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# MODELLING OF AUTO-AGGLOMERATION OF COHESIVE POWDERS

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## ABSTRACT

Fine particles in the micron size range or smaller are usually so cohesive that they cannot exist as individual entities and are in cluster form, the size of which depends on the stress history. During handling, transportation or storage, the powder is subjected to mechanical vibration and/or agitation and, as a result of which clumping of particles or "snowballing" can occur even without the presence of any binder. This is an undesirable feature, as it is responsible for poor flow behaviour, cohesive arching, segregation of lumps and inducing flaws in products. Nevertheless, the mechanism of auto-agglomeration of cohesive powder has not received due attention and the conditions under which such clusters/lumps form, their size, structure and strength has not been analysed extensively. In this work we present a preliminary model to predict the equilibrium cluster size based on two separate energy balances to predict the granule solid fraction and equilibrium size, respectively. Despite some broad approximations, this approach can capture the trend of variation of the agglomerate size with the vibration intensity for some data reported in the literature. The proposed model also identifies the mechanism controlling the growth of the agglomerates as the balance between the cohesive energy of the particles and the disruptive energy of vibration.

## KEYWORDS

Auto-agglomeration, Cohesion, Granulation, Modelling

## 1. INTRODUCTION

Auto-granulation is the agglomeration of fine powder without any additives, due to the highly cohesive nature of the inter-particle contacts. With fine and cohesive powder stable agglomerates can be formed by mechanical vibration or fluidization [1-7]. However, the mechanism by which clusters form has not been elucidated sufficiently. In wet granulation, granule growth occurs when like-sized entities collide with an impact kinetic energy lower than the dissipation across the binder layer [8]. In the absence of a binder, the mechanism by which the collisional energy is dissipated is necessarily related to inter-particle cohesion. Based on this consideration, Ku et al. [2] proposed a snow-balling mechanism for granule growth, where fine particles are taken up by larger granules by sticking onto their surface. According to this interpretation, the agglomerates would comprise identical building blocks, i.e. the individual particles of the powder.

Attempts have been made to predict the equilibrium cluster size in pressure swing agglomeration operations [9-11]. Similar to auto-granulation, pressure swing granulation does not require the presence of a binder. In a typical cycle, the powder is first compacted by a downward flux of air, following which the formed compact is broken into clusters by reversing the direction of the air flow and fluidising the clusters, thus producing round granules. For this application, Iwadate and Horio [9] suggested a model based on a force balance between the cohesion holding together two granules and the disruptive force associated with the passage of bubbles during the fluidisation stage. However, a limitation of this approach is the need for empirical inputs such as the granule crushing strength and agglomerate density as well as for the description of the bubble phase. The model of Mooroka et al. [7] for dry granulation is based on an energy balance between the collisional energy, augmented by

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the energy transferred through laminar shear, and the energy required to break the granules. The applicability of their model is also affected by the presence of parameters difficult to estimate and, as in the previous case, no description of the granule fracture mechanics is included. In this regard, Golchert et al. [12] suggested that the breakage behaviour is strongly dependent on the structure of the granule, as crack propagation depends on the network of contacts between the particles within a granule.

Models to predict the granule size in vibro-fluidised beds have also been proposed by writing both force [3] and energy balances [6]. In both cases the variation of the granule solid fraction with the process conditions was not considered important, with the aggregate density assumed close to the vibrated density of the powder. However, the development of an accurate macroscopic model for dry granulation requires a description of the processes of densification and breakage [13-18]. Ku et al. [1] found that mechanically vibrated fine titania particles cluster to a maximum equilibrium size which increase with the level of vibration, i.e. by increasing the vibration amplitude and/or frequency. The same authors carried out measurements of the granule solid fractions and found a core-rim microstructure with the outer layer of the granules exhibiting higher density compared to the inner core. In contrast to previous studies, the densification of the outer layer with the vibration power was therefore considered to be responsible for the increase in the strength of the particle structure which allowed the granules to grow to a larger equilibrium size.

In view of the above, a satisfactory mathematical model for auto-agglomeration has yet to be developed. We present a preliminary mathematical model of auto-agglomeration based on the description of granule densification and breakage.

## 2. MODEL

A model is derived below to estimate the equilibrium granule size when fine cohesive powders are subjected to mechanical vibration. It is assumed that an equilibrium granule size is reached as a result of the balance between the mechanisms of densification and breakage brought about by the bed agitation.

### Granule densification

At the start of the process, the agglomeration and subsequent increase in packing density is assumed to occur rapidly, with all the impacts between primary particles being effective in creating new contacts. The kinetic energy of the particles is therefore totally converted into bonding energy inside the agglomerate structure:

$$\frac{1}{2} N_p m_p V^2 = N_b \Gamma A_c \quad Eq (1)$$

where  $N_p$ ,  $m_p$  and  $V$  are the number, mass and velocity of the primary particles,  $N_b$  is the number of bonds formed,  $\Gamma$  the surface energy and  $A_c$  the contact area. The number of bonds and particles in the agglomerate can be related to the coordination number  $Z$  [13]:

$$Z = \frac{N_b}{2N_p} \quad Eq (2)$$

The coordination number can also be related to the agglomerate solid fraction by the empirical equation [19]:

$$Z = \frac{\alpha}{(1-\phi)^n} \quad \text{Eq (3)}$$

with  $\alpha=1.61$  and  $n=1.48$ . Substitution of Eqs 2 and 3 into Eq. 1 yields:

$$\phi = 1 - \left( \frac{4\alpha A_c \Gamma}{m_p} \right)^{1/n} \frac{1}{V^{2/n}} \quad \text{Eq (4)}$$

which provides the agglomerate solid fraction as a function of the primary particles properties and velocity. Eq.4 can become negative below a certain velocity threshold, which could be interpreted as the minimum operating velocity for agglomeration to occur. The velocity of the particles is assumed to be related to the vibration amplitude A and frequency f as:

$$V = 2\pi A f \quad \text{Eq (5)}$$

The contact area is estimated from the JKR model [13] as:

$$A_c = \left( \frac{3}{4} \right)^{4/3} \pi^{5/3} (1-\nu^2)^{2/3} \left( \frac{d_p^2 \Gamma}{E} \right)^{2/3} \quad \text{Eq (6)}$$

with  $d_p$  denoting the primary particle size,  $\nu$  the Poisson ratio and E the Young modulus.

### Equilibrium granule size

It is now assumed that the size of the granules is limited by the onset of breakage. The granule tensile strength is given by Kendall [14] as:

$$\sigma = 15.6 \phi^4 \Gamma_c^{5/6} \Gamma^{1/6} (d_p c)^{-1/2} \quad \text{Eq (7)}$$

where  $\Gamma_c$  is the fracture energy and c the crack length. It is now assumed that  $\Gamma_c \sim \Gamma$  and that the crack length c varies proportionally with the primary particle size  $d_p$ . The work required to break the granule is also roughly estimated as:

$$W = \sigma \frac{\pi d_a^2}{4} \lambda \quad \text{Eq (8)}$$

where  $\lambda$  is a characteristic length assumed to be again proportional to the primary particle size  $d_p$ . Substituting Eq.7 into Eq.8 therefore yields:

$$W \propto \phi^4 \Gamma d_a^2 \quad \text{Eq (9)}$$

The work to break the granule given by Eq.9 is also considered to be proportional to the incident kinetic energy of the agglomerates, following the work of Moreno-Atanasio and Ghadiri [13]:

$$W \propto \frac{1}{2} \rho_p \phi \frac{\pi d_a^3}{6} V^2 \quad \text{Eq (10)}$$

Finally, the equilibrium granule size is obtained by equating Eqs 9 and 10, obtaining:

$$d_a = k \times \phi^3 \frac{\Gamma}{\rho_p V^2} \quad Eq (11)$$

where k is a proportionality constant. Eq. 11 thus provides an expression to estimate the equilibrium granule size from the primary particle properties and the agglomerate solid fraction.

A non-dimensionalisation of the previous equations will now be carried out to identify the main dimensionless numbers which govern the process. Eq.11 can be written in dimensionless form as:

$$We_a = \frac{\rho_p V^2 d_a}{\Gamma} = k \times \phi^3 \quad Eq (12)$$

where  $We_a$  is the agglomerate Weber number. Inserting Eq. 4 for the agglomerate solid fraction and rearranging gives:

$$We_a = k \times \left[ 1 - \left( \frac{56.5}{Ie_p^{2/3} We_p} \right)^{1/n} \right]^3 \quad Eq (13)$$

where the adhesive index number  $Ie_p$  and primary particle Weber number  $We_p$  are defined as:

$$Ie_p = \frac{Ed_p}{(1-\nu^2)\Gamma} \quad Eq (14)$$

$$We_p = \frac{\rho_p V^2 d_p}{\Gamma} \quad Eq (15)$$

Apart from the uncertainty on the value of k, Eq.13 can be the starting point for a generalised relationship for the equilibrium agglomerate size as a function of the primary particle properties.

### 3. VALIDATION

The model is first validated with the experimental data provided by Ku et al. [1]. The primary particle properties are given in Table 1.

Table 1. Titania particle properties

Property	Value
Density $\rho_p$ , kg/m <sup>3</sup>	3953
Particle size $d_p$ , $\mu$ m	0.1
Young's Modulus, GPa	288
Poisson's Ratio -	0.27

An approximate average value of the surface energy of  $0.3 \text{ J/m}^2$  has been used, which is compatible with values provided by molecular dynamics simulations [20]. The primary particle size is reported to be around  $0.1 \text{ }\mu\text{m}$ , although it is likely that the powder will be endowed with a certain degree of agglomeration even before starting the vibration process. In Figure 1, the solid fractions calculated with  $d_p=2.1 \text{ }\mu\text{m}$  from Eq.4 are reported as a function of  $V$ . In the same figure, the average values of the experimental solid fractions obtained from Ku et al. [1] are shown for comparison. Inspection of Figure 1 reveals that the proposed model is capable of capturing the variation of the packing fraction with the vibration intensity apart from one data point. The underlying physical justification for assuming a much larger value, compared to the real primary particle size, is based on the fact that cohesive sub-micron particles cannot exist as individual entities and the powder will be in a granular form before the vibration process starts. This initial granule size could be somewhat controlled by coarse sieving the particles and breaking the agglomerates larger than a given size (e.g. above few microns). It should be also observed that the value of  $d_p$  mainly affects the calculation of the solid fraction. Experimental values of solid fractions, such as those presented in Figure 1, could be directly substituted into Eq. 11.

In Figure 2, the predictions of Eq. 11 using  $k=2.43$  along with the values of solid fraction given by Eq.4, are compared with the experimental granule size given by Ku et al. [1]. After selecting a suitable value for  $k$ , the approach proposed proves successful in capturing the variation of the granule size with the intensity of vibration. Although the value of  $k$  could appear low, this could stem from some imprecision in calculating the work to break the agglomerate by Eq. 8. Furthermore, the model is sensitive to the value of surface energy, which is difficult to estimate with accuracy. The determination of the surface energy by inverse gas chromatography or, indirectly, by mechanical tests at single particle or bulk solid level would probably provide a good estimate of this property. However, as the model relies on many simplifying assumptions, an accurate determination of the surface energy is probably not necessary at this stage. Using a different value of surface energy would determine a variation of the value of the fitting parameter  $k$ . The main objective of this work is to obtain more insight into the mechanism of auto agglomeration, before attempting at the development of a more rigorous and fully predictive model at a later stage.

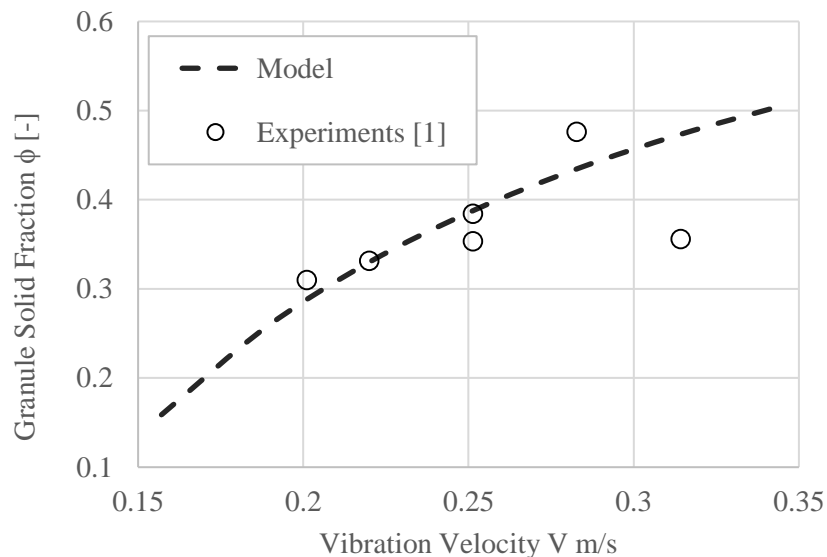


Figure 1. Comparison between predictions from Eq. 4 and experimental values of solid fractions [1] measured as a function of vibration velocity.

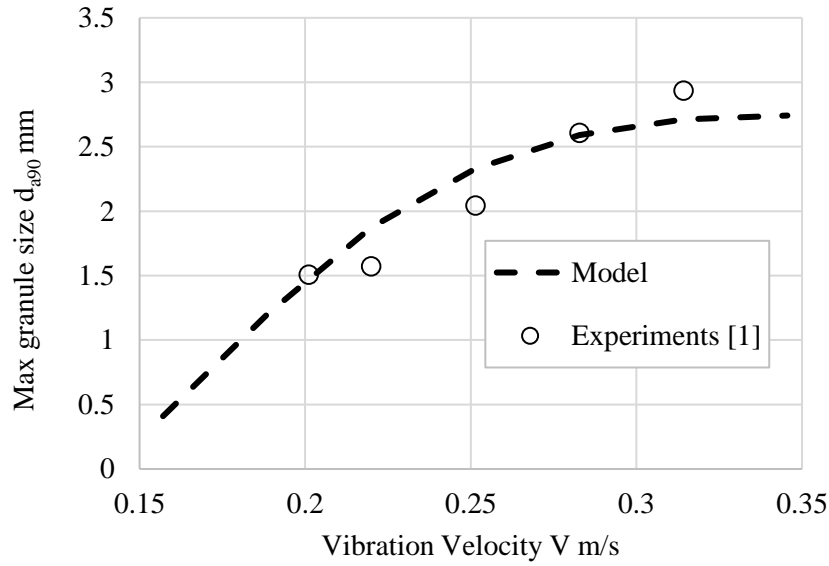


Figure 2. Comparison between predictions from Eq. 11 and experimental values of equilibrium granule size [1] measured as a function of vibration velocity.

We shall now explore using the proposed model for the prediction of the agglomerate size for the system studied by Barletta and Poletto [3]. In their study, agglomeration of silica particles was obtained in vibro-fluidised beds; however, the authors reported that aggregates formed at a gas velocity much lower than that necessary for their fluidisation and, accordingly, fluid dynamic forces can be neglected. Our approach should therefore be applicable. The properties of the silica particles are shown in Table 2. The experimental granule size at the different acceleration due to vibration are reported in Figure 3. The acceleration is calculated from the vibration frequency and amplitude by the expression:

$$a = (2\pi f)^2 A \quad \text{Eq (16)}$$

Table 2. Silica particle properties

Property	Value
Density $\rho_p$ , kg/m <sup>3</sup>	2650
Particle size $d_p$ , $\mu\text{m}$	2.8
Young's Modulus, GPa	70
Poisson's Ratio -	0.19

For our calculations, an average value of surface energy of  $25 \text{ mJ/m}^2$  will be used, although there is some scatter in the values reported in the literature [21]. As these particles are less cohesive than titania, a somewhat smaller value of the primary particle size is used, namely  $1 \mu\text{m}$ .

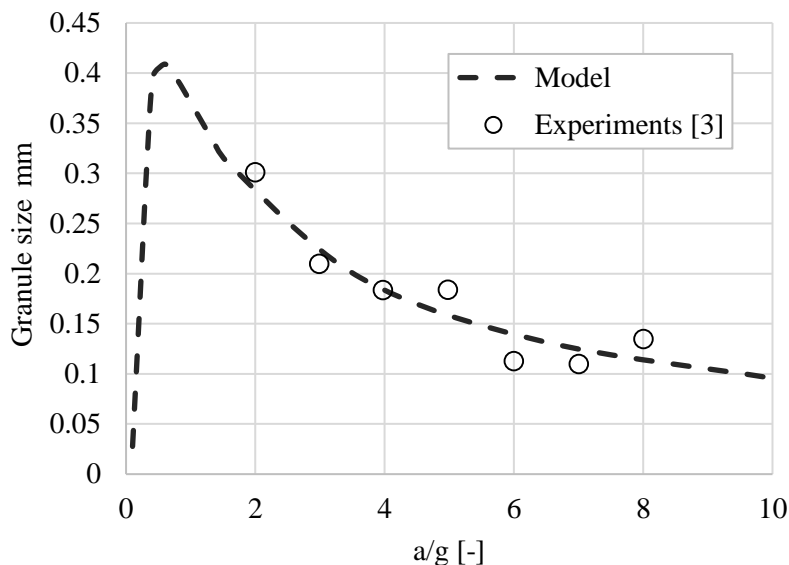


Figure 3. Comparison between predictions from Eq. 11 and experimental values of equilibrium granule size [3] measured as a function of acceleration relative to gravity.

The model predictions calculated with  $k=0.047$  are reported in Figure 3. A much smaller value of  $k$  is therefore needed to fit the data compared to titania particles. However, the value of  $k$  is highly sensitive to the primary particles properties which should be determined accurately. The larger variation of the parameter is probably due to the large number of simplifying assumptions and the inaccuracy of the physical properties introduced into the model. A better comparison between the values of  $k$  obtained can be obtained only by carrying out systematic experiments on a sufficiently large number of powders. These experiments need to determine with reasonable accuracy agglomerate solid fractions, agglomerate crushing strength, initial granule size) and single particle mechanical properties (e.g. Young modulus and surface energy). Once a large number of data is produced, a general correlation in terms of dimensionless numbers, such as the one expressed by Eq. 13, can be tested. Nevertheless, inspection of Figure 3 reveals an important feature of the model, i.e. that under certain conditions the equilibrium granule size can be maximum at an intermediate level of agitation. This stems from the fact that once the granule has already reached a high level of densification, any increase in the vibration intensity will only lead to an intensification of breakage. The equilibrium granule size is therefore the result of a balance between the cohesive tendency of the powder and the inherent granule strength. Once the granules have reached their maximum level of densification, an increase in vibration intensity will result in a larger number of collisions with sufficient energy to produce breakage. This suggests that a critical vibration intensity exists beyond which the equilibrium agglomerate size decreases as more agitation energy is transferred to the solids.

The model developed suggests that the maximum achievable granule size is limited by the onset of breakage at high impact velocity. It can be argued that the same modelling approach can be applied to fluidised beds, provided that the granule impact velocity can be estimated. In this regard, Ennis et al. (1991) [8] have provided the following relationship which depends on the properties of the bubble phase:



$$V_{\max} = 12 \frac{u_B d_a}{d_B} \quad \text{Eq (17)}$$

where  $u_B$  is the bubble velocity and  $d_B$  is the bubble size. For group A solids the bubble size can be assumed to be constant, whereas the bubble velocity varies almost linearly with the excess gas velocity  $u - u_{mf}$  (assuming that the minimum fluidisation velocity  $u_{mf}$  is not very different from the minimum bubbling velocity). In the light of these assumptions, substitution of Eq.17 for  $V$  in Eq. 11 gives:

$$d_a = k' d_B^{2/3} \times \phi \left( \frac{\Gamma}{\rho_p (u - u_{mf})^2} \right)^{1/3} \quad \text{Eq (18)}$$

where  $k'$  is a proportionality constant which includes the effect of the bubble phase properties.

In Figure 4, some experimental granule sizes obtained in a fluidised bed of fused alumina [22] are shown as given in [9]. The solid fraction is reported equal to 0.35 and assumed constant in the following analysis. With a surface energy of  $0.01 \text{ J/m}^2$  the experiments are compared with the predictions of Eq.18 in Figure 4, using the data given in Table 3. With a 10 mm bubble size, the value of the fitting parameter  $k'$  is 0.86. This reveals that the same approach could also be capable of capturing the trend of the agglomerate size with the excess gas velocity.

Table 3. Fused alumina particle properties

Property	Value
Density $\rho_p$ , kg/m <sup>3</sup>	3970
Young's Modulus, GPa	211
Poisson's Ratio -	0.27

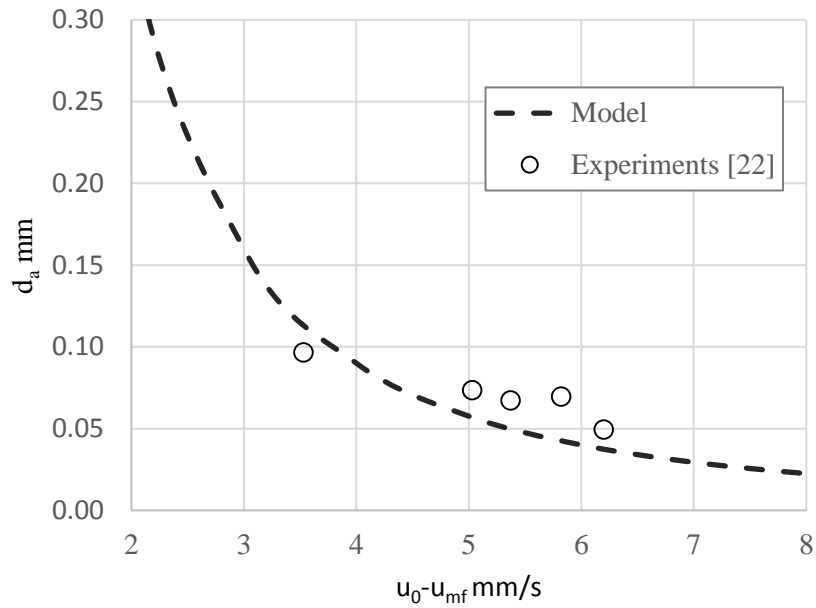


Figure 4. Comparison between predictions from Eq. 18 and experimental values of equilibrium granule size [22] measured as a function of the excess gas velocity.

#### 4. CONCLUSIONS

A simple model has been proposed which seems capable of reproducing the experimental trend of the agglomerate size and solid fraction with vibration intensity. Despite broad approximations, the approach developed has shown that an equilibrium granule size is reached as a result of the balance between densification and breakage. The analysis also reveals that a maximum agglomerate size can be obtained at an intermediate vibration intensity, to be confirmed experimentally. The model still needs improvement, especially with respect to providing more insight into the meaning of the parameter which, in this paper, has been related to the crack length. Validation with further experiments after an accurate determination of the physical properties will be undertaken in future work, as well as extension to other applications such as pressure swing granulation.

#### LIST OF SYMBOLS

$a$	acceleration due to vibration	[m/s <sup>2</sup> ]
$A$	vibration amplitude	[m]
$A_c$	adhesive contact area	[m <sup>2</sup> ]
$c$	crack length	[m]
$d_a$	agglomerate size	[m]
$d_{a\ 90}$	agglomerate size at 90 % of the cumulative distribution	[m]

$d_B$	bubble size	[m]
$d_p$	primary particle size	[m]
$d_{p10}$	primary particle size at 10 % of the cumulative distribution	[m]
$E$	Young modulus	[Pa]
$f$	vibration frequency	[Hz]
$g$	gravity acceleration	[m/s <sup>2</sup> ]
$I_{ep}$	primary particle adhesive index number	[-]
$m_p$	primary particle mass	[kg]
$n$	parameter in Eq. 3	[-]
$N_b$	number of bonds between primary particles	[-]
$N_p$	number of primary particles	[-]
$k$	proportionality factor between crack length and agglomerate size	[-]
$u$	gas velocity	[m/s]
$u_B$	bubble velocity	[m/s]
$u_{mf}$	minimum fluidisation velocity	[m/s]
$V$	vibrational velocity	[m/s]
$We_a$	agglomerate Weber number	[-]
$We_p$	primary particle Weber number	[-]
$Z$	coordination number	[-]
$\alpha$	parameter in Eq. 3	[-]
$\Gamma$	surface energy	[J/m <sup>2</sup> ]
$\Gamma_c$	fracture energy	[J/m <sup>2</sup> ]
$\nu$	Poisson ratio	[-]
$\rho_p$	particle density	[kg/m <sup>3</sup> ]
$\sigma$	tensile strength	[Pa]
$\phi$	granule solid fraction	[-]

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