Advanced Powder Technology 26 (2015) 309–314

Contents lists available at ScienceDirect

Advanced Powder Technology

journal homepage: www.elsevier.com/locate/apt

Original Research Paper

The effect of seeding on the phase separation phenomenon in a solidifying molten drop

A. Abouzeid *, S. Petersen, J. Ulrich

Martin Luther University Halle-Wittenberg, Center for Engineering Science, Thermal Process Engineering, D-06099 Halle, Germany

ARTICLE INFO

Article history:
Received 24 July 2014
Received in revised form 15 September 2014
Accepted 23 October 2014
Available online 7 November 2014

Keywords:
Seeding
Melt crystallization
Phase separation
Eutectic system

ABSTRACT

Considered as being one of the building blocks of the purification schemes used in industrial applications, melt crystallization is an accurate, energy saving, and a reproducible approach to the purification of materials. The separation between two mixed materials, lutrol and ibuprofen, is achievable here through tightly optimizing the highly versatile crystallization conditions. The so called, phase separation, is the end result of a melt crystallization process and its applicability on the molten drop scale is further discussed in detail. Analysis in the form of light microscopy and ultra-violet spectrometry are also introduced to quantify the final phase separation in the drops and to compare the results. The advantage of using lutrol seeding to control the separation within the molten drops is demonstrated through producing successful phase separation at the drop scale. Together with changing the cooling temperatures, seeding is also proven a successful method to produce a better phase separation and hence lower levels of impurities in the outer layer. Crystallizing the molten drops on lutrol or starch coated surfaces also enhances the quality of the phase separation by a different method, but significantly since it alters the final solidified drops geometry along with changing the cooling conditions. Finding an explanation to the different separation qualities founded on a theoretical basis for a full understanding of the process and giving the basis of a possible future optimization.

1. Introduction

As a separation technique, crystallization from the melt has many advantages such as, lower energy and running costs consumption, and that no additional substances are needed for its operation [1]. Moreover, it provides high efficiency per separation stage delivering a high degree of purity effectively [2]. Furthermore, melt crystallization is a well known industrial purification process that is known for many useful applications such as the fractionation of milk fats and sugars [3]. Many melt crystallization techniques have been already well developed, such as, crystal layer formation, suspension growth, high pressure crystallization, and distillative freezing [2]. The success of any crystallization process (from melt or solution) depends on the co-existence of several sequential energy dependent phenomena. Melt crystallization shares the same kinetics and transfer phenomena [4]. In fact, the solubility diagram that provides the major platform for understanding a solution crystallization system is only a part of the phase diagram used to study a melt crystallization system [4,5]. Nucleation, crystal growth, and mass and heat transfer are all mandatory stages in the purification, melt crystallization based, process [4]. The rate determining step, which is the heat transfer, is the slowest and the most important phenomenon in controlling most melt crystallization processes [6]. Moreover, the driving force in any crystallization process is expressed in the difference of the chemical potential between the forming crystal and the supercooled melt. The higher this difference is, the higher is the supersaturation of the crystallization system. A successful mass transport means crystal growth then the chemical potential difference decreases and as a result, the system is known to have a lower supersaturation. In a primary nucleation process, the atoms forming the crystal building blocks have to overcome a certain energy barrier, or reach a certain supersaturation, before the crystal growth is started. The progress of the crystallization process depends on an ongoing thermodynamic competition between nucleation and crystal growth to reach equilibrium [2]. This is where seeding makes an impact on the crystallization process. Seed crystals provide a surface at which new nuclei can be formed [7]. Furthermore, it provides fixed point at which crystal growth commences, and so, it is an excellent way to produce high quality

* Corresponding author. Tel.: +49 3455528 410; fax: +49 3455527358.
E-mail addresses: ahmed.abouzeid@iw.uni-halle.de (A. Abouzeid), joachim.ulrich@iw.uni-halle.de (J. Ulrich).

http://dx.doi.org/10.1016/j.apt.2014.10.013
0921-8831/

crystals reproducibly [6]. This is of course possible when seeds are introduced at a certain supersaturation level, or at the same melt temperature. This goes in hand with the success and the quality of the phase separation between the, here presented, two mixed molten components; lutrol and ibuprofen. The melt crystallization happens then at the drop scale. Moreover, the eutectic nature this system posseses, as shown in Fig. 1, gives the theoretical information necessary to ascertain the separability of the respective mixed components. The mechanism of the solid phase separation (followed in Fig. 1) along with the dynamic concentration change has been explained in detail in previous work [9]. Most importantly, the choice of the most suitable seeding material is a mandatory prerequisite upon only which a further optimization is possible. Therefore, the main objective is to process the best phase separation of the mixed molten materials through seeding. In order to quantify this phase separation, the ibuprofen concentration measurements of different crystallized drop layers are performed. This analysis is then matched with the corresponding crystallized drops' geometrical differences and the further microscopic investigation of the respective crystallized layers. This interconnected set of data will help to complete the picture of the critical phase separation phenomenon taking place within the industrial melt crystallization process and will serve as a starting point from where further optimization can take place. In addition, it will provide a fixed platform for the choice of possible future applications.

2. Materials

The active ingredient used in the melt mixture, ibuprofen, was purchased as a white fine powder, 99% purity from Caelo, Hilden, Germany. The other material used in the melt mixture, Lutrol F68 (Kolliphor P188), was purchased as a white crystalline solid powder, ⩾99% purity from Sigma Aldrich, St. Louis, United States. Corn starch that is used as a seed bed was provided as a white fine powder, with a mean particle size of 10 μm by Cargill, Krefeld, Germany.

3. Experimental methodology

Considering the sensitivity of the phase separation process, tight control of the experimental parameters has to be taken into account. This is why it is necessary to follow a specific number of steps in the right order to achieve the separation. As to be shown in the scheme displayed in Fig. 2, careful choice of the chemical

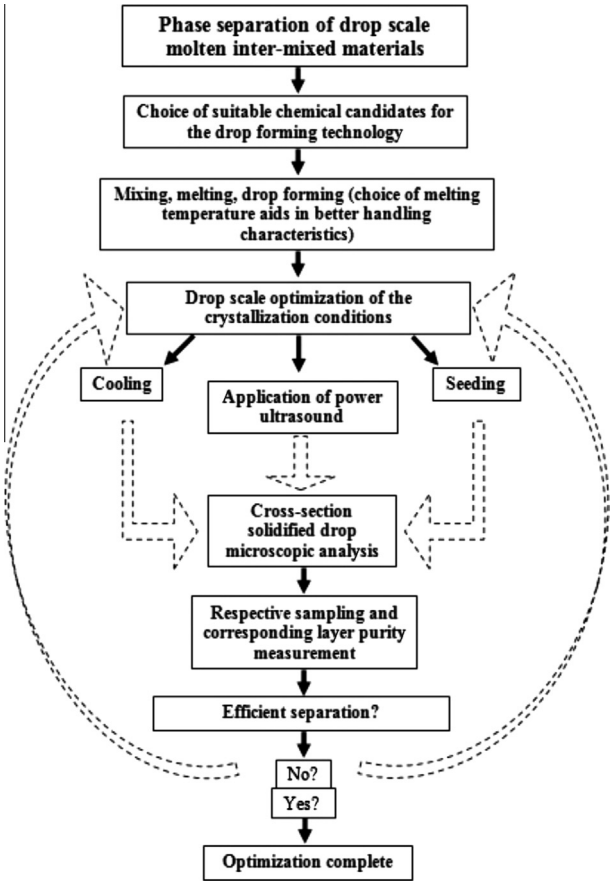

Fig. 2. A sequential step by step guide to obtaining the most optimized solidified layer phase separation on a drop scale.

candidates that make up the molten system is the first and most important step. A system satisfying a eutectic behavior such as lutrol and ibuprofen, as shown in Section 1, is sufficient to consider for the future experimental trials. Using the lutrol-ibuprofen mixture in the wt% ratio of 90:10, the drop forming method, discussed before [9], is applied to form the crystallized structures. However, during the solidification of the drop structures, several crystallization optimization options are applied. As to be seen in Fig. 2, a step

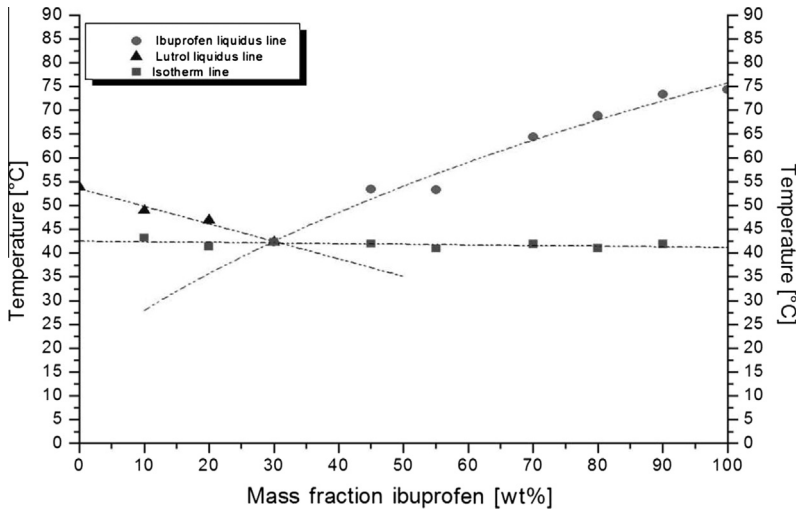

Fig. 1. A theoretical binary phase diagram highlighting the eutectic nature of the lutrol-ibuprofen mixture [8].