# CHAPTER III
# EXPERIMENTAL

## 3.1 Materials and Equipment

### 3.1.1 Equipment:

- Scanning Electron Microscope (SEM: JSM-6400)
- Optical microscope (Olympus: GX41)
- Surface profilometry (Surtronic 25 and dial indicator)

### 3.1.2 Instrument:

**Water loop:**

- Centrifugal pump
- Ball valves
- Needle valve
- Flow meter
- 1"-Vinyl pipe
- 1"-PVC pipe
- ¼"-Stainless steel pipe
- Filter

**Test section:**

- Union cross
- Heat shrink Teflon
- Stainless steel cover
- Teflon sleeve
- Jet nozzle
- Teflon disk
- Reducing union
- Rulon Support
- Stainless steel holder

3.1.3 Chemicals:

- Water supply, Fredericton city water

- Single crystal of Gypsum ($CaSO_4 \cdot 2H_2O$)

- Plaster of Paris (90+%:$CaSO_4 \cdot 1/2H_2O$)

- Plaster of Paris (97+%:$CaSO_4 \cdot 1/2H_2O$)

- Trans-cinnamic acid ($C_9H_8O_2$)

- Potassium hydrogen tatrate ($C_4H_5O_6K$)

- L-Aspartic acid ($C_4H_7NO_4$)

- De-ionized Water

## 3.2 Experimental Procedures

After manufacture, pellets for insertion in the jet-impingement apparatus were oven-dried at 70 °C for 1 hour. After each experiment the pellets were also dried at 70 °C for 1 hour before being weighed. Teflon molds were used to contain all pellets. The size of the Teflon mold is 7.94 mm diameter and 6 mm length.
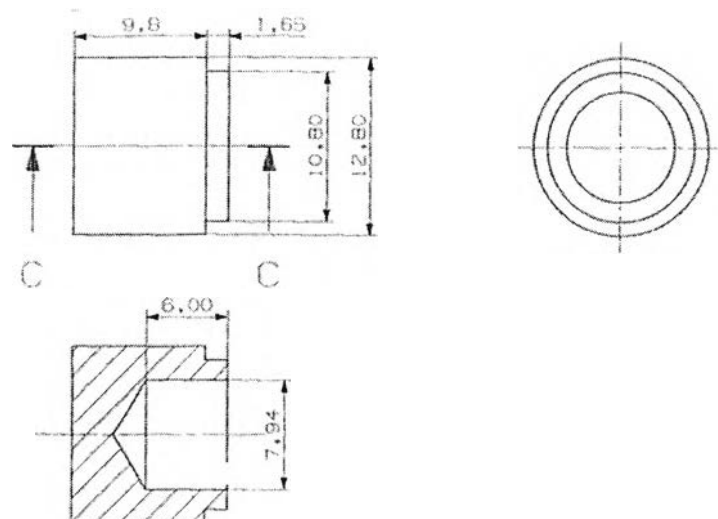


Figure 3.1 Drawing of teflon holder/mold (in millimeter unit).

### 3.2.1 Gypsum Pellet Preparation

#### *3.2.1.1 Plaster of Paris (CaSO$_4$·1/2H$_2$O)*

Normally, the hydration reaction between plaster of Paris and water requires 18.6% of water, but for easier casting and for higher fluidity, an excess of water is added. The porosity of plaster can be controlled during the drying process. Air bubbles are often trapped during mixing; they cause brittleness of the material, which will affect the test. Therefore, to minimize the formation of air bubbles, the two different purities of plaster were thoroughly mixed with de-ionized water in vacuum chamber (Figure 3.2). Slurries prepared vacuum are significantly denser and have fewer air bubbles than those prepared under open atmospheric conditions.

Commercial plaster of Paris (90+%:CaSO$_4$·1/2H$_2$O) and pure plaster of Paris pellets (97+%:CaSO$_4$·1/2H$_2$O) were prepared by the same method. The ratio of water-to-powder, R$_o$, (R$_o$ = M$_{water}$/M$_{plaster}$) affects the mechanical properties. The final consistency was optimized by varying the ratio R$_o$ during the mixing. A small R$_o$ led to a very compact and dense material but the plaster was too thick to be cast. On the other hand, when the fluidity of plaster was increased by adding water, the plaster was more brittle at larger R$_o$. A compromise was found when the plaster powder was mixed with de-ionized water in the ratio R$_o$ = 0.7 as a regular procedure; most of the air bubbles in plaster were easily removed (Sinthuphan, S., 2008).
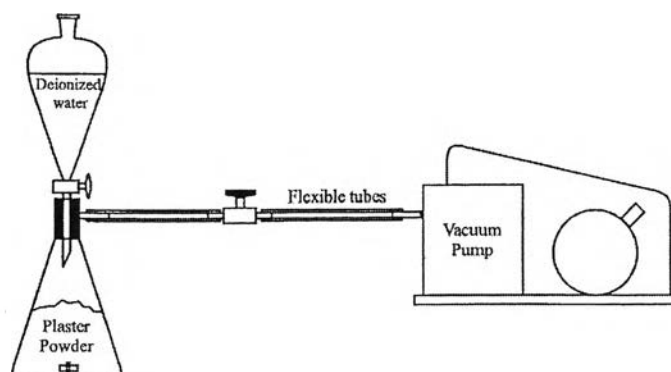


**Figure 3.2** Schematic of the mixing apparatus (Villien et al., 2001).

After the plaster powder was mixed with water, the wet solution of gypsum was poured into the Teflon holder and dried at room temperature for 30 minutes (setting time of gypsum). Then it was dried in the oven at 70 °C for 1 hour before being put into the test section. After each experiment the pellet was also dried 70 °C for 1 hour before being weighed.

### 3.2.1.2 Single Crystal of Gypsum

The single crystal was cut into a cylinder shape with a lathe machine as two sizes: 5 mm-diameter and 6 mm-long and 8 mm-diameter and 6 mm-long.
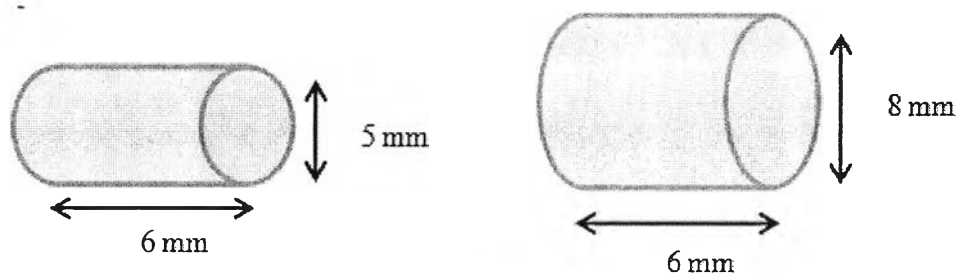


**Figure 3.3** Schematic of gypsum pellet.

### 3.2.1.3 Trans-Cinnamic Acid

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

### 3.2.1.4 Potassium Bitartrate

Potassium bitartrate powder was pressed into pellets at 5000 psi pressure for 10 minutes and then sintered at 170 °C for one hour before being mounted in Teflon holders.

### 3.2.1.5 Aspartic Acid Powder

Aspartic acid powder was pressed into pellets at 2000 psi pressure for 10 minutes and then sintered at 210 °C for one hour before being mounted in Teflon holders.

## 3.3 Test Loop
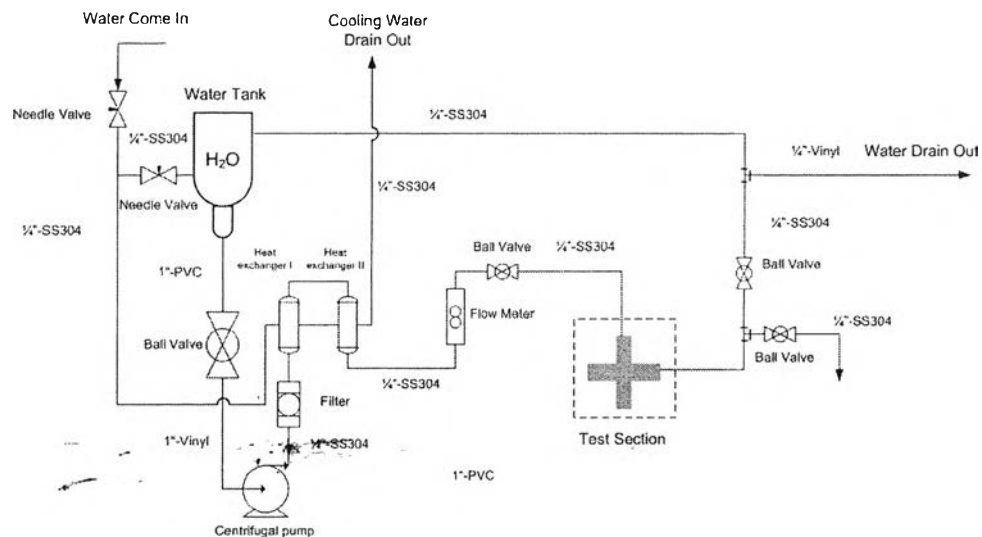


**Figure 3.4** Schematic of water loop for the dissolution rate experiment.

The experiments were carried out in a stainless-steel and PVC water loop operating at atmospheric pressure and room-temperature (Figure 3.4). First, water from the service system passed through a ball valve and went to the 50 liter water tank reservoir. Second, water passed through the centrifugal pump, along the vinyl pipe. Third, water went through a filter to get rid of contamination. Fourth, water directly flowed to the flow meter that was used for controlling the flow rate. Fifth, water went to the heat exchangers I and II that were used for controlling the temperature. Sixth, water went to another ball valve; this valve was usually turned off after each experiment for stopping water flow. Seventh, water entered the top of the test section contained the pellet. Finally, water passed through the ball valve before passing to a 3-way connection and draining out. In addition, another ball valve was used to calibrate the water flow. Cooling water that was used in heat exchanger came from the service water. Then, cooling water passed to the needle valve that was used for controlling the flow rate. The water tank was connected with a pipe to drain out the water for preventing an overflow.

## 3.4 Test Section

The test section is shown in Figure 3.5. The main body material is ¼" 316L stainless steel union cross (A) connected to several fittings. The water enters from the top, passes through a 1/4" 316SS tubing (B) and passes directly to a 0.5 mm-diameter jet nozzle (C). A Rulon piece (D) is used to support the shoulder between the cross and the end of a ¼ "SS tube (E).

Each pellet is sheathed with heat-shrink Teflon (F) and surrounded by its Teflon holder (G). The whole is covered in a machined 304SS piece (H). This (H) piece is on top of a 304SS rod (I) and is tightly held in position between the shoulder and the end of the cover (J).

The jet region is adjusted by adding a 2 mm thick shim in the right leg of the union cross before swaging the fitting. The ¼" 316SS tubing (K) is connected to a reducer (L) which directs the water flow to drain. The other side of the union is shut with a cover (M).
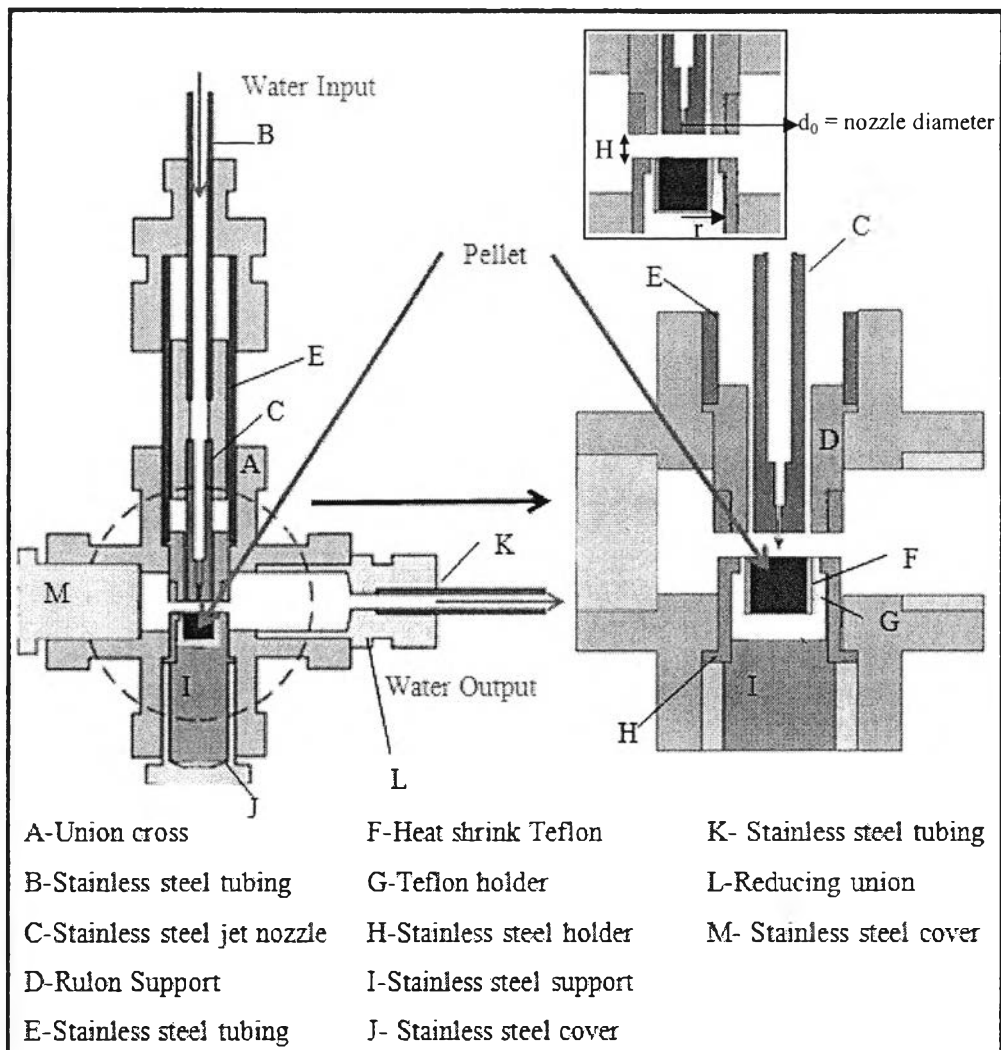
Figure 3.5 Schematic diagram of the test section.

## 3.5 Test Conditions

### 3.5.1 Plaster of Paris (CaSO$_4$·1/2H$_2$O)

#### 3.5.1.1 Commercial Plaster of Paris (90+%:CaSO$_4$·1/2H$_2$O)

Different flow rates were used for investigating the dissolution of the gypsum pellets (Plaster of Paris) at room temperature. Flow rates ranged from 40 to 200 ml/min, so they caused different values of Reynolds number. Pressure, temperature and pH remained the same.

**Table 3.1** Runs for studying the effect of flow rates by using Plaster of Paris mixed with de-ionized water (pH 7 ± 1, pressure 1 atm, 3 mins, 25 °C)

| Run | Flow rate (ml/min) |
|-----|--------------------|
| 1 | 200 |
| 2 | 180 |
| 3 | 160 |
| 4 | 140 |
| 5 | 120 |
| 6 | 100 |
| 7 | 80 |
| 8 | 60 |
| 9 | 40 |

*3.5.1.2 Pure plaster of Paris (97+%:CaSO4·1/2H2O)*

The experiments were performed at 35 °C at flow rates ranging from 40 to 180 ml/min.

**Table 3.2** Runs for studying the effect of flow rates by using pure plaster (pH 7 ± 1, pressure 1 atm, 10 mins)

| Run | Flow rate (ml/min) |
|---|---|
| 1 | 180 |
| 2 | 160 |
| 3 | 140 |
| 4 | 120 |
| 5 | 100 |
| 6 | 80 |
| 7 | 60 |
| 8 | 40 |

### 3.5.2 Single Crystal of Gypsum

Two different diameters were used for investigating the dissolution of the single crystal of gypsum pellets at room temperature. Pressure, temperature, flow rate (120 ml/min) and pH remained the same.

**Table 3.3** Runs for studying the effect of flow rates by using single crystal (pH 7 ± 1, pressure 1 atm, 30 mins, 120 ml/min, 20 °C)

| Run | diameter (mm) |
|---|---|
| 1 | 8 |
| 2 | 5 |

### 3.5.3 Trans-Cinnamic Acid and Potassium Bitartrate

Different flow rates were used for investigating the dissolution of the Trans-cinnamic acid pellets and potassium bitartrate at 20 °C for 10 mins. Flow rates ranged from 40 to 180 ml/min (same as pure plaster), so they caused different values

of Reynolds number. Pressure, temperature and pH remained the same (pH 7 ± 1, pressure 1 atm).

### 3.5.4 L-Aspartic Acid

Aspartic acid experiments were performed at 20 °C for 5 minutes at flow rates ranging from 40 to 180 ml/min same as pure plaster (pH 7 ± 1, pressure 1 atm).

## 3.6 Analytical Techniques

### 3.6.1 Characterization of Pellet Surface

Typical surface morphology of pellets was determined by using a digital camera and optical microscope. Photographs were taken at any given time and after each experiment. In addition, the structure and the condition of the pellet were examined with a scanning electron microscope (SEM). Surface profilometry (Surtronic 25 and dial indicator) was used to determine the roughness of pellet surfaces and to measure the extent of dissolution.

### 3.6.2 Dissolution Rate and Mass Transfer of Pellet

The mass loss was obtained by weighing a pellet before and after the test. The value was compared with that obtained by profilometry. The data from profilometry can be used to calculate volume loss of pellet by using the integration, followed by equation (3.1).

$$\text{Volume} = 2\pi \int_0^x xy\,dx \qquad (3.1)$$

where y is the depth at x position. Then the volume loss can be converted to mass loss by using the density of each material. In addition, the dissolution area can also be calculated from integration as equation (3.2).

$$\text{Area} = \int_0^\theta \int_0^r \left[ \sqrt{\left(cos\theta \frac{dz}{dr} - \frac{1}{r}\sin\theta \frac{dz}{d\theta}\right)^2 + \left(\sin\theta \frac{dz}{dr} + \frac{1}{r}\cos\theta \frac{dz}{d\theta}\right)^2 + 1} \right] r\, dr\, d\theta \quad (3.2)$$

where z is the depth at r position and $\theta$ is angle at r position. Then the average rate of dissolution (kg /m$^2$sec) can be determined from equation (3.3).

$$\text{Dissolution Rate} = \frac{\text{weight losss}}{\text{time·dissolution area}} \quad (3.3)$$

The overall rate constant for dissolution (K; m/s) was derived from equation (3.4).

$$\text{Dissolution rate} = K(C_s\text{-}C_b) \quad (3.4)$$

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

The correlation of the mass transfer coefficient $k_m$ and dissolution rate constant $k_d$ is shown in equation (3.5).

$$K = k_m k_d/(k_m + k_d) \quad (3.5)$$

Sherwood number can be calculated via equation 3.6.

$$Sh = \frac{k_m d_0}{D_{AW}^\circ} \quad (3.6)$$

where $k_m$ is mass the transfer coefficient (m/s), $d_0$ is the nozzle diameter (m) and $D_{AW}^\circ$ is the diffusivity of species A in water (m$^2$/s). Sherwood number (Sh) depends on the Reynolds (Re) and Schmidt (Sc) numbers, the relative jet length (H/$d_0$) and the relative radial distance from the stagnant point (r/$d_0$). The Reynolds number and Schmidt number equation can be followed by equations 3.7 and 3.8.

$$Re = \frac{\rho VL}{\mu} = \frac{VL}{\nu} \qquad (3.7)$$

where V is the mean jet velocity, L is a characteristic linear dimension (m), $\mu$ is the dynamic viscosity of the fluid (Pa·s or N·s/m² or kg/(m·s), $\nu$ is the kinematic viscosity ($\nu = \mu / \rho$) (m²/s) and $\rho$ is the density of the fluid (kg/m³).

$$Sc = \frac{\nu}{D} = \frac{\mu}{\rho D} \qquad (3.8)$$

The Reynolds number in these experiments should be around 1,600 to 10,500 so we can compare the values of Sherwood number by using equations in Table 2.7. These mass transfer correlations were evaluated and the appropriate one chosen.