



Dissolution kinetics of natural magnesite in acetic acid solutions

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Abstract

Dissolution of magnesite in acetic acid solutions was investigated. The influence of various parameters such as reaction temperature, particle size and acid concentration was studied in order to elucidate the kinetics of magnesium carbonate. The leaching rate increased with decreasing particle size and with increasing temperature. Initially, the dissolution in terms of acid concentration increased until a definite concentration and then fell with increasing concentration. A kinetic model was researched to describe the dissolution and to analyse the kinetic data, basically. Dissolution curves were evaluated in order to test shrinking core models for fluid–solid systems. Consequently, it was determined that the dissolution of natural magnesite was controlled by chemical reaction, i.e., $1-(1-x)^{1/3}=kt$. The apparent activation energy of leaching process was found as $78.40 \text{ kJ mol}^{-1}$.

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1. Introduction

Magnesite ore, still, is the basic raw material for manufacturing of magnesium and its compounds. Also, these products have rather wide usage fields and their costs are high (Bengtson, 1999; Maniocha, 1997; Bukovisky, 1997). In order to recover magnesium and its compounds from magnesite ore, the

hydrometallurgical methods are usually used (Kovacheva et al., 2001).

In this direction, as leaching agent, generally used are the chemical compounds such as inorganic/organic acids or bases and their salts. Although magnesite dissolution has been examined with inorganic reagents (Fredd and Fogler, 1998; Economou et al., 2002), the studies concerning dissolution kinetics of magnesite in organic acid have nearly been limited (Demir et al., 2003).

Organic acids have high selectivity although their dissolving abilities are weak. Therefore, it is advantageous for particularly dissolution of carbona-

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ceous compounds. Also, in scale-up studies conducted with inorganic acid, high CO₂ pressure and froth forming owing to fast dissolution can lead to some risks. Organic acids can be an attractive extracting agent because the extraction is performed at mildly acidic conditions (pH 3–5); their degradation is biologically easy (Veeken and Hamelers, 1999). Additionally, in the industrial processes, organic acids can cause a little corrosion effect (Bilgiç, 2002). However, the organic acids could not be generally used as leaching agent for hard dissolving compounds. Also, use of organic acids at high temperature may be limited because of low boiling temperatures and their decomposition.

In aquatic solutions, acetic acid is weakly dissociated ($pK_a=4.76$) and at concentrations of 1.0, 0.1 and 0.01 M the resulting pH is 2.4, 2.9 and 3.4, respectively. Many metals, as well as their oxides and carbonates, dissolve in aqueous solutions of acetic acid to give simple salts. The reactions are considerably slower than those of hydrochloric acid or sulfuric acid, but the rate is still higher 10–11 times than with most other organic acids (Wagner, 1978). Today, acetic acid is also widely used as a solvent in the chemical industry and a raw material for many organic syntheses such as the manufacture of vinyl acetate and cellulose acetate. In addition, calcium magnesium acetate can be used as an additive to coal-fired combustion units, for example, boilers used by electrical utilities.

Numerous studies for the dissolution of magnesite were found in the literature (Jordan et al., 2001; Ekmekyapar et al., 1993) and some of them shall be briefly discussed at this point. The dissolution kinetics of magnesite in water saturated by chlorine gas was studied by Özbek et al. (1998) and it was found that the dissolution process was also controlled by surface reaction. Also, Demir et al. (2003) found that the leaching kinetics of magnesite in citric acid solutions was controlled by chemical reaction in developing semi-empirical model. The activation energy of the process was determined as 61.35 kJ mol⁻¹.

Abali et al. (1992) examined the reaction kinetics of magnesite with SO₂ gas in aqueous medium. The results obtained from experiments showed that the dissolution rate was controlled by surface reaction and the activation energy for process was 81 kJ mol⁻¹.

Also, Kurtbaşı et al. (1992) investigated the dissolution kinetics of magnesite in HCl solution and it was determined that the dissolution rate was controlled by surface reaction. Another interesting search discussed here was the comparative study by Chou et al. (1989), who investigated the dissolution of various carbonates (including calcite, magnesite and dolomite) in HCl solutions at 25 °C by using a continuous fluidized bed reactor and samples of relatively coarse particle size. A chlorination study of magnesium carbonate by Kennedy and Harris (2000) was performed in a stirred tank reactor. Chlorination rates were measured over a range of temperatures from 740 to 910 °C. Activation energy of process was calculated as 80 kJ mol⁻¹ over the temperature range from 740 to 825 °C and the fastest chlorination was reached at a temperature of 860 °C.

The present research aimed to study the leaching kinetics of natural magnesite in the acetic acid solutions.

2. Methods and materials

The magnesium carbonate ore used in the work was provided from Erzurum-Oltu in Turkey. After crushing and washing, the sample was ground, and its chemical composition was analyzed by standard gravimetric and volumetric methods (Furmann, 1963). The results were given in Table 1. An X-ray diffractogram illustrating the contents of the sample was given in Fig. 1. The ore was sieved using ASTM standard sieves, giving particle size fractions of 138, 215, 478 and 855 µm.

The acetic acid, CH₃COOH, used as leachate was of reagent grade. The main production routes for acetic acid were liquid-phase oxidation of *n*-butane and methanol carbonylation, both with feedstocks derived from natural gas or petroleum (Busche, 1990;

Table 1
Chemical analysis of the natural magnesite

Component	[wt.%]
MgO	45.95
CaO	1.40
Fe ₂ O ₃	0.52
SiO ₂	1.98
Loss on ignition [at 850 °C]	50.15

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