Oct. 15, 1968 J. KOSAR ET AL 3,406,119

ENCAPSULATION

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FIG. 1

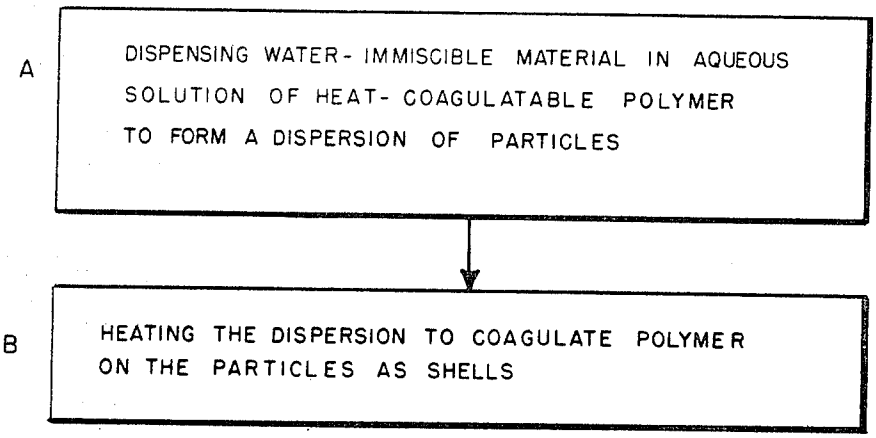
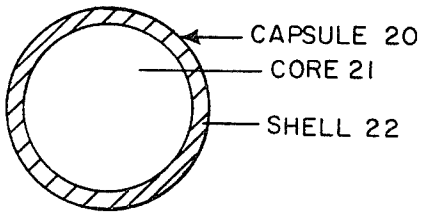


FIG. 2



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ENCAPSULATION

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ABSTRACT OF THE DISCLOSURE

The method of forming capsules by the heat coagulation of an albumin about particles or droplets of water-immiscible oil dispersed in an aqueous solution of albumin is improved by including in the oil droplets an infrared absorber and exposng the emulsion or dispersion of oil droplets to infrared radiation. The radiation absorbed by the infrared absorber is converted to heat and raises the temperature of the oil droplets sufficiently to coagulate the albumin at the surface of the droplets. In this manner, heating of the mass of aqueous vehicle and extraneous albumin coagulation are avoided.

The present invention relates to encapsulation and refers more particularly to capsules and methods for making capsules.

In many instances it is convenient to have two or more reactive agents in close proximity to each other in order to react promptly and uniformly. One prior art material provides for two layers of reactants separated by a third fusible intermediate layer. Heating the composite layers causes melting of the fusible intermediate layer, thereby allowing the two outer layers to commingle and react. The reactants are thus separated by the intermediate layer until needed. This provides convenience for the user since one of the reactants need not be held in reserve and added at the desired time.

Another method of isolating at least one of the reactants is to encapsulate the item in a shell or envelope of unreactive material. When needed, the reactant is made available by breaking the shell. One form of capsule is the developer pod used in the photographic art. These pods or capsules contain the active ingredients necessary to rapidly develop exposed photographic images. The capsules are broken physically by squeezing.

The size of the capsules presents a handicap for some applications, and therefore, capsules of fine size have been developed. These find use in such specific and varied applications as food flavorings, medicines, and photography. Some of the advantages afforded by the use of such capsules in the photographic art are isolation of reactants until desired, coating simplicity, convenience in development, indefinitely long storage life for sensitized materials, (and fine resolutions).

Previous methods for making microcapsules have used high molecular weight polymers and natural colloids for shell materials. When two colloids having different isoelectric points are used, proper adjustment of the pH produces compound capsules.

The present invention provides a novel method for making simple capsules. This method involves the discovery that certain materials can be dissolved in water at room temperature and be precipitated from the water at elevated temperatures. Thus, for example, polymer A is dissolved in water at one temperature. Water-immiscible material B is then added to the solution and dispersed to a fine size. The dispersion is then heated until the polymer coagulates around the particles or globules. The resuplting capsules may then be filtered, hardened, and dried.

Therefore, one object of the present invention is to provide capsules and novel methods for making capsules.

Another object is to provide methods for encapsulating material by means of heat, and providing capsules so formed.

Other objects of the present invention will become apparent in the course of the following specification.

The objects of the present invention may be realized by dissolving a heat-coagulatable polymer or colloid in cold water, dispersing a water-immiscible oil or solid in the solution, and then heating the dispersion to coagulate the polymer around the particles in the form of a shell.

In the drawing:

FIGURE 1 is a flow sheet of the present method; and

FIGURE 2 is a capsule of the present invention.

The process of FIGURE 1 shows two steps. In step 1A, a water-immiscible material is dispersed in an aqueous solution of heat-coagulateable polymer to form a dispersion of particles. In step 1B, the dispersion is heated to coagulate polymer on the particles as shells, thereby producing discrete capsules. Optional steps not shown in FIGURE 1 include separation of the capsules from the solution, washing the capsules, and drying the capsules.

The capsule **20** of FIGURE 2 comprises core **21** and shell **22**. Core **21** comprises water-immiscible material which may be solid or liquid. Shell **22** comprises heat-coagulated polymer completely surrounding core **21** and substantially impervious to the core material **21**.

Egg albumin coagulates at temperatures of 60–70° C., but it was found that lower temperatures are suitable for the present invention. Blood albumin was also suitable.

Albumin capsules do not form the hard agglomerates that casein capsules do and therefore offer greater advantages in coating applications where uniformity is desirable. As with casein capsules, however, use of an anti-foaming agent is helpful. Silicones have served effectively as an anti-foam agents for albumin sols.

Ratios of 7.5 parts oil to one part albumin have been successfully used. The range may vary from 1:1 to 10:1. As can be seen in the examples, coagulation by means of heat can be effected in any of several ways. The size of the capsule is determined by the size of the oil droplet in the oil-in-water emulsion. Particle sizes preferably range from 1 micron to 50 microns in diameter. The particles may be solid or liquid at the time of shell-formation.

The following examples further illustrate the present invention and are not intended to limit the scope of the inventive concept.

Example 1

10 grams of egg albumin were dissolved in 200 ml. of water at room temperature. Seven drops of a 30% silicone anti-foam (Antifoam 60) were added to the albumin sol. Added next was 20 ml. of cotton seed oil in which was dissolved 0.5 gram of an oil black dye (CI Solvent Black 12). An emulsion was made using an air-powered mechanical stirrer. An electrical hot plate was used to heat the emulsion to 70° C. and thereby form capsules. Stirring was continued throughout the heating process. The capsules were then filtered from the solution.

The capsules were spherical and ranged from 5 to 50 microns in diameter. They were placed in acetone to test the imperviousness of the capsule shell. No color was imparted to the acetone phase. The capsules were then crushed to release the dyed oil. The acetone then took on the color of the dye.

It should be evident that the presence of the oil soluble dye is not necessary for the encapsulation of material. It was used here to show that the oil could be a carrier for another material and as a marking material to demonstrate the imperviousness of the capsule shell.

Example 2

10 grams of egg albumin were dissolved in cold water and 7 drops of a 30% silicone anti-foam agent were added. 0.5 gram of carbon tetrabromide and 0.2 gram of Cyasorb IR–117 infrared radiation absorber were dissolved in 20 ml. of cotton seed oil and added to the albumin sol. An air powered stirrer was used to emulsify the oil in the aqueous phase. With continued stirring, the emulsion was exposed to an infrared lamp (reflecting drying lamp, 250 watt) in order to cause coagulation of the egg albumin as shells around the oil droplets. Samples to be inspected under the microscope were obtained with a pipette. Capsules were already present when the temperature of the aqueous phase reached 35° C. When the temperature of the aqueous phase reached 40° C., the mixture was filtered. The capsules thus obtained were washed with acetone and the filtrate mixed with diphenylamine and exposed to a UV light source (500 watt projection lamp). No color developed. The capsules were then crushed in acetone, the filtrate mixed with diphenylamine, and the mixture exposed to the light source. A blue color was formed. Carbon black was also suitable and an infrared absorber.

Example 3

10 grams of egg albumin were dissolved in 200 ml. of water. Five drops of a 30% silicone anti-foaming agent were added to the albumin sol. Two grams of carbon tetrabromide were dissolved in 20 ml. of castor oil and placed, along with the albumin sol, into a Waring Blendor (model CB–4). An oil in water emulsion was prepared by operating the blender at 17,000 r.p.m. After two minutes of operation, 500 ml. of boiling water were added to form the capsules and the stirring was continued for another 1½ minutes. The capsules were then filtered from the solution.

The capsules were spherical and about 10 microns in diameter. They were washed with benzene and the filtrate was mixed with diphenylamine and exposed to a strong light source. No color was formed. The capsules were then crushed in the benzene and the filtrate was mixed with diphenylamine and exposed to light source. A deep blue color developed as in Example 2.

It is apparent that the described examples are capable of many variations and modifications. All such variations and modifications are to be included within the scope of the present invention.

What is claimed is:

1. A method of making capsules comprising:
dispersing between about one and ten parts of a water-immiscible oil containing a carbon black infrared absorber into an aqueous solution comprising about one part of a heat-coagulatable albumin to form a dispersion of oil particles; and
exposing said dispersion to infrared radiation for a time sufficient to coagulate said albumin at the surface of said particles.

2. A method of making capsules comprising:
dispersing between about one and ten parts of a water-immiscible oil containing a carbon black infrared absorber into an aqueous solution comprising about one part of egg albumin to form a dispersion of oil particles in the aqueous vehicle; and
exposing said dispersion to infrared radiation for a time sufficient to heat said particles to the coagulation temperature of said albumin, whereby said albumin coagulates on the surface of said particles.

3. A method of making capsules comprising:
dispersing between about one and ten parts of a water-immiscible, carbon black-containing oil in an aqueous solution comprising about one part of egg albumin to form a dispersion of oil particles in the aqueous vehicle; and
exposing said dispersion to infrared radiation for a time sufficient to heat the albumin at the surface of said particles to a temperature of about 60°–70° C., thereby coagulating said albumin to form capsule walls about said particles.

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