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Effect of ultrasonic irradiation on morphology and polymorphic transformation of glycine

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ABSTRACT

The polymorphic phase transformation of β-glycine to α-glycine was analyzed in the absence and presence of three different intensities of ultrasonic irradiation in a batch system at 25 °C. The experiments were performed to explore the effects of ultrasonic irradiation and its intensity on phase transformation time, morphology, storage stability, and filtration characteristics. The crystals obtained with and without ultrasound were characterized by X-ray diffraction (XRD). The XRD results showed that the ultrasound tended to reduce the transformation time. According to the scanning electron microscopy (SEM) images, the ultrasound had a significant effect on the crystal morphology. The highest ultrasound power applied yielded more rounded crystals compared to the crystals prepared in the absence of ultrasound. In addition to SEM analysis, detailed crystal shape analysis was performed and shape factors, namely the circularity, elongation, and convexity values, were determined quantitatively to get more information about the morphological changes related to the variations in the size and shape of the end products. From the morphological point of view, the circularity values of the crystals obtained under ultrasound were higher but the elongation values were smaller compared to the crystals obtained in the absence of ultrasound. It was also found that higher ultrasound decreased the length and width of the crystals, and the application of ultrasound increased the aspect ratio value from 0.483 to 0.657.

1. Introduction

Glycine is the simplest amino acid and has widespread use in the chemical, cosmetics, pharmaceutical, and food industries. Its three polymorphs under atmospheric temperature and pressure are the γ, α, and β forms, which are the most stable polymorph of glycine, the metastable polymorph of glycine, and the unstable polymorph of glycine, respectively [1–5]. α-Glycine at room temperature is stable and spontaneously crystallizes from aqueous solution. Both β and γ nucleation are usually obtained only under certain conditions, such as in the presence of alcohol and additives, or with technical processes at extreme temperatures [6,7]. Polymorphs generally have different chemical and physical properties, including stability, crystal shape, compressibility, melting point, thermal conductivity, heat capacity, density, and dissolution rate [8]. These differences in chemical and physical properties result in the need to handle and process the materials differently [9]. Thus, the control of polymorphism is of great importance for efficient crystal production [10]. Ultrasound is used to improve the quality of the end-products and control polymorphism. Use of ultrasound in crystallization processes is reported to alter the crystal habit and crystal size distribution, help or hinder agglomeration, and facilitate product handling [9]. Some studies have indicated that different polymorphs can be formed with different physical properties when systems are treated with ultrasound. Nii and Takayanagi [11] performed a study to investigate the impact of high-frequency ultrasound on the crystallization behavior of the polymorphs of glycine in a water–ethanol system. The transformation between β and α forms was carried out in non-sonicated and sonicated media. Ultrasonic transducers operating at 1.6 MHz and 20 kHz were used and the influence of ultrasonic frequency was investigated by comparing the resulting crystals produced by these two ultrasound frequency. They found that the crystal size was reduced at low frequency ultrasound; however, ultrasound enhanced both the growth rate of α-glycine crystals and the incorporation of microcrystals into larger crystals. Renuka et al. [12] studied to determine the higher ultrasonic frequency effects on the nucleation and growth characteristics of glycine. The experiments were performed at different ultrasound frequencies in the range of 1–10 MHz. They found that higher ultrasound yielded a higher nucleation rate but with a smaller crystal size compared to the crystals obtained in the absence of ultrasound. A similar study performed by Louhi-Kultanen et al. [13] showed that ultrasound affected the crystal morphology and crystal size distribution of glycine. These studies were

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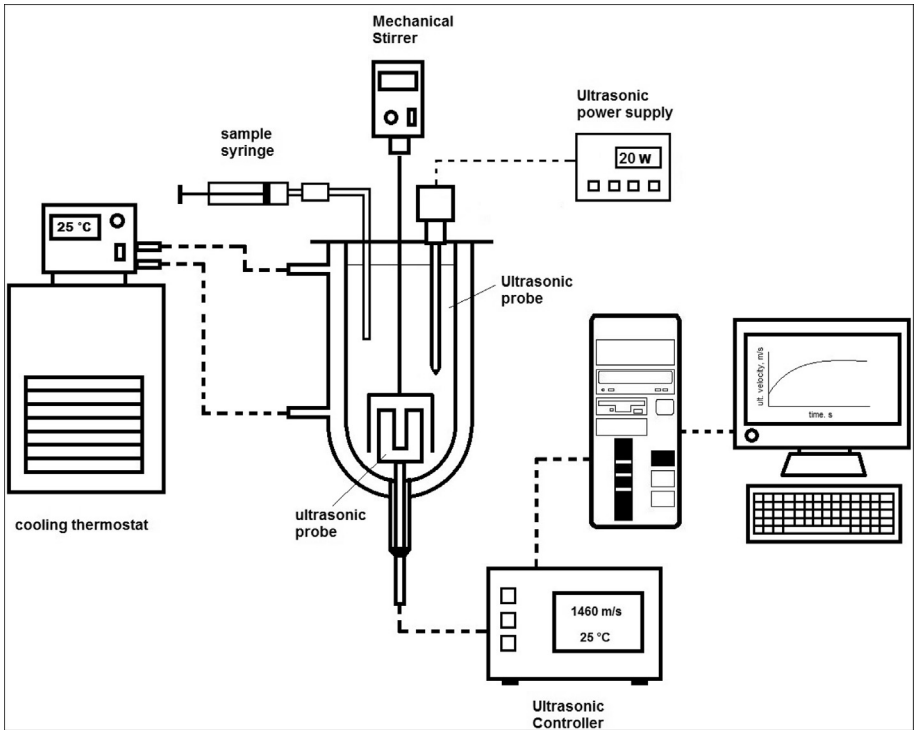

Fig. 1. Experimental setup.

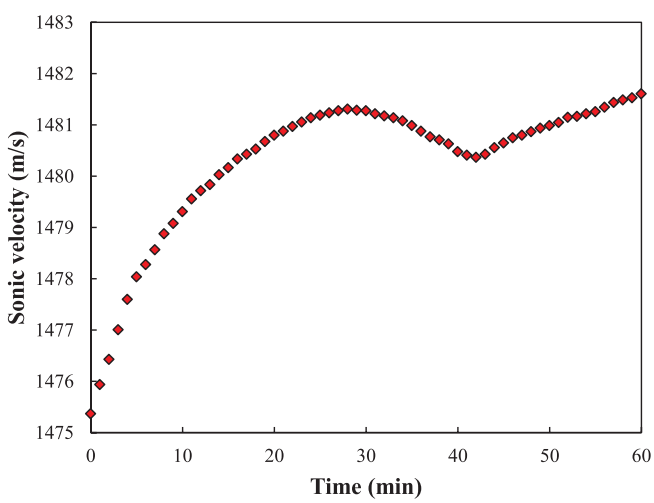

Fig. 2. The variation of ultrasonic velocity versus time during the transformation process in the absence of ultrasound.

performed to investigate the influence of ultrasonic irradiation on the particle size and distribution. However, the crystal shape factors, such as circularity, elongation, and convexity, have not been studied and there is a lack of studies focusing on particle shape which has similar importance degree. Therefore, the effects of ultrasonic irradiation on the morphology and crystal shape of glycine were investigated. The measurement of particle shape in addition to size is important in order to improve the operation conditions efficiently and reproducibly [14]. The operation conditions, such as flowing, mixing, wetting, and drying of solids, are dependent on the characteristics of the solids. In order to produce a quality product with the desirable properties, such as flowability and filterability, particle shape factors should be known. Detailed identification of crystal shape with interpretation and representation of shape parameters would provide useful information for various industries where glycine is used. Furthermore, as an important physical property of polymorphic crystals, the stability of the crystals was examined to improve the quality of the as-obtained glycine crystals and to obtain detailed information on the effects of ultrasonic irradiation on the glycine polymorphs formed. To date, no previously reported study has focused on the storage stability of glycine polymorphs precipitated using a liquid antisolvent method in the presence of ultrasonic irradiation. A better understanding of the stability of crystals is of great importance to increase the productivity of the glycine transformation process. Therefore, the purpose of this study was threefold: to determine the influence of ultrasonic irradiation on the morphology and storage stability of glycine, to describe the particle shape parameters quantifiably, and finally, to detect the impact of sonication on the time taken for the phase transformation from the β to the α form of glycine.

2. Experimental methods

Crystallization experiments were carried out in a 2-litre double-jacketed glass crystallizer in the absence and the presence of an ultrasonic source. Pure powdered α-glycine (Merck), ethanol (Merck), and triple distilled water were used in all experiments. A schematic representation of experimental setup is given in Fig. 1. The inner temperature of the crystallizer was kept constant at 25 ± 0.1 °C by means of circulating water from the thermostated bath into the crystallizer jacket. Stirring was performed with a specially designed paddle-type Teflon stirrer. The stirring rate was set to 75 rpm and kept constant. A Bandelin ultrasound homogenizer Sonopuls HD 2200 equipped with a 3 mm diameter immersion probe was used as the ultrasonic source, working at 20 kHz frequency and 200 W maximum power. The experiments were conducted at three different ultrasonic powers of 20, 30, and 40 W.

Firstly, the transformation of β-glycine to α-glycine was carried out in the absence of ultrasonic irradiation. At the beginning of the experiment, according to solubility data at 25 °C, the appropriate amount of α-glycine was dissolved in 500 ml of triple distilled water and filtered using a membrane filter with a nominal pore size of 0.45 μm. The saturated and filtered solution of α-glycine at 25 °C was placed in the crystallizer, stirred well at 75 rpm, and kept at 25 °C in media without the application of ultrasonic irradiation. After reaching thermal

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