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HYDROXYETHYLCELLULOSE PELLETS
AND PROCESS

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This invention relates to cellulose which has been graft substituted with ethylene oxide to produce hydroxyethyl-cellulose (HEC) at various levels of substitution, and has for its object the provision of an improved product of HEC, and a process of producing the product. The HEC which is treated according to the invention is a fibrous, fluffy crumb of low bulk density, and the object of the invention is the conversion of this bulky crumb to a product of much higher density without impairing its utility for further conversion into other HEC end-products. The object of the invention is achieved by pelletizing the crumb under carefully controlled conditions.

The invention is based on our discovery of a critical relationship between the bulk density, the moisture content, internal added fiber lubricant and dispersant, pre-fluffing, and the degree of fiber substitution. The pellets of HEC produced according to the invention are in an excellent form for marketing in that they may be packaged, shipped economically, stored in a relatively small space, and can be processed by the ultimate user, manufacturer of products therefrom with substantially the same ease of dispersal and solubilization as the original loose crumb.

During the course of our investigations, we encountered problems in producing a pelleted product of adequately high bulk density that would redisperse in water, return to a loose fibrous mass, without undergoing objectionable fiber consolidation, resulting in glazing or case hardening of the surface. This glazed part does not readily disperse in water and become amenable to rapid solution in dilute caustic. We have found that prefluffing with a fiber lubricant that inherently is a dispersing agent as well as lubricant is a necessary adjunct to a narrow range of moisture content for the HEC crumb and a narrow range of pellet density, within which pellets of HEC can be produced economically and contrive to have the dispersibility which is necessary for their practical utility in conversion to dispersible or soluble forms.

The preferred HEC crumb which is processed in this invention has such an amount of ethylene oxide substitution that it is essentially water-insoluble, but is soluble in dilute caustic soda and suitable for filament-forming and film-forming solutions for conversion into filaments or clear tough films of HEC, and other products. It is important that the amount of ethylene oxide substitution be such that the HEC is in a fibrous state and that the lower limit be above about 2% to enable the HEC to dissolve in dilute sodium hydroxide solution, and that the upper limit of substitution be not over about 8% by weight to prevent swelling and at least partial solubility in water.

The HEC crumb can be produced from cotton linters or wood pulp, preferably from cellulose having a uniform chain length, which is converted to alkali cellulose and reacted with ethylene oxide. In the interest of product quality and control it is important that the substitution be uniform, at any given level and to this end we prefer to form the HEC according to the process of the Mitchell et al. Patent No. 2,847,411.

It is important to obtain a high density for economy in shipping and yet have a product that can be dispersed freely in single fiber state in water as the preliminary step in processing the HEC by the ultimate user. In order to disperse or dissolve the HEC in a practical manner it is necessary that in a first stage the pellet will rapidly dis-

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tegrate uniformly in water into a fibrous or dispersed slurry, usually with the aid of some mechanical agitation. Alkali may then be added to solubilize in a second stage. When needed, a third stage of chilling will improve the quality of the solution. If the moisture content of the crumb, dispersant additive, prefluffing and the amount of compression are not carefully controlled and balanced, the pellets become glazed or case hardened and as a result the fibers do not separate during redispersal and the attempt to dissolve them in dilute caustic results in a slimy mass having very poor filterability.

There are various factors entering into the operating procedure and conditions which in the aggregate determine the commercial feasibility of producing the pellets and are expressible by the term "processibility," which encompasses such operating characteristics as power consumption during pelletizing, amount of material recycled in the pelletizing operation, actual pelletability, i.e., whether a compact pellet that can be subjected to further processing is formed, ease of drying the pellet, and the amount of material recycled during drying, and product degradation during pellet formation. The pellet characteristics important to the eventual user of the HEC pellets are expressible under the term "dispersibility." One of the most important characteristics in this category is dispersibility in water because the first step in preparing an alkali solution of HEC is dispersion of the pellet in water to a single fiber state, free of fiber clumps. Directly related to the dispersibility within realistic mixing time is the filtration potential of the resulting alkaline solution of HEC.

The problem which this invention has overcome is the production of dense pellets under conditions of processibility that are economic for the producer of the pellets, and the provision of pellets which have the chemical and physical properties of good dispersibility and, therefore, capable of practical processing by the ultimate user.

We have found that if the moisture content of the initial HEC of about 4% substitution is less than 45% the bulk density cannot be carried to practical values without glazing and impairing the dispersibility of the HEC and that if the moisture content be over 60% the HEC cannot be compressed into a stable pellet.

Below about 40% moisture content in the HEC feed stock, pellets can be formed, but processibility and subsequent dispersibility are sacrificed. As the moisture content drops from below about 40%, extremely hard pellets are formed (with excessive power consumption) which are badly glazed and the resulting pellets lack utility. The practical outside range of moisture content is from 40 to 60%, but the optimum commercial range for pelletizing HEC crumb is within the range of 50 to 55% moisture content. The bulk density of the pellets decreases with increasing moisture content of the HEC feed stock. Beyond about 60% moisture content in the HEC feed stock no pellet formation occurs, with the crumb going through the pellet mill virtually unchanged.

We have also found that the incorporation of a small amount of a fiber lubricant in the HEC fibers is conducive to easy fluffing, and uniform formation of the pullet with low power consumption as well as to ready redispersal behavior. A surfactant material such as Tetronic 304, Pluronic L-64 or Carbowax 1540 not only increases the processibility by reducing the power for pelletizing the crumb and increasing the usable bulk density but it also provides a built-in penetrating agent which increases the dispersibility of the pellet in subsequent use. The fiber lubricant enables the pellet to be compressed to a higher bulk density with less effort and without damage in the quality of the pellets (glazing). The addition of the fiber lubricant, in amounts

varying from 0.05 to 0.20% on the cellulose modifies these characteristics and increases the useful area of pelletizing. This in effect gives improved performance for any pellet produced within the aforementioned ranges.

Following adjustment of moisture and addition of the double acting fiber lubricant and dispersant it is essential to shred or fluff to provide a uniform feed of fibrous material to the die surface, such material being substantially knot-free; it is more knot-free and uniform if an additive of the type mentioned above is used.

The desired surfactant is one that not only will lubricate during shredding and pelleting but will act as a wetting agent, penetrating aid and fiber dispersant when the pellet is to be subsequently redispersed. The surfactant need also be compatible and should be beneficial to the process in which the pellet is to be used. Materials pre-eminently suitable for such use are the simple polyethylene glycols, and complex polyethylene-polypropylene glycols such as Carbonwaxes, Pluronic and Tetronics.

The density of the pellets is measured in pounds per cubic foot, and varies from the high range of from 30 to 35 for a moisture content of 40% to the low range of from 20 to 25 for a moisture content of 55%. The bulk densities for intermediate moisture contents vary proportionately between these upper and lower limits, and the addition of a fiber lubricant comparable to Pluronic L-64, permits one to uniformly increase the density about 10 to even 20 percent without detrimental effect on pellet dispersibility at each moisture level.

The ease of dispersibility can be determined by visual inspection as the pellets of the invention disintegrate or separate into loose fibers in water; good pellets become separated into a fibrous slurry in matters of seconds or minutes; poor pellets may take hours. However, a more technical measure of dispersibility of alkali soluble types is determined by measuring the filterability of a caustic soda solution of the HEC pellets following standard dispersal procedures. This is done by measuring the amount of solution in grams that will pass through a square cm. of filter area before it becomes plugged with insoluble residues.

The percentage of moisture in the HEC crumb referred to herein is determined by ordinary methods relating difference in "as is" weight to "oven dry" weight.

The following examples illustrate the production of pellets according to the invention and their comparative properties:

Example I

An HEC feed stock (4% ethylene oxide substitution) of uniform moisture content (45% moisture, 55% bone dry HEC) and containing 0.1% of Pluronic L-64 surfactant was fluffed to a uniformly divided fibrous mass by a single pass through a 12" laboratory shredder. This fluffed crumb was then fed to a laboratory model pellet mill fitted with a 1/4" diameter hole size die. The lubricated fluff was pressed into the 1/4" die orifices and cut off in about 1/2 inch lengths following extrusion. A clearance of 0.006 inch between the die and a pressure roller was employed. A uniform feed rate was maintained with accompanying uniformity of product discharge. Well-formed pellets without surface glazing were formed without over loading the pelletizer and without excessive power consumption. These pellets were dried in a rotary-type dryer to a 7% moisture content (93% bone dry HEC).

These dried pellets had a bulk density of 30 lbs. per cubic foot and a fines content of less than 1%. The product had a brightness (reflectivity) equal to that of the original crumb indicating freedom from excess heat generation due to friction. The dispersibility, as measured by time to disintegrate in water into a single fiber form, was less than one minute. The filterability of an alkaline solution (8.0% HEC/7.0% NaOH) prepared from these pellets had a plugging value of 500 grams

per sq. cm.; the filterability of the original feed stock was also 500 grams per sq. cm.

Pellets similarly formed from 8% substituted material performed best in giving a density of 30-35 pounds per cubic foot when the moisture was lowered to 45% to 48%.

When Carbowax was substituted for the Pluronic L-64 behavior was similar in pelleting but dispersion of the pellets so formed was slower and filterability was 450 rather than 500.

Example II

The same conditions employed in Example I were used except the crumb feed stock did not contain any added Pluronic L-64 surfactant. Power consumption of the pellet mill was 1/3 higher than that encountered in Example I. Bulk density of the pelletized product was 32 pounds per cu. ft. Temperature of the pellet die during the operation was definitely higher than Example I and the exiting product temperature was consequently higher also; some pellets showed signs of scorching. Dispersibility in water of these pellets was definitely poorer than those of Example I, requiring about 3 minutes high speed mixing in order to achieve a complete fiber dispersion. The filterability of a standard 8.0/7.0 solution was also somewhat lowered, giving a plugging value of 400 grams per square cm.

Example III

HEC crumb of 4% ethylene oxide substitution and 60% moisture content (40% bone dry HEC) containing no Pluronic L-64 surfactant was fed to the pellet mill after fluffing, at the same rate as in Example I. The fibrous material went through the die without any appreciable compaction and little or no pellet formation occurred. The material coming from the mill was still in crumb form with only a slightly higher density (12 pounds/cu. ft.) than the feed stock (10 pounds/cu. ft.). The material from the pellet mill was not much different from the feed stock and was basically incapable of being pelletized.

Similar stock at 58% moisture pelletized to a bulk density of 20-22 pounds per cu. ft.

Example IV

Pre-fluffed HEC crumb (4% ethylene oxide substitution level, at a moisture content of 45%) containing no surfactant was fed to the pellet mill at same rate as Example I. The power to operate the pellet mill was more than twice as much as that required in Example I. Considerable heating within the die chamber occurred during the run and the product emerging from the mill was at a considerably higher temperature than the feed stock. Severe glazing occurred causing some pellets to appear almost translucent. Bulk density was high (38 pounds per cu. ft.), but pellet dispersibility was extremely poor with most pellets remaining partially intact even after 20 minutes' mixing in water. The surface of the pellets, in addition to being glazed, showed an increase in color due to the high temperature developed in the pelletizing operation. This was reflected in a lower level of brightness in the pelletized product. In addition to the poor dispersibility exhibited by these pellets, a much longer time was required for mixing a standard alkali solution. The deleterious effect upon product quality was also apparent from the filterability of the standard alkali solution (100 grams per sq. cm.).

Similar material, but containing 0.1% Pluronic L-64 and 50% moisture pelletized to a bulk density of 32 pounds per cu. ft. and had excellent dispersibility.

We claim:

1. An alkali soluble hydroxyethylcellulose pellet product formed of hydroxyethylcellulose fibrous crumb having from 2% to 8% of ethylene oxide substitution and compressed to a bulk density which varies from the high

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range of from 30 to 35 when the moisture content is 40% to the low range of from 20 to 25 when the moisture content is 58%, said pellets being free of surface glazing and readily dispersible in water.

2. An hydroxyethylcellulose pellet product defined in claim 1 which comprises a fiber lubricant and dispersant added to the crumb prior to shredding and pelleting and the bulk density is increased from 10% to 20%.

3. An hydroxyethylcellulose pellet product according to claim 1 in which the pellets are rod-like extrusions having diameters varying from $\frac{1}{8}$ to $\frac{3}{8}$ inch and length of about $\frac{1}{2}$ -1 inch.

4. An hydroxyethylcellulose pellet as defined in claim 1 in which the bulk density varies from the high range of from 32 to 34 when the moisture content is about 50% to the low range of from 20 to 22 when the moisture content is about 55%.

5. The process for producing hydroxyethylcellulose pellets which comprises incorporating a fiber lubricant having dispersing properties in fibrous hydroxyethylcellulose crumb having from 2% to 8% of ethylene oxide substitution, compressing the crumb to a bulk density

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varying from 33 to 42 when the moisture content is 40% and extending to from 22 to 30 when the moisture content is 58% to produce pellets free of surface glazing which are dispersible in water.

6. The process as defined in claim 5 which comprises forcing the crumb containing the dispersant additive through a die to produce rod-like pellets varying in diameter from $\frac{1}{8}$ to $\frac{3}{8}$ and about $\frac{1}{2}$ -1 inch in length.

7. The process as defined in claim 5 which comprises fluffing the crumb compressing the crumb to a high range of bulk density varying from 32 to 34 when the moisture content is 50% to a low range of from 28 to 30 when the moisture content is 55%.

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