Study of Solid Dissolution in Fixed-bed with Downward Flow of the Liquid

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The mass transfer coefficient in solid dissolution in fixed bed of spherical particle with downward flow of the liquid has been studied experimentally in urea-distilled water system. The influence of different parameters, such as: particle size, height of the bed, liquid flow rate, on the mass transfer in dissolution in packed bed are also investigated. The experimental values of the mass transfer coefficients were in good accordance with those calculated from the Cussler and Cho equations.

Keywords: solid dissolution, mass transfer coefficient, packed bed, active sphere, liquid flow

Applications of fluid-solid systems are often encountered in practice and are used for a long time. Fluid-solid systems are an alternative for achieving a good contact between the phases and, depending on the bed structure, have different names [1-3]: particles in fixed bed systems, systems with moving particles (fluidized bed, moving bed, hydraulic transport).

Contacting the solid-phase with a liquid in fixed bed is characterized by a large area of contact between the phases which results in a very good heat and mass transfer and has applications in chemical industry, petroleum industry, food industry, pharmaceutical industry, energy industry, metallurgy, building materials, nuclear industry, waste processing etc.

Study of mass transfer in solid-liquid systems in fixed and fluidized bed can be achieved by dissolution, adsorption, ion exchange, electrochemical methods [4-17].

The dissolution technique is used, essentially at atmospheric pressure to determine the mass transfer coefficients between the solid particles from fixed bed and a fluid moving upward or downward.

To investigate the solid-liquid mass transfer in fixed and fluidized bed dissolution, several methods may be used:
- columns with short or long bed of active particles,
- columns with long bed from inert particles (glass spheres, sand) in which is inserted a single active particle, are dispersed more particles or a bed of active particle is placed between two beds of inert particles.

Active particles may be made from molted substance or by coating a spherical or cylindrical core made from different materials (sand or glass) with a thin layer of molted substance. Active substance may be urea, benzoic acid, salicylic acid, naphthalene, β-naphthol etc.

Distilled water or an aqueous solution of the active substance may be used as dissolution medium.

The heterogeneous mass transfer is influenced by hydrodynamics of the system parameters: fluid flow rate, bed porosity, concentration of particles in the layer.

The methods for determining the concentration of the solution used depend on the nature of the solute: by titration, with UV spectrometer.

In this paper, the solids dissolution study in fixed bed was carried out with downward flow of the dissolution medium. Through this study we aimed to determine the mass transfer coefficients; follow the influence of various parameters on the dissolution (particle diameter, the height of the particles bed, the flow rate of the dissolution medium); compare the experimental results with calculated values based on existing models in the literature.

Experimental part

Study of solid dissolution in fixed bed with downward flow of the dissolution medium was carried out on a laboratory device (fig. 1) consisting of a cylindrical glass column with an internal diameter of 78 mm and a height of 480 mm provided at bottom with a perforated plate which supports the granular material, centrifugal water pump, an electric motor for driving the pump, rotameter for determining the flow of water, distilled water tank and solution of urea tank, digital thermometer, valves.

Experimental determinations have been made using urea particles having a diameter of 2.25 and 2.825 mm. Physical and flow properties of urea were investigated experimentally [18]: particle size, bulk and tapped densities, bed porosity and specific surface area.

Urea dissolution study was conducted in fixed bed at atmospheric pressure and a temperature of 25°C using distilled water as the dissolution medium. Height of the fixed bed has been altered by the amount of material used: 100, 200, 300 and 400 g.

The liquid flow rate was: 50 L h⁻¹, 75 L h⁻¹, 100 L h⁻¹, 150 L h⁻¹. The minimum amount of fluid flow has been established in order to ensure the recommended minimum strength of the spraying material [19].
To determine the mass transfer coefficient, urea concentration in the effluent and urea concentration of the solution in the storage tank were determined at different time intervals. Sample analysis was performed using a refractometer and a calibration curve experimentally determined.

**Results and discussions**

The experimental results are presented in figures 2 and 3 as the variation of urea concentration in effluent in time for several values of liquid flow rate and for several values of amount of urea in column.

Increasing the flow rate of the dissolution medium decreases the urea concentration in the effluent and reduces the dissolution time of the sample.

Increasing the amount of the material in fixed bed (the height of the bed) results in an increase in urea concentration in the effluent (due to the increased distance traveled by the liquid) at all values of the liquid flow rate. Also, there is an increase of the dissolution time of the material.

The quantity of urea from the solution storage tank was calculated based on the volume of the solution and the concentration of urea in the solution.

Depending on the amount of urea in the solution tank we calculated:
- the quantity of urea dissolved,
- the amount of urea remaining in the column (non-dissolved).

Figure 4 presents the variation of the three quantities (the amount of urea in the solution tank, the amount of urea in the dissolution column and the dissolved quantity of urea) in time for samples of 200 g urea with particle diameter of 2.25 mm and 2.825 mm at a flow rate of 100 L liquid h⁻¹.

The dissolution of urea with a particle size of 2.25 mm occurs faster because the solid-liquid contact area is bigger than in the case of particles with 2.825 mm.

The degree of dissolution of the material was calculated from relation:

\[
\eta = 1 - \frac{m_f}{m_0}
\]

where:
- \(m_f\) – the amount of material in column;
- \(m_0\) - the amount of urea introduced into the column.

The obtained results are presented in figure 5 for dissolution of the sample with 400 g urea and particle of 2.25 mm in diameter.

In order to determine the mass transfer coefficient of the following simplifying hypotheses have been considered: urea particles are spherical of the same diameter and the same rate of dissolution, the number of particles remains constant during the dissolution and change in the form and the particle size is uniform.

The number of particles in the column is determined from the amount of material introduced into the column:

\[
N_p = \frac{6 \cdot m_0}{\pi \cdot \rho_p \cdot d_0^3}
\]

where:
- \(N_p\) - number of particles;
- \(m_0\) - the amount of urea introduced into the column, [kg];
- \(\rho_p\) - urea density [kg m⁻³],
- \(d_0\) - initial diameter of the urea particle [m].

The particle diameter at a certain time of the process may be determined from the amount of material non-dissolved in the column:

\[
d_p = \left(\frac{6 \cdot m_{\text{res}}}{\pi \cdot \rho_p \cdot N_p}\right)^{1/3}
\]
The changes in particle diameter during dissolution are shown in figure 6.

The mass transfer coefficient can be determined from the experimental results using the relation:

\[ k = \frac{\Delta m}{A \cdot \Delta t \cdot \Delta C_{med}} \]  

where:
- \( k \) – the mass transfer coefficient, \([\text{m s}^{-1}]\);
- \( \Delta m \) - the amount of dissolved substance, \([\text{kg}]\);
- \( A \) - contact surface area \([\text{m}^2]\);
- \( \Delta t \) - dissolution time \([\text{s}]\),
- \( \Delta C_{med} \) - mean driving force of the mass transfer \([\text{kg m}^{-3}]\).

The mass transfer area is variable during the dissolution and can be calculated depending on the number and the particle diameter of the column:

\[ A_i = \pi \cdot N_p \cdot d_i^2 \]  

where:
- \( A_i \) - the particles surface in the column at a certain time \([\text{m}^2]\);
- \( d_i \) - the average particle diameter in the time interval calculated as the arithmetic mean of the value at successive times of measurement of the concentration \([\text{m}]\).

Cussler [20] has shown that the mass transfer coefficient at the dissolution in fixed bed using pure solvent is about the same for the driving force for the log mean and for the driving force determined by the difference between the saturation concentration and the final concentration of the solute in the solution, but the log mean driving force is recommended.

\[ \Delta C_{med} = \frac{(C^* - C_i) - (C^* - C_f)}{\ln \frac{C^* - C_i}{C^* - C_f}} \]  

where:

- \( C^* \) - the saturation concentration \([\text{kg m}^{-3}]\),
- \( C_i \) - concentration of urea in the dissolution medium at the entry into the column (0 if the fluid is pure) \([\text{kg m}^{-3}]\),
- \( C_f \) - concentration of urea in effluent \([\text{kg m}^{-3}]\).

Figure 7 shows the mass transfer coefficient values for urea dissolution in fixed bed with downward flow of liquid.

The values of the coefficient of mass transfer vary slightly over time. Increasing the flow rate of the liquid has a positive effect on the mass transfer coefficient which can be explained by greater liquid velocity in intergranular spaces.

Increasing the height of the fixed bed increases urea concentration values in the effluent, as well as the mass transfer driving force and reduces the mass transfer coefficient values (fig. 8).

Theoretical values of mass transfer coefficient have been determined from the Cussler [20] relation:

\[ k = 1.17 \cdot v_0 \left( \frac{d}{v} \right)^{0.42} \left( \frac{D}{V} \right)^{0.66} \]  

and from the Chu [1, 2, 20] correlation which applies to the mass transfer in fixed and fluidized bed:

\[ j_m = 5.7 \cdot Re_m^{0.78} \text{ for } 1 < Re_m < 30 \]  

where:
- \( k \) - mass transfer coefficient, \([\text{m s}^{-1}]\);
- \( d \) - the particle diameter, \([\text{m}]\);
- \( v_0 \) - speed of the fluid in the column, \([\text{m s}^{-1}]\);
- \( v \) - kinematic viscosity of the fluid, \([\text{m}^2 \text{s}^{-1}]\);
- \( D \) - diffusion coefficient, \([\text{m}^2 \text{s}^{-1}]\);
- \( j_m \) - Chilton-Colburn mass transfer factor
- \( \text{Re}_m = \frac{M_m \cdot d}{\nu} \) Reynolds number modified
- \( \text{Sc} = \frac{\nu}{D} \) Schmidt number
Experimental and theoretical values of the mass transfer coefficient determined from relations (4), (7) and (8) are shown in figure 9. The correlation between experimental and theoretical values calculated using the equation (7) is good in the most part of the dissolution process. Theoretical values determined from the relation (8) are in good agreement with the experimental results for the start of the dissolution and then increase significantly. This disparity is the result of the characteristics changes of the material layer, in particular, at small values of the particle diameter.

Conclusions

The dissolution of urea spherical particles was studied in fixed bed by downward flow of the dissolution medium. The experimental results led to the following conclusions:
- the urea dissolution is influenced by the particle size of urea, by the bed height, by flow rate of dissolution medium;
- the dissolution time increases with particle size and bed height increase and decreases with liquid flow rate increase;
- the mass transfer coefficient has been positively influenced by increase of the flow rate of the liquid and by decrease of the height of the material bed;
- the correlation between experimental and theoretical values of the mass transfer coefficient is better for the Cussler relation and for the first part of the dissolution when the bed characteristics are not significantly affected.

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