



## Determination of Impinging Jet Correlation and Dissolution of Materials

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The rate constant for the dissolution in water of a range of materials may be important for designing process equipment or determining the mechanisms of chemical reactions. A jet-impingement apparatus has been used to study dissolution rates by directing a jet of water onto a pellet surface of the material of interest at high velocity to ensure that dissolution is controlling at the given conditions. The apparatus has been used to measure the dissolution rate constant of magnetite under the conditions of power system coolants – often a controlling parameter in steel corrosion. For unequivocal measurements of the dissolution rate constant, the mass transfer characteristics of the apparatus need to be known in order to extrapolate mass transfer coefficients to the conditions of interest. Experiments have been performed using pellets of cast plaster of Paris of different purities and single crystals of the same material (gypsum –  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ), as well as pellets of trans-cinnamic acid, potassium bitartrate and aspartic acid. The dissolution rate constants for single crystal gypsum and aspartic acid were determined and found to be in good agreement with literature values and the commercial plaster result led to a mass-transfer correlation for the jet-impinging apparatus in reasonable agreement with a published correlation. However, the commercial plaster tended to have a higher solubility than pure plaster or gypsum crystals; its dissolution rates were higher than those of the other materials studied, which were in the same range.

### 1. Introduction

There are many industrial processes that rely on measurements of dissolution. Examples are nuclear fuel reprocessing and ore leaching, also, the dissolution behaviour of gypsum is required for studies of geological processes and for determining contamination in oil and gas production processes. Similarly, the dissolution of aspartic acid is studied in the pharmaceutical industry, because the rate of dissolution affects the bioavailability of drug crystals while for trans-cinnamic acid and potassium bitartrate, the study of dissolution rate constants is useful for industrial food production.

Such processes are often controlled by mechanisms that involve dissolution in series with mass transfer. Designing the process equipment for optimum performance entails knowing the dissolution rate separately from the mass transfer rate. Dissolution rates are also important in other situations; for example, the corrosion of metals in liquids may depend on the dissolution of the protective oxide film. A submerged, impinging jet may be used for determining dissolution rate constants if the mass transfer coefficient of the jet apparatus has a high enough mass transfer coefficient to ensure that dissolution is controlling.

The rate constant for the dissolution in water of magnetite, the predominant oxide on carbon steel in cooling water systems, is an important parameter in the mechanism of steel corrosion. Magnetite dissolution kinetics are being studied in a jet-impingement apparatus, where a high-velocity jet of water at the chemistry and temperature of interest is directed onto a compact pellet of the oxide. The high mass transfer of the jet ensures that dissolution is controlling. To measure the mass transfer characteristics of the apparatus, solids of the type mentioned earlier have been dissolved in water at ambient temperatures with the submerged, impinging jet. Dissolution rate constants have been measured along with a correlation that will help the interpretation of measurements of the dissolution of magnetite at temperatures and pressures of interest.

## 2. Experimental

### 2.1 Pellet Preparation

Pellets of several materials were prepared in different ways. Cast and pressed pellets were cylindrical, 7.94 mm in diameter and 6 mm long.

Commercial plaster of Paris (90+%:  $\text{CaSO}_4 \cdot 1/2\text{H}_2\text{O}$ ) and pure plaster of Paris (97+%:  $\text{CaSO}_4 \cdot 1/2\text{H}_2\text{O}$ ) were made into pellets by the same method. Plaster of Paris was mixed with water with the ratio of water-to-powder equal to 0.7. Then the wet solution of gypsum was poured into a Teflon mould (which also served as a holder) and dried at room temperature for 30 minutes, resulting in the cast gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) pellets.

A single crystal of gypsum was cut with a lathe into cylindrical pellets of two sizes: 5 mm-diameter and 6 mm-long and 8 mm-diameter and 6 mm-long. The cylinders were mounted in Teflon holders.

Trans-cinnamic acid powder was melted and kept at the melting point (150 °C) for one and a half hours before being poured into Teflon holders to be cast as pellets. After solidification, the top surface of each pellet was polished flat with sand paper.

Potassium bitartrate powder was pressed into pellets at 5,000 psi pressure and then sintered at 170 °C for one hour before being mounted in Teflon holders.

Aspartic acid powder was pressed into pellets at 2,000 psi pressure and then sintered at 210 °C for one hour before being mounted in Teflon holders.

After manufacture, all pellets for insertion in the jet-impingement apparatus were oven-dried at 70 °C for 1 h. After each experiment the pellets were also dried at 70 °C for 1 h before being weighed.

### 2.2 Test section and conditions

The experiments were carried out in a stainless-steel and PVC water loop operating at atmospheric pressure. The water enters the impingement test section from the top and passes directly to the 0.5 mm-diameter nozzle, causing a high-velocity jet of water to be directed onto the pellet. Different flow rates were used for investigating the dissolution of the pellets.

Commercial plaster: experiments were performed at 25 °C for 3 min at flow rates ranging from 40 to 200 mL/min (jet velocities of 3.4 to 16.9 m/s).

Pure plaster: experiments were performed at 34°C for 5 minutes at flow rates ranging from 40 to 180 ml/min (jet velocities 3.4 to 15.3 m/s).

Single-crystal gypsum: experiments with pellets of different diameters were performed at 20°C for 30 minutes for investigating the dissolution at one flow rate (120 mL/min – jet velocity 10.2 m/s).

Trans-cinnamic acid and potassium bitartrate: experiments were performed at 20 °C for 10 min at flow rates ranging from 40 to 180 mL/min (jet velocities 3.4 to 15.3 m/s).

Aspartic acid: experiments were performed at 20 °C for 5 min at flow rates ranging from 40 to 180 mL/min (jet velocities 3.4 to 15.3 m/s).

### 2.3 Pellet characterization

Structures and morphologies of cast pellets were investigated with a scanning electron microscope (SEM) and single-crystal specimens were examined with an optical microscope. Surface profilometry (using a Surtronic 25 and a dial indicator) was used to determine the roughness of pellet surfaces and to measure the extent of dissolution.

## 2.4 Dissolution Rate and Mass Transfer of Pellet

The weight loss of commercial plaster was determined from the volume loss, which was obtained by profilometry and from the density of material, because the measurement by weighing tended to be inaccurate because of inadvertent loss of material from the bulk of the pellet during handling. Then the mass loss was converted to average rate of dissolution ( $\text{kg}/\text{m}^2 \cdot \text{s}$ ) via Equation (1).

$$\text{Dissolution Rate} = \frac{\text{weight loss}}{\text{time} \cdot \text{dissolution area}} \quad (1)$$

The overall rate constant for dissolution ( $K$ ) was derived from the expression:

$$\text{Dissolution rate} = K(C_s - C_b) \quad (2)$$

where  $C_s$  = solubility (literature value) and  $C_b$  = bulk concentration (zero in this experiment).

In terms of the mass transfer coefficient  $k_m$  and dissolution rate constant  $k_d$ :

$$K = k_m \cdot k_d / (k_m + k_d) \quad (3)$$

## 3. Result and Discussion

### 3.1 Pellet characterization

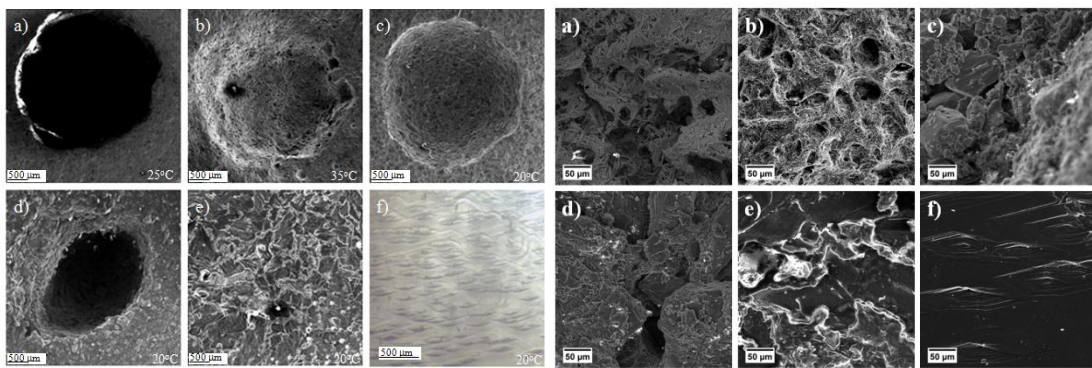


Figure 1: Surfaces of pellets of different materials (a) Commercial plaster at 199 ml/min (b) Pure plaster at 180 ml/min (c) Potassium bitartrate at 180 ml/min (d) Aspartic acid at 180 ml/min (e) Trans-cinnamic acid at 180 ml/min and (f) Single crystal gypsum at 120 ml/min.

The surfaces of commercial plaster, pure plaster, potassium bitartrate and aspartic acid pellets were similar. There was a crater inside each pellet from the driving force of the impinging jet (Figure 1). The trans-cinnamic acid pellet has surface cracks and a shallow crater. The gypsum single crystal surface has numerous shallow, aligned pits, some of which overlap. These pits, related to the crystalline axes, are commonly referred to as being parallelogram-elongated (Chunfang and Henry, 2007).

Inside the crater the porosity of each material is revealed. Figure 2 indicates that commercial plaster has large pores; they presumably affected the dissolution pattern. Pure plaster pores are smaller but their distribution is more regular. Potassium bitartrate porosity is low, no doubt because the powder was pressed at high pressure. On the other hand, aspartic acid (which was pressed at lower pressure) has large cracks inside the crater that appear to divide the solid into large particles. These observations suggest that cast pellets may loosen and then release particles to different degrees as dissolution proceeds; at these higher flows, commercial plaster, pure plaster, aspartic acid and potassium bitartrate apparently tend to release particles more than trans-cinnamic acid and gypsum single-crystal.

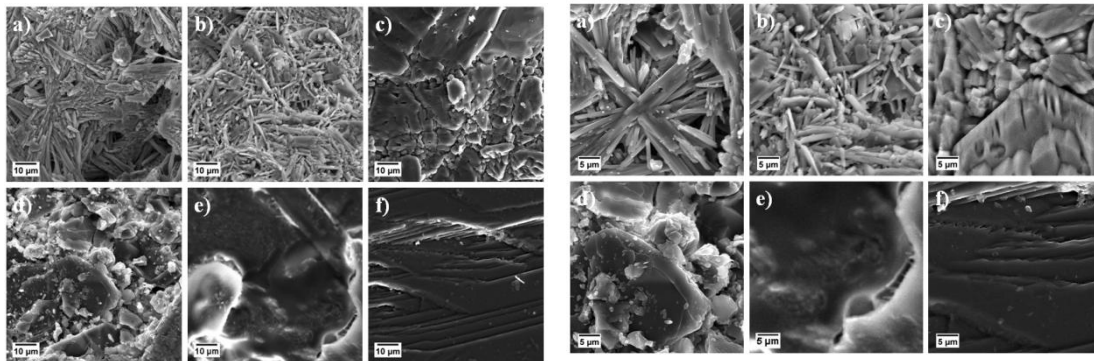


Figure 2: Surfaces of pellets of different materials (a) Commercial plaster at 199 ml/min b) Pure plaster at 180 ml/min c) Potassium bitartrate at 180 ml/min d) Aspartic acid at 180 ml/min e) Trans-cinnamic acid at 180 ml/min and f) Single crystal gypsum at 120 ml/min.

### 3.2 Dissolution rate and mass transfer results

The highest dissolution rates are of commercial plaster; it has values tending to level-off at the highest flow rates (Figure 3). These suggest that the control of dissolution of the cast plaster changed from mass transfer to surface dissolution. The dissolution rates of the other materials (at 20°C, except the pure plaster, which was at 34°C) are lower. The dissolution rate of L-aspartic acid, potassium bitartrate and pure plaster are in the same range. The trans-cinnamic acid and gypsum crystal have rates that

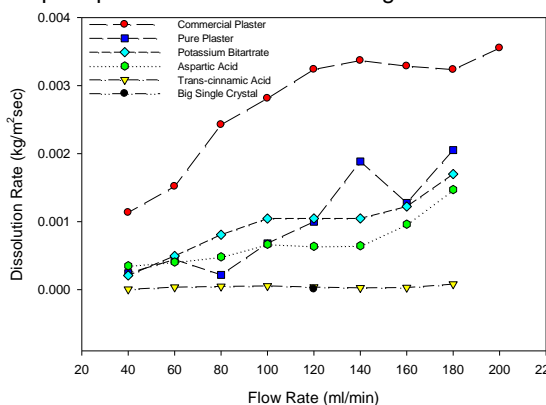


Figure 3: Dissolution rates of pellets of various materials

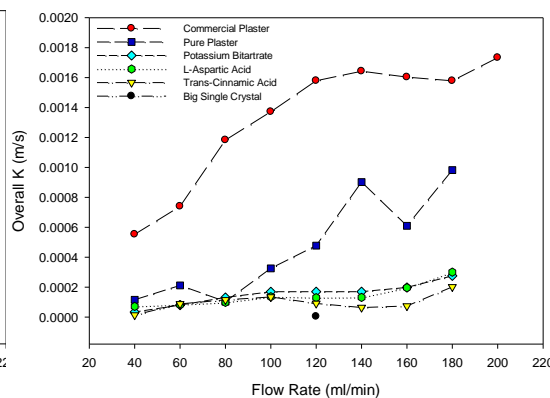


Figure 4: Overall rate constants of pellets of various materials

are obviously lower than other material. The overall rate constant of commercial plaster is higher than that of other materials (Figure 4). However, these results should be in the same range as those of other materials. It can be concluded that the effective solubility of commercial plaster is higher than that of pure plaster or single- materials (except commercial plaster) are crystal gypsum, even though the pure plaster was studied at a higher temperature. At the  $6.45E-05 \pm 5.10E-05$  m/s. This would be lowest flow rate, the overall rate constants of close to each other with an average value of unusually low for a mass-transfer controlled process, so is likely to be influenced mostly by dissolution with any flow effects caused by particle or crystallite removal. Also, our value of the dissolution rate constant of the single crystal gypsum ( $3.37E-6$  m/s) is very close to published values of the dissolution rate constant of mineral gypsum at the same temperature (e.g.,  $3.36E-6$  m/s from Colombani and Bert, 2007). The average dissolution rate of aspartic acid at 100, 120 and 140 mL/min is 1.33 m/s. This corresponds to the value of 1.22 m/s from Shan et al. (2001) at the same temperature. The pure plaster tend to have mixed control between dissolution and mass transfer and the potassium bitartrate dissolution constant is unknown; these two materials are not suitable to determine the apparatus correlation.

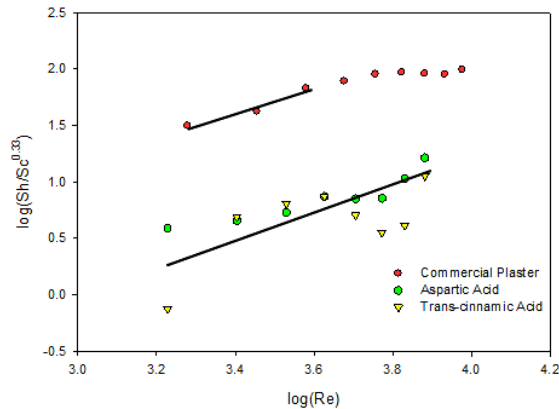


Figure 5: Sherwood number plots for dissolution of cast pellets of various materials various materials

As the experiments were carried out at different temperatures and with different materials, which affects the Schmidt number value, the log-log plots of Sherwood number divided by Schmidt number raised to the power 0.33 against Reynolds number for the liquid jet can lead to the apparatus correlation (Figure 5). The Schmidt number dependence in these correlations is obtained from published impinging-jet correlations; for example, the electrochemical study of jet impingement of Chin and Tsang (1978). In Figure 5, the plot shows that Sherwood numbers divided by Schmidt number for the aspartic acid and trans-cinnamic acid are in the same range.

The correlation can be found as:

$$Sh = 2.29E-03 Re^{0.94} Sc^{0.33} \quad (4)$$

On the other hand, for the commercial plaster the data at lower flows (where mass transfer rate approaches the limiting step) produce the correlation:

$$Sh = 9.05E-03 Re^{1.07} Sc^{0.33} \quad (5)$$

These two equations can be compared with the correlations of Rao and Trass (1964):

$$Sh = 0.046 Re^{1.06} (H/d_0)^{-0.09} \quad (6)$$

and Chin and Tsange (1978) :

$$Sh = 1.12 Re^{0.5} (H/d_0)^{-0.057} \quad (7)$$

because they involved a submerged jet similar to the experimental apparatus [note :  $(H/d_0)$  is relative jet length and for our jet geometry  $H$  was 3.0 mm and  $d_0$  is 0.5 mm]. The Schmidt number of  $CaSO_4 \cdot 2H_2O$  at 25°C (983) and the average Schmidt number of aspartic acid and trans-cinnamic acid at 20°C (1122) can be used to transform our correlation. The Reynolds numbers at 180 ml/min and 20°C for aspartic acid and trans-cinnamic acid correlation and 80 ml/min and 25°C for commercial plaster correlation are also used to determine Sherwood number. The Sherwood number from the aspartic acid and trans-cinnamic acid correlations ( $Sh = 103$ ) is much lower than that from published expressions (Rao and Trass;  $Sh = 895$ , Chin and Tsange;  $Sh = 509$ ) for similar equipment, no doubt because of dissolution's having the major effect. On other hand, the Sherwood number from the commercial plaster correlation ( $Sh = 595$ ) is closer to the Sherwood number from the Chin and Tsange correlation ( $Sh = 606$ ) than to that from the Rao and Trass correlation ( $Sh = 244$ ), although the power dependence of the Reynolds number seems low in the Chin and Tsange expression. Deviations from the published expression are probably due to the ranges of Reynolds number in the literature (Chin and Tsange;  $4,000 < Re < 16,000$ , Rao and Trass;  $20,000 < Re < 125,000$ ) that are higher than in this experiment ( $1901 < Re < 3,802$ ), as well as to the effects of particle/crystallite release.

#### **4. Conclusion**

In the jet-impingement apparatus at temperatures close to ambient, commercial plaster of Paris gave a mass-transfer correlation in reasonable agreement with a published correlation, although presumably particle or crystallite release gave a somewhat higher power of Reynolds number than the published value. The levelling-off of dissolution rate at higher flows was attributed to the involvement of control by dissolution. The apparatus gave values for the dissolution rate constant of single-crystal gypsum and cast aspartic acid in good agreement with literature values. Cast/pressed pellets of pure plaster of Paris, potassium bitartrate, aspartic acid and trans-cinnamic acid dissolved slowly by comparison with commercial plaster and were largely under dissolution control; flow effects were then attributed to the release of particles or crystallites.

The apparatus would appear to have predictable mass-transfer characteristics and be appropriate for measuring the dissolution rates of sparingly-soluble materials. A more precise correlation should be obtained by further experiments over a wider range of experimental variables, in particular temperature.

#### **Acknowledgement**

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