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## Monitoring industrial crystallization using Acoustic emission

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### Abstract

Acoustic emission (AE), which has been successfully applied for monitoring a rather wide variety of solids elaboration processes, was almost never evaluated in the field of industrial crystallization for pharmaceutical or chemical products. Few papers reported that crystallization processes give rise to acoustic emission signals that could be related to the development of the basic crystallization phenomena. This study is intended to demonstrate new perspectives opened up by the possible use of acoustic emission (AE) as a non-intrusive and non destructive sensor for monitoring crystallization with a particular focus being put on real industrial processes. The preliminary results of the study will be reported in this work namely concerning the ability of the EA to follow the crystallization processes under different operating conditions and different types of crystallisation, and the obtained crystals in terms of particle size distribution and agglomeration degree.

**Keywords:** particle size, crystallisation, monitoring, industrial processes

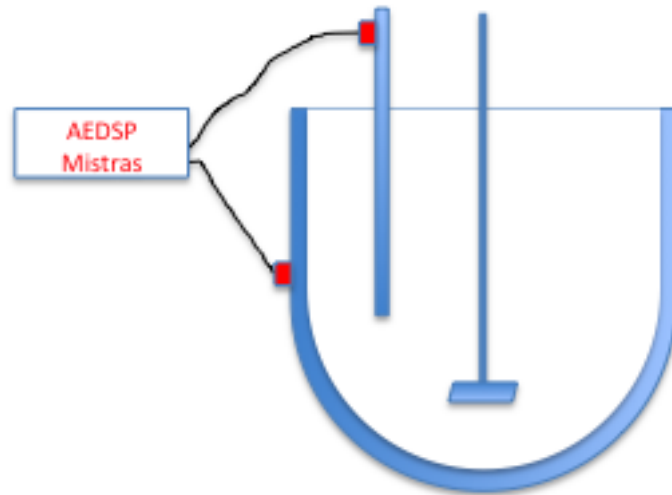
## 1. Introduction

We show that acoustic emission (AE) is a very attractive non-intrusive technique for monitoring crystallization processes. AE has been successfully applied in many fields of material sciences [1] and it was also used in the pharmaceutical industry for monitoring various chemical engineering processes [2] including fluidized bed granulation [3], fluidized bed coating [4], powder compaction [5-6], etc. However, few studies deal with the use of AE to monitoring crystallization processes [7-10]. The objective of this work is to evaluate the potential for using acoustic emission to monitor batch crystallization. The basic concept behind AE monitoring of crystallization processes is that the phase transitions occurring during crystallization in solution induce physicochemical changes in the suspension. Such changes release energy and therefore generate acoustic elastic waves propagating in the liquid medium [11]. Furthermore, as crystal particles are generated, the elastic properties of the dispersed phase also change. The latter changes affect the acoustic emission caused by the particle collision impacts and inter-particles and/or particles-wall frictions. In this present work, -we will only consider particles impacts and the influence of several parameters on it.

## 2. Materials and Method

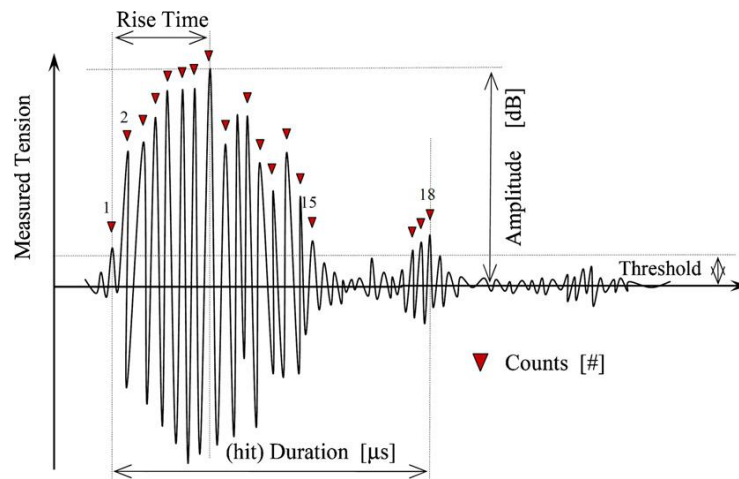
### *2.1 Acoustic emission on crystallisation vessel*

As displayed in Fig.1, a 3 L glass vessel equipped with a jacket was used for the experiments. The jacket is baffled and a centrifugal pump forces the circulation. Stainless –steel baffles and a high efficiency propeller (Mixel TT TM) are used to maintain a good homogeneity of particles in the slurry. The temperature is set at 20°C and the rotating speed of the impeller is 300 rpm.



**Figure 1 :** Experimental apparatus

During this work, we had recourse to system MISTRAS (Massively Instrumented Sensor Technology for Received Acoustic Signal) developed by Euro Physical Acoustics (EPA) for the acquisition and the treatment of the signals of EA. The system uses the chart of digitalization AEDSP 32/16 which allows digital acquisition high speed and high-resolution of the forms of wave and gives access to the parameters (Fig.2) of acoustic emission such as the number of counts, duration, rise time and amplitude (dB).



**Figure 2 :** Main characteristics of acoustic emission

The sampling rate is of 4 MHz. Two sensors: a WD and a F15 provided by Mistras whose Eigen frequency are respectively of 450 KHz and 150 KHz are used in this work. They are both connected to a preamplifier of 40 dB and of a filter [20, 1200] KHz. The PDT (peak time definition), HDT (hit time definition), HLT (hit lockout time) are of 100 100 and 200  $\mu$ s, respectively.

The energy of reference (600 atto Joules) and a threshold of acquisition (30 dB) were given starting from measurements taking of account the electromagnetic environment, the agitation and the flow of coolant on the level of dual envelope.

## 2.2 Powders

Two powders we used in this study, adipic acid and also citric acid. Sigma Aldrich provided both powders and the purity is above 99%. In the table 1, the list of the physical properties is given:

|             | Density                | Solubility              | D50    |
|-------------|------------------------|-------------------------|--------|
| Adipic acid | 1,36 g.cm <sup>3</sup> | 8 g .L <sup>-1</sup>    | 250 μm |
| Citric acid | 1,66 g.cm <sup>3</sup> | 2000 g .L <sup>-1</sup> | 500 μm |

**table 1: physic-chemical properties of powders**

The solubility is given by the work done by Apelblat et al [12]. The density is obtained by helium pycnometer (accupyc from micrometrics) and the D50 by laser granulometry using the sirocco device of the Mastersizer 2000 (Malvern).

## 2.3 Experimental procedure

We only want to study the influence of several parameters of crystallisation on acoustic emission, that's why the solutions must be at a saturation state. The first step of experiments is the dissolution of the two products in order to achieve the saturation stage. Sufficient amount of powder is dropped into the water in order to dissolve all particles. We consider that this stage is achieved when no particles are observed in the vessel. This stage is at least three hour for citric acid and two hours for adipic acid. The next step of experiment is to drop a certain amount of powder inside the solution and then change the volume fraction, the rotating speed or the particle size and records the associated acoustic emission.

## 3. Results and discussion

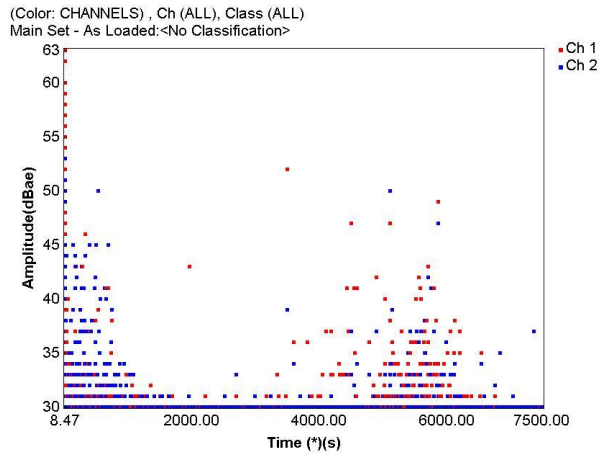
This work on acoustic emission on monitoring industrial crystallization will only consider the impacts of particles and the sensibility of the AE to several parameters such as volume fraction, rotating speed and also particle size. Previous work [13] shows that crystallization processes like nucleation and growth is also responsible for AE, but this will not be included in this work.

### 3.1 Acoustic emission during dissolution

First we discuss on some acoustic event that occur during the dissolution stage of citric acid and then of adipic acid.

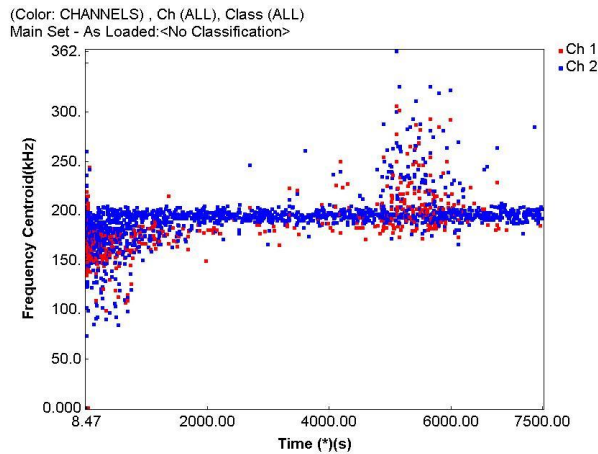
#### 3.1.1. Citric acid

The first experiment on citric acid is recorded after dissolving a large amount of powder (2kg) in 1L of pure water. We show the evolution of AE amplitude during three hours after the introduction of 500 g of citric acid in order to reach a supersaturated state. This evolution is plotted in figure 3. Two periods (from 0 to 1000s and from 5000s to 6000s) of acoustic activity is observed. The first one is the dissolution and the particles impacts of the powders. The other one is not obvious. Normally nothing changes during this period of dissolution.



**Figure 3 : Amplitude of AE (dB) versus time(s).**

In order to have an idea of what may happen during this period, let's have a look on the evolution of centroid frequency versus time.



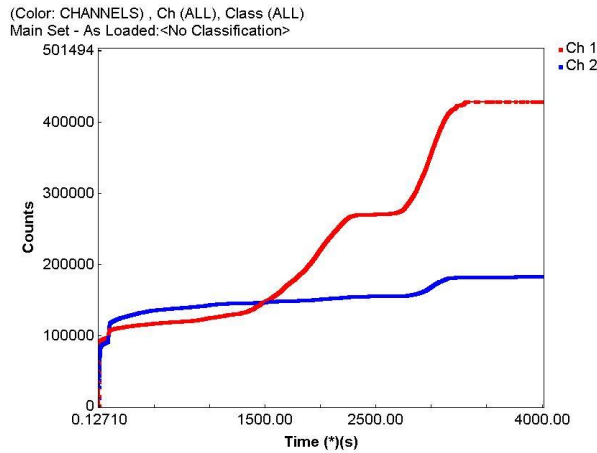
**Figure 4 : Centroid frequency (kHz) versus time (s)**

The centroid frequency of dissolution phenomena is generally under 200 kHz but for the second part of AE activity the frequency is above 200 kHz. In oxalate ammonium crystallisation in Gherras work [13] is generally near 250 kHz for nucleation and/or growth of particles. We can consider that, taking into account the fact that we are above the supersaturation stage, we have a crystallisation of citric acid during this period.

For this reason and also for the strong viscosity of the solution due to the high concentration of citric acid, we change to adipic acid system for the following results.

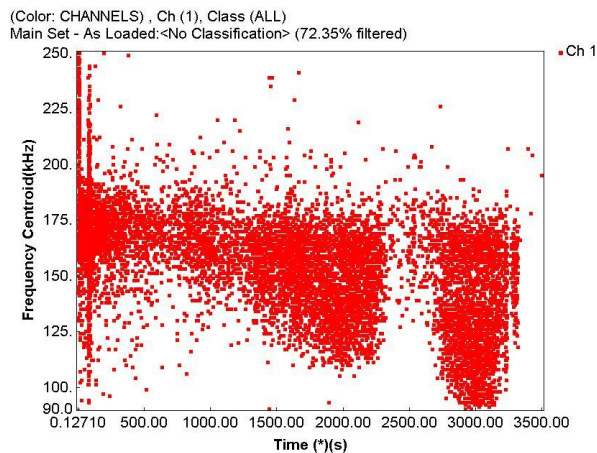
### 3.1.2. Adipic acid

The first experiment on adipic acid is recorded after dissolving 16g of powder in 2L of pure water. We show the evolution of AE amplitude during three hours after the introduction of 8 g of adipic acid in order to reach a supersaturated state. This evolution is plotted in figure 5.



**Figure 5: Cumulative counts versus time (s)**

In this case, we observe in the vessel that the powder is on the surface of the water; the wettability of the adipic acid is not good. That's why a large number of AE counts are recorded by 2000s and 3000s when the powder sink into the water and the dissolution occur. The dissolution and particles impacts are responsible of this AE. In order to have a better idea of this AE, the evolution of centroid frequency versus time is plotted in figure 6.



**Figure 6: Frequency centroid (khz) versus time (s)**

At the beginning the AE frequency is centred around 175 kHz, and then the frequency dropped and is between 100 and 175 kHz. This may be the related frequency of dissolution of adipic acid that is different to the frequency of particle impact. So we can characterize both AE. In the following results we change the experimental procedure by adding surfactant to overcome the poor wettability of adipic acid.

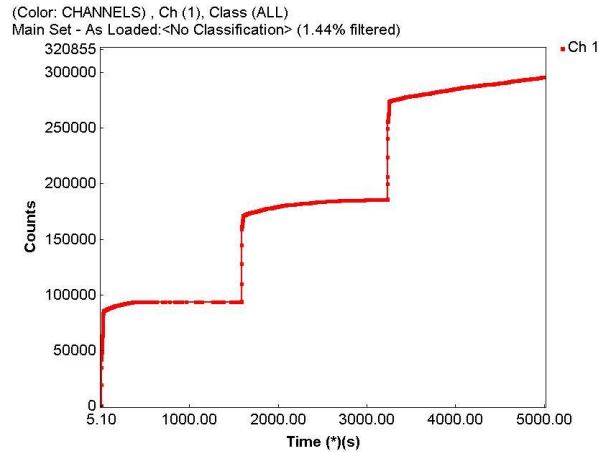
### ***3.2 Influence of volume fraction***

We want now know the sensibility of AE to the volume fraction  $\phi$  of particles inside the vessel. After dissolving 16g of adipic acid with surfactants in 2L of pure water and waiting for total dissolution, we introduce different quantity of powder by adding it with a funnel. After several tests, we decide to introduce from 0,001 up to 0,02 of volume fraction. In table 2, we have the experimental procedure of the different stages of volume fraction, introduced mass of adipic acid versus time.

| Time (s)   | Volume fraction | Total introduced mass (g) |
|------------|-----------------|---------------------------|
| 0-1800s    | 0,001           | 2,72                      |
| 1800-3600s | 0,005           | 13                        |
| 3600-5000s | 0,01            | 27,5                      |

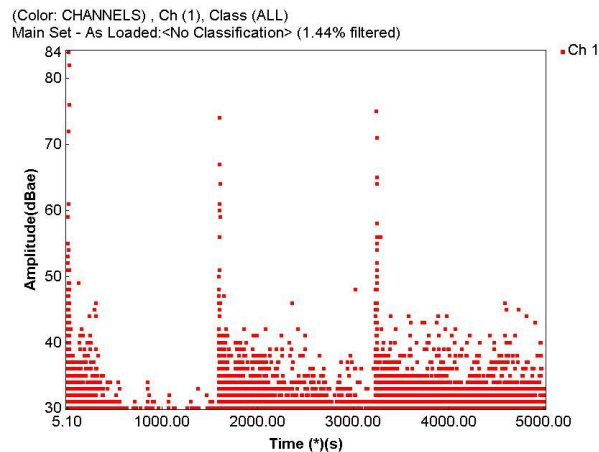
**table 2: experimental procedure**

In figure 7, cumulative counts versus time of this experiment are plotted.



**Figure 7: Cumulative counts versus time (s)**

First, we notice that around 100000 counts are recorded when we introduce the particle inside the vessel. This is the AE of the sliding particles in the funnel. We only want the AE between those two very emissive events. That's why we will analyse the AE amplitude versus time of figure 8.



**Figure 8: Amplitude of AE (dB) versus time(s).**

We have also the AE of the introduction of particles with amplitude up to 80dB. For the first volume fraction, AE is recorded in the beginning then weak AE is detected (may be the background noise).

### ***3.3 Influence of rotating speed***

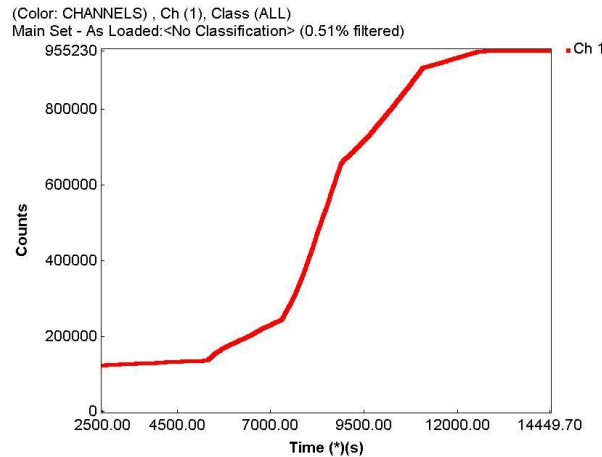
We want now know the sensibility of AE to the rotating speed of the impeller. After dissolving 16g of adipic acid with surfactants in 2L of pure water and waiting for total

dissolution, we change rotating speed from 300 rpm to 500 rpm. In table 3, we have the experimental procedure of the different steps of rotating speed versus time.

| Time (s)      | Rotating speed |
|---------------|----------------|
| 2500-5200 s   | 300 rpm        |
| 5500-7200 s   | 400 rpm        |
| 7200-9000 s   | 500 rpm        |
| 9000-10800 s  | 400 rpm        |
| 10800-12600 s | 300 rpm        |

**table 3 : experimental procedure with evolving rotating speed**

In figure 9, cumulative counts versus time of this experiment are plotted.



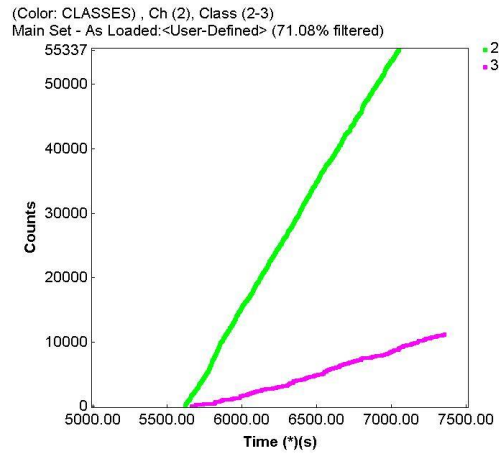
**Figure 9: Cumulative counts versus time (s)**

We easily notice that the number of counts increases with increasing rotating speed. With higher rotating speed, the velocity inside the vessel is higher and the particle impacts are more numerous and more energetics. The stability of AE is quite good because we can draught straight line of number of counts versus time. The slope of the line may be in relation with the number of particles and the size of particles. This will be discussed in the last part of this work.

### ***3.3 Influence of particle size***

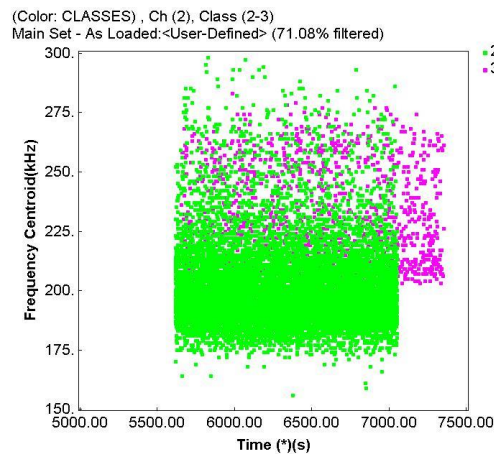
In this part, we sieve the adipic acid powder into two different granulometrics classes, big particles (250-500  $\mu\text{m}$ ) and small particles (50-125 $\mu\text{m}$ ). After dissolving 16g of adipic acid with surfactants in 2L of pure water and waiting for total dissolution, we change rotating speed from 300 rpm to 500 rpm with the sieved particles .In figure 10 cumulative counts versus time is shown of the experimental part of 500 rpm in order to have the better differences between the AE of particles. The big particles are represented by the cluster in green dots 2 and the small one by the purples ones 3. We have more counts for big particles and we have also a linear slope for AE counts versus time like in figure 9.





**Figure 10: Cumulative counts versus time (s)**

The AE is related to the particle size, and we try to assign one AE characteristic by looking at the variation of centroid frequency versus time plotted in figure 11. In this figure, the big particles are represented by the cluster in green dots 2 and the small one by the purple ones 3.



**Figure 11: Frequency centroid (kHz) versus time (s)**

We notice that the centroid frequency is lower for big particle and the range is broader (175 to 300 kHz) than those for smaller particles. So we can use AE to estimate particle size distribution by using may be several sensors and filters in order to have specific response to precise particle size. In this case, we can monitor crystallisation in order to stop the process when a certain size is reached.

## 4. Conclusions

In this study of monitoring industrial crystallization using acoustic emission, several interesting results are obtained. We can notice that we can distinguish different phenomena with different centroid frequencies. The crystallisation can be characterized with frequencies above 200 kHz (figure 4), the dissolution with frequencies down to 100 kHz and for all the particles impacts (figure6). We can detect particles (with this sensor and this experimental procedure) when the volume fraction inside the vessel is above 0,05. Further work must be done in this field to try to detect particle with lower volume fraction. One way can be by increasing rotating speed because the AE is far higher with a speed of 50 rpm for example. To conclude we can distinguish the particle size with AE because the AE signature is changing with particle size.

This work must be proceed by further investigation with other products, other sensors and also vessel to better understand all the AE sources and the AE signature.

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