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Impact of particle size and surface charge density on redispersibility of spray-dried powders

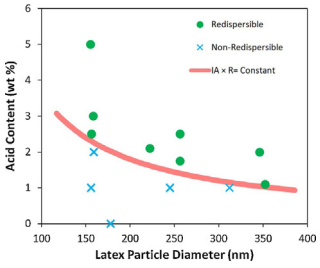
Liang Hong[a,*], Liang Chen[a], Mladen Ladika[a], Yongfu Li[b], Linda Kim-Habermehl[c], Roger Bergman[d]

[a] Formulation Science, The Dow Chemical Company, Midland, MI 48674, United States
[b] Analytical Science, The Dow Chemical Company, Midland, MI 48674, United States
[c] Dow Construction Chemicals, The Dow Chemical Company, Midland, MI 48674, United States: Currently at Xilatec, LLC
[d] Dow Emulsion Polymers, The Dow Chemical Company, Midland, MI 48674, United States; Currently at Styron Corporation

HIGHLIGHTS

- Studied impact of particle size and acid content on redispersibility.
- Larger particle can achieve redispersibility with lower acid content.
- Surface charge density is strongly correlated with redispersibility.
- DLVO analysis confirmed electrostatic repulsion is critical.
- Conclusions provide new means to control performance.

GRAPHICAL ABSTRACT



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ABSTRACT

To understand the impact of surface charge density on the redispersibility of spray dried powders, the redispersibility of styrene-butadiene (SB) powders prepared from latexes with various particle sizes and acid contents was investigated. The redispersibility of these powders showed a step function from fully redispersible to non-redispersible. The phase boundary between these two regimes is determined by the product of the latex particle size and acid content, which is strongly related to the surface charge density of the latex particle. Results indicate that to achieve fully redispersible polymer powder, a proper balance of latex particle size and surface charge is needed. For the class of SB latexes studied in this work, with a certain acid level, it is possible to predict the minimum latex particle size that enables redispersibility of its spray dried powder. This phenomenological observation is unique and it may help us to reveal the fundamentals which govern the redispersibility. To maintain the redispersibility of a spray dried powder, a colloidal stabilizer that adsorbs onto the surfaces of latex particles is required. When latexes are brought to close proximity, strong van der Waals forces may deplete the surface stabilizers, causing irreversible latex agglomeration at spray drying temperatures. A qualitative DLVO analysis was employed to study the impact of various colloidal interactions in this system, including van der Waals, electrostatic, and steric interactions. Zeta potential measurements of the mixture of SB latex and polyvinyl alcohol (PVOH, used as a colloidal stabilizer) suggest that the surface charge density plays a significant role on redispersibility, and strong electrostatic repulsive interactions are needed to overcome the attractive van der Waals interaction. Size exclusion chromatography of serum phase analysis indicated that PVOH adsorption onto latex particles did not change significantly at different acid levels, suggesting low impact of steric repulsion on redispersibility. Conclusions from this work provide additional means to control the performance of redispersible materials.

* Corresponding author. Tel.: +12156191409.
E-mail address: LHong@Dow.com (L. Hong).

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1. Introduction

Redispersible materials are materials in a powdered form that can be redispersed to produce dispersions/emulsions with a particle size essentially the same as that in the original emulsions [1]. These materials have found broad applications ranging from food and pharmaceuticals, to industrial applications such as construction materials. For example, in food applications various emulsion systems (e.g. flavor oil and milk) have been converted into powdered forms to preserve the food [2]. During consumption, these food materials are required to readily redisperse in water. In pharmaceutical products, nanoparticular formulations have attracted a lot of attention due to their high surface/volume ratio which enables higher bioavailability and better delivery efficiency [3–11]. In order to turn such formulation into a solid dosage form that is suitable for oral or nasal delivery, a drying process is necessary to convert it into a powder form (micrometer to millimeter). The resulting powder must be readily redispersible in contact with water to maintain the benefits of nano-formulation. In construction applications, redispersible powders have been one of the game-changing technologies to enable a single-pack dry mix system for cementitious formulations [12–17]. Various latex systems, i.e. vinyl acetate, acrylics, and styrene butadiene (SB) latexes were successfully used in this approach to improve cement performance.

The critical challenge in the preparation of redispersible materials is the prevention of irreversible aggregation during the drying process. The drying process has a significant impact on the properties of the resulting materials. Precise process control and a well-defined processing window are typically required for the preparation of redispersible materials. In industry, two drying processes are commonly used to prepare redispersible materials: freeze drying and spray drying. In a freeze drying process, a liquid formulation is frozen at low temperature and the continuous phase is removed at a reduced pressure. This process is less temperature sensitive than spray drying; however emulsion systems may aggregate due to solvent crystallization during the freezing process. This type of aggregation could often be reduced with the addition of cryoprotectants [18–22]. The main disadvantage of freeze drying is high process cost. In food and construction applications, spray drying is widely used for the preparation of redispersible materials due to its low cost and high throughput. During a spray drying process, a liquid formulation is atomized into micron-size droplets and the continuous liquid phase is quickly evaporated at elevated temperatures. The key challenge during this fast evaporation is to maintain colloidal stability of individual particles, especially for polymers with low glass transition temperatures (T_g) [23]. There are many interactions contributing to the stability of emulsions during this process, including van der Waals interactions, electrostatic interactions, steric interactions, etc.

To prepare spray dried redispersible powders from an emulsion formulation, one of the critical requirements is that through the process, high T_g water soluble shell materials will need to remain at the latex polymer interfaces. Several strategies have been discussed in the literature. One strategy includes dense chemical linkage of water soluble oligomers/polymers at the surfaces of the original particles in emulsion. The covalent bonding ensures that the water soluble oligomers/polymers remain at the interface throughout the drying process. This strategy was widely used for preparation of spray-dried redispersible powders from vinyl acetate-ethylene (VAE) latexes [16,24]. Another strategy is to construct core-shell geometries with a high T_g shell which could be soluble in a medium used as a continuous phase. For example, redispersible acrylic powders with a core-shell morphology were produced by a two-stage sequential emulsion polymerization process, where the high T_g shell is an alkali-soluble emulsion polymer [25]. Inorganic hybrid latex was prepared by emulsion polymerization in the presence of inorganic pigments such as TiO_2, iron oxides, and the water-redispersibility of the dried material was achieved by controlling the monomers with proper pH response [26]. These approaches were very effective in the preparation of redispersible powders from certain polymer types, but for many classes of redispersible powder precursors they are not practical due to limited selection of chemistries feasible for this process (and therefore not economically viable).

The other strategy that enables good redispersibility includes the use of a colloidal stabilizer that can physically adsorb at the surfaces of particles. In this case, the combination of electrostatic repulsions and steric repulsions needs to overcome the van der Waals interactions to maintain the stability of colloidal systems through the drying process. In order to design this complex system, it is necessary to consider many parameters such as charge density, physical adsorption, and particle size. The main challenge in this strategy is potential desorption of colloidal stabilizers that leads to particle agglomeration and formation of non-redispersible powder. One example of a colloidal stabilizer that can very effectively absorb at the surfaces of almost any class of redispersible powder precursors is poly(vinyl alcohol) (PVOH) [16]. This paper will discuss the design strategy to enable this type of redispersible powders.

To prepare this class of spray dried redispersible materials, the system has to be stable at any point of the processing conditions, and a relatively higher energy barrier will be desired to stabilize the system during the drying process. A very high stabilizer load (>60%) was used in order to provide a matrix of materials for nano-crystals in the formulation [27]. Charged surfactants with high T_g could also be employed to increase the surface charge density and hence to improve the stability. These surfactants remain at the interfaces to provide strong electrostatic repulsion, while the high T_g nature of the materials provides strong barriers at elevated temperature during the drying process [28,29].

To the best of our knowledge, previous studies of redispersible powders did not systematically investigate the effect of particle size and the surface charge density of particles on the redispersibility of spray dried powders. The focus of this article is to provide fundamental understanding of the interplay of van der Waals, electronic and steric interactions in an example carboxylated SB latex/PVOH system. The impact of latex particle size and acid content on the redispersibility was studied to understand the fundamental principles for redispersible powder technology for SB latex systems.

2. Experimental procedure

2.1. Materials

Styrene and DOWFAX™ 2A1 surfactant are Dow Chemical products. Butadiene was obtained from Airgas. Itaconic acid, acrylonitrile, and tert-dodecyl mercaptan (TDDM) were obtained from Sigma–Aldrich. Polyvinyl alcohol (Mowiol™ 4-88) was purchased from Kuraray Inc. and Kaolin clay (KaMin™ HG90) was purchased from Kamin LLC.

2.2. Latex synthesis and characterization

Carboxylated SB latexes were prepared using a previously described procedure [30–33]. Lab free radical emulsion polymerizations were carried out in stainless steel pressure vessels. Main variables in this series of latexes were the surface charge of latex particles controlled by the level of itaconic acid (IA) and the target particle size.

The particle sizes of SB latexes were collected using the Microtrac Nanotrac™ 150 particle size analyzer. The glass transition