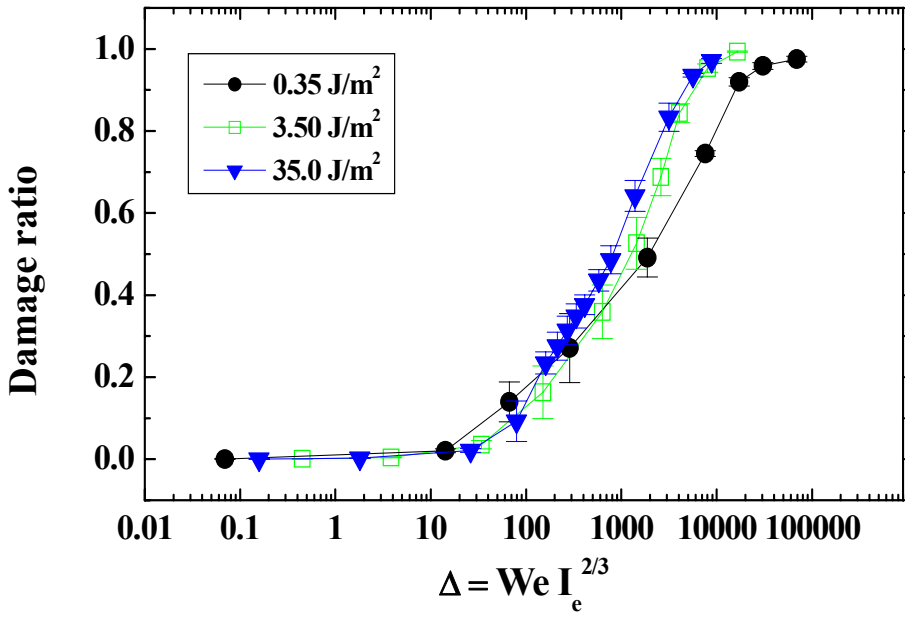

Fig. 3. Relationship between damage ratio and new dimensionless group, Δ, for different values of surface energy. The data points correspond to the average damage ratio for the impact of Agglomerates A-D (Table 2).