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Dry particle coating using magnetically assisted impaction coating: modification of surface properties and optimization of system and operating parameters

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Abstract

The feasibility of using the magnetically assisted impaction coating (MAIC) device to coat fine silica guest particles onto the surface of larger cornstarch and cellulose host particles was examined. This was done to simultaneously improve the flowability of the host particles, as well as reduce their hydrophilicity, making them more suitable for use in foods and pharmaceuticals.

The success of coating achieved by MAIC depends on the degree of ''fluidization'' of the host/guest particle system caused by the motion of the magnetic particles. To better understand the factors influencing this fluidizing behavior, several critical system and operating parameters were investigated. This was done using a model system consisting of PMMA host particles and alumina guest particles. The system parameters examined were magnetic particle size, mass ratio of magnetic particles to powder (host and guest particles) and guest particle size. The operating parameters were processing time, current (or voltage) and frequency. In addition to varying these parameters, enhanced image processing was used to measure the motion of the magnetic particles in order to study its effect on coating efficiency. The magnetic particles were observed to have both rotational and translational motion. © 2000 Elsevier Science S.A. All rights reserved.

Keywords: MAIC; Cornstarch; Cellulose; Magnetic particles; Dry coating

1. Introduction

Surface modification to alter the properties of powders (such as flowability, wettability, flavor, color, etc.) is very important to many industries [1]. Typically, surface modification of particles to form a barrier or film between the particle and its environment has been done by wet coating methods such as pan coaters and a variety of fluidized bed coaters or by wet chemistry-based techniques such as coacervation, interfacial polymerization and urea/formaldehyde deposition. However, wet coating methods have become less desirable because of environmental concerns over the resulting waste solutions and possible VOC emissions. Dry particle coating, which directly attaches fine materials (guest particles) onto the surface of larger core particles (host particles) by mechanical means without using any solvents, binders or even water, is a promising alternative approach [2]. Apart from forming a barrier as in wet coating, dry particle coating can be used for making significant changes in the functionality or the properties of the original host particles, and thus creating engineered particulates.

Several dry coating methods have been developed [3–5]. These methods generally allow for the application of high shearing stresses or high impaction forces to achieve coating. The strong mechanical forces and the accompanying heat generated can cause layering and even embedding of the guest particles onto the surface of the host particles. However, some dry coating methods may not be appropriate for certain applications if there are special constraints such as allowable temperature range, material hardness and cost factors that need to be considered. Many food and pharmaceutical ingredients, being organic and relatively

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soft, are very sensitive to heat and can quite easily be deformed by severe mechanical forces. Hence, soft coating methods that can attach the guest particles onto the host particles with a minimum degradation of particle size, shape and composition caused by the build up of heat, are the better candidates for such applications. The magnetically assisted impaction coating device (henceforth called MAIC) can coat soft organic host and guest particles without causing major changes in the material shape and size [6]. Although there is some heat generated on a microscale due to the collisions of particles, there is negligible heat generation on a macroscopic level and hence no increase in temperature of the material during processing in the MAIC. This is an added advantage when dealing with temperature sensitive powders such as pharmaceuticals.

Certain materials, such as cornstarch and cellulose, are important ingredients in food and pharmaceutical products. Cornstarch is frequently used as a food-thickening agent and as an inactive component of pharmaceuticals [7]. Cellulose is also a commonly used component in the composition of several processed foods. However, their cohesiveness (especially cornstarch) and hydrophilicity are undesirable. For example, cohesiveness causes problems in handling and hydrophilicity limits shelf life due to premature biodegradation or the growth of molds and other microorganisms on the surface. In addition, materials can become sticky due to exposure to humid environments and there can be a loss of mechanical properties due to plasticization [8].

Fine powders are often used as flow aids by simply mixing the fines with the core material [9,10]. However, simple mixing cannot change the materials' hydrophilicity. Hence, the ability to modify these materials to simultaneously obtain better flow properties and also make them hydrophobic is not only advantageous, but also necessary to create composite materials with unique functionality.

Cornstarch is composed of two basic types of polymers — amylose and amylopectin. Amylose is a linear polymer where each of the monometric units (except the terminal units) contains one primary and two secondary hydroxyl groups. The hydroxyl groups impart hydrophilic properties to the polymer, which leads to an affinity for moisture, i.e., the hydroxyl groups act as sites for adsorbing moisture. Cornstarch has been chemically modified by introducing hydrophobic ester groups at low levels of substitution when it is desirable to impart improved flow properties to the powder [11]. This modified cornstarch is a free flowing powder, which is also remarkably water repellant.

Cellulose fibers, similar to cornstarch, also contain hydroxyl groups [11] on their surfaces that are responsible for moisture absorption. To reduce the hydrophilicity of these materials, it is necessary to remove some of these hydroxyl groups either by chemical esterification or by some other surface modification process. In a series of publications, Senna [12–15] has convincingly shown that dissimilar metallic species, notably complex oxide powders, can be cross-linked by oxygen using a soft-mechanochemical process (mechanical stressing of the powders) by proton transfer through OH groups, and subsequent electron transfer. The mechanochemical reaction is not restricted to inorganic materials, but is also applicable to complex formation between inorganic–organic or organic–organic materials and can be accomplished using easily available machines for grinding or comminution. Therefore, it is quite possible that particle processing by MAIC will promote a mechanochemical reaction between the almost neutral hydroxyl groups of cornstarch/cellulose and another material's more acidic hydrophilic OH groups by oxygen linkages and the removal of water molecules to form hydrophobic groups (see Fig. 9). Silica is one such candidate that can react with cornstarch/cellulose as it possesses acidic hydrophilic silanol groups (–Si(OH)–) on its surface [16].

In addition, the fine silica particles coated onto the surface of the cornstarch can act as a flow aid by reducing the Van der Waals forces between the larger host particles [17]. We have used the MAIC device [18,19] to coat fine silica particles onto the surface of cornstarch to simultaneously modify its flowability and hydrophilicity. We have also coated silica particles onto the surface of cellulose (fiber-like particles with an aspect ratio of about 4–5) for the same purpose.

Cornstarch and cellulose were chosen as host particles for several reasons. Firstly, both materials are organic and the ability to process these materials in the MAIC without appreciably altering their properties based on shape and size will be a significant advantage. Secondly, as mentioned before, both materials are widely used commercially, but their poor flow and their high moisture absorption surfaces limit their applications. Hence, these materials were chosen to examine the feasibility of surface enhancement by MAIC as a way to broaden their usage.

There are several critical system and operating parameters affecting the coating performance of the MAIC device. Once the host and guest particles are specified, the key system parameters are magnetic particle size and magnetic particle to powder (host and guest particles) mass ratio. The major operating parameters are frequency, current (or voltage) and processing time. To study the effects of these parameters on the coating efficiency, a model system consisting of 200 μm spherical PMMA host particles and several sizes of alumina guests particles was chosen and experiments were conducted which systematically varied all of the parameters mentioned above. PMMA was chosen as the host material in our ''model system'' because the particles are both spherical and smooth, and therefore the added variable of surface irregularities is eliminated in evaluating coating performance. In addition, several previous investigators [19] have used the PMMA (host particles), coated with alumina (guest particles) system as an advantage for dry particle coating studies, because of the favorable difference in hardness between the two materials (soft-hard spheres).

The motion of the magnetic particles was also examined using a high-speed digital camera. This was done to study the effect of the motion of the magnetic particles on the quality of the surface coverage obtained. Based on the results of the optimization and the magnetic particle motion studies, a preliminary mechanism of coating by MAIC is proposed.

2. Experimental

A schematic of the MAIC device (batch mode) is shown in Fig. 1. With some modification, the device can also be operated continuously and has been scaled-up to process up to 800 lb/h [20]. The device used in this study operates in a batch mode. A weighed amount of host and guest particles are placed into a processing vessel (200-ml glass bottle). The mass percentage of guest particles used in an experiment is usually calculated based on the assumption of 100% surface coverage of the host particles with a monolayer of guest particles. However, when coating with silica, only 1% by mass of silica is used to conform to Food and Drug Administration (FDA) standards. Therefore, only very discrete coatings are obtained.

A measured mass of magnetic particles is also placed in the processing vessel. The magnetic particles are made of barium ferrite and coated with polyurethane to prevent contamination of the coated particles. An external magnetic field is created using a series of electromagnets surrounding the processing vessel. When a magnetic field is present, the magnetic particles are agitated and move furiously inside the vessel, resembling a fluidized bed system. These agitated magnetic particles then impart energy to the host and guest particles, causing collisions and allowing coating to be achieved by means of impaction or peening of the guest particles onto the host particles.

The experimental study was divided into two parts. Firstly, the feasibility of the MAIC to modify the surface properties of cornstarch and cellulose host particles by coating with silica guest particles was studied. The physical properties of the materials and the experimental operating parameters are given in Tables 1 and 2, respectively. Secondly, the critical parameters affecting the coating efficiency of the MAIC device were studied using the model system of PMMA as host particles and alumina as guest particles. The physical properties of PMMA and alumina are also given in Table 1. The variations in system and operating parameters investigated in the optimization study are given in Table 3.

In the first study, the coated cornstarch and cellulose products were examined with a scanning electron microscope (SEM) to study the surface morphology and particle shape after coating. Energy dispersive X-ray spectroscopy (EDX) was used to study the surface composition of the coated products. In some cases, an API Aerosizer was also used to measure the particle size distribution and mean particle size of the coated sample as a function of processing time. The flowability of the products was analyzed by measuring the angle of repose (AOR) using a fixed base method. Wettability tests were conducted by using the penetration rate method [21,22] to evaluate the hydrophilicity of the coated products. In the penetration rate method,

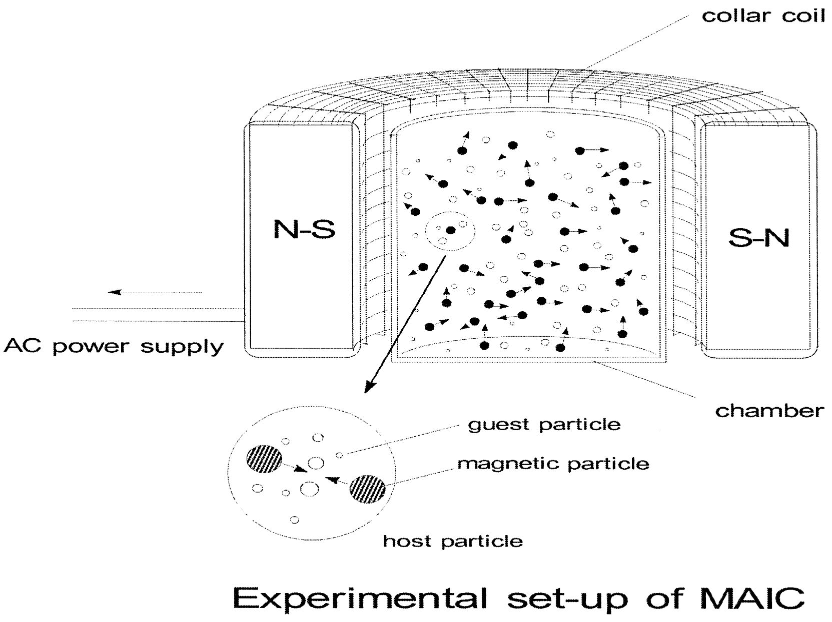

Fig. 1. Schematic of the MAIC process.

Table 1
Physical properties of materials

System 1	
Host particle size: cornstarch (μm)	15
Guest particle size: silica (μm)	0.3
Agglomerate guest particle size: silica (μm)	35
System 2	
Host particle size: cellulose (μm)	Aspect ratio of 4–5 (180/40 μm)
Guest particle size: silica (μm)	0.3
System 3	
Host particle size: PMMA (μm)	200
Guest particle size: alumina (μm)	0.05, 0.2, 0.4 and 1.0

powder is filled into a column, and the change in the amount of liquid penetrating into the powder layer is measured. The powder was compacted to the same voidage by controlling the mass of powder used and the height to which the powder was compressed. The controlled voidage for both materials studied was 0.35. Fourier transform infrared spectroscopy (FTIR) was also used to study the changes in O–H groups of the samples before and after coating to help interpret the hydrophilicity results.

In the second study, a series of experiments were conducted to obtain optimum operating parameters. The parameters examined were processing time, current, frequency, magnetic particle to powder mass ratio, magnetic particle size and guest particle size. The coated products obtained were evaluated by several characterization techniques. A SEM was used to examine the surface morphology of the coated PMMA products. A statistical image analysis was conducted on each SEM micrograph for different operating conditions to obtain the percentage of surface area covered by guest particles.

The speed and the behavior of the magnetic particles during processing in the MAIC device were obtained by using a Kodak EktaPro1000 high-speed digital camera with an intensified imager (capable of recording at up to 1000 frames per second) to capture the motion. The movement of magnetic particles was examined as a function of the frequency of the external field. By combining the results of the speed and the parameter optimization studies, the surface coverage as a function of the magnetic particle speed was obtained.

Table 2
Operating parameters for the surface modification study

Primary particle size: magnet (mm)	1.4
Mass susceptibility of magnetic particle (emu/g)	24.66
Mass ratio of magnets/powder	1, 2
Mass ratio of guest/host	1%
Average magnetic field strength (mT)	40
Processing time (min)	5, 10, 20
Volume of processing chamber (ml)	200

Table 3
Variations in the system and operating parameters used for the optimization study

Magnetic particle to powder mass ratio	0.5, 0.75, 1.0, 1.5, 2.0
Magnetic particle size (μm)	180, 800, 2700
Processing time (min)	1, 3, 5, 7, 10
Current (A)	1, 2, 3, 4, 5
Frequency (Hz)	45 to 110

3. Results and discussion

3.1. Study I: modification of the surface properties of cornstarch and cellulose

Experiments were conducted to evaluate the effectiveness of the MAIC device in modifying the surface properties of cornstarch and cellulose. The coated products were characterized by different methods as discussed below.

3.1.1. Surface morphology and surface elemental mapping

SEM micrographs of cornstarch before and after coating (10 min processing time) are shown in Fig. 2a and b, respectively. An elemental mapping of silicon on the surface of the 10-min coated product is shown in Fig. 2c. After processing in the MAIC device, small silica agglomerates (≤ 3 μm) are observed on the surface of cornstarch. Due to the very small size of the primary particles of silica, these particles have a natural tendency to form very large agglomerates, approximately 35 μm in diameter as observed by SEM. The absence of these large agglomerates after coating suggests that they are broken up into smaller primary sizes (de-agglomeration) during the MAIC process. This observation is in contrast to our previous study [23] using large PMMA particles (mean size of 200 μm) as the host particles and silica as the guest particles. In that study, large silica agglomerates were still seen on the surface of the PMMA particles after being subject to various processing times in the MAIC device.

The difference in these two systems shows the importance of the host to guest particle size ratio in the coating mechanism. When the primary guest particles are in the sub-micron range, the attraction forces (Van der Waals, electrostatic, etc.) among the primary particles are relatively strong and require larger forces to separate them. Smaller host particles can obtain larger velocities than larger host particles from collisions with the magnetic particles, resulting in higher forces of impaction, sufficient to break the agglomerated guest particle structure. In addition to the de-agglomeration of the guest particles, it should be emphasized that cornstarch still maintains its

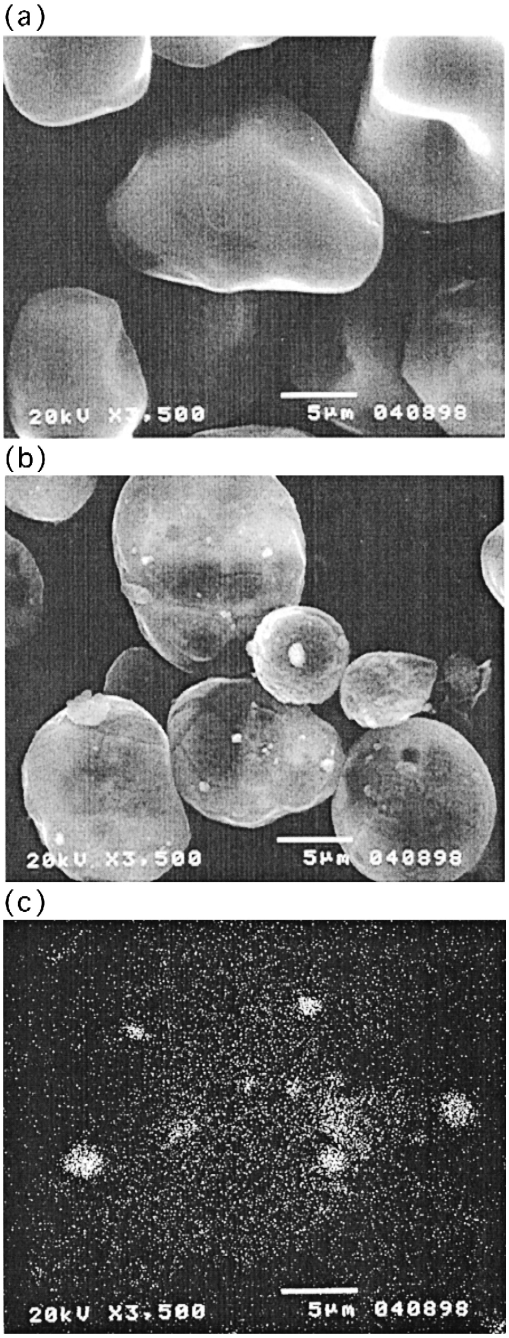

Fig. 2. SEM micrographs of (a) unmodified cornstarch, (b) cornstarch processed for 5 min and (c) EDX mapping of silicon on the surface of modified cornstarch.

disc-like shape after processing. This is also a unique feature of the MAIC device, in that after processing, soft organic materials still maintain almost their original shape and size. The elemental mapping (Fig. 2c) confirms the small particles on the surface of cornstarch as silica.

A SEM micrograph of unmodified cellulose is shown in Fig. 3a. The particles are fiber-like with an aspect ratio of 4–5. SEM micrographs showing the surface morphology of cellulose coated with silica for processing times of 5 and 10 min are shown in Fig. 3b and c, respectively. Observation of cellulose coated products also showed the presence of silica on the surface (Fig. 3b and c). As the processing time increased from 5 to 10 min, the area of surface coverage also increased. At a higher processing time of 20 min, there was no visible change in the surface coverage of cellulose as compared to the 10-min cellulose product.

3.1.2. Flowability

The AOR is a commonly used index for flowability, hence it is used in this work to evaluate the coating

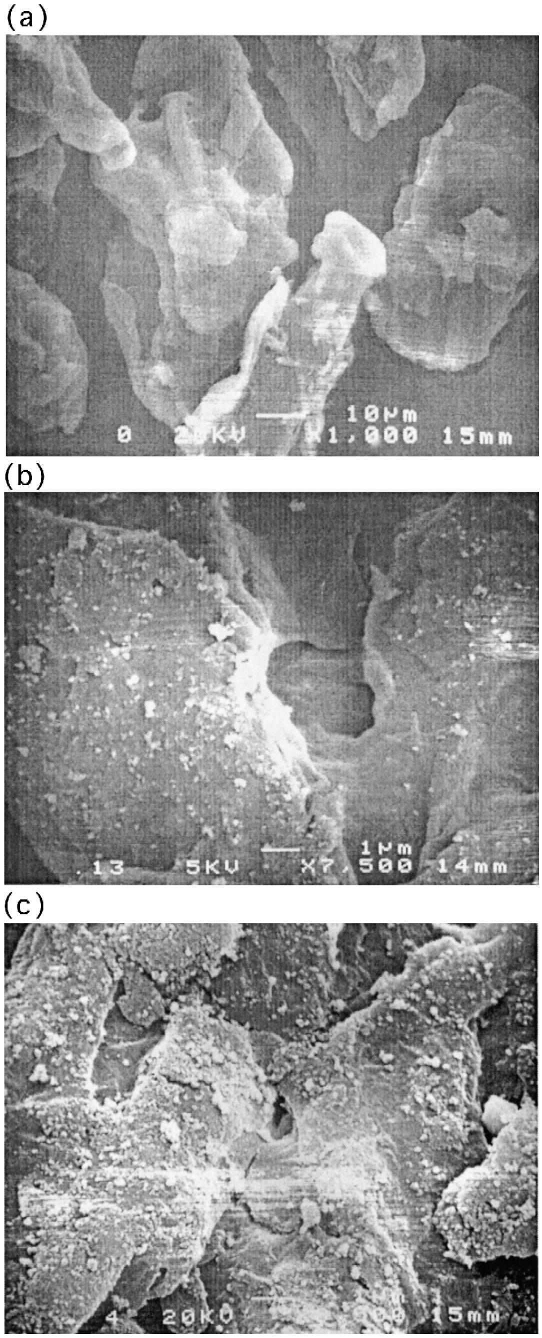

Fig. 3. SEM micrographs of (a) unmodified cellulose, (b) cellulose coated with silica for 5 min and (c) cellulose coated with silica for 10 min.

effectiveness in terms of improving flow properties. The results for the AOR of cornstarch products for two different magnetic particle to powder mass ratios are shown in Fig. 4. The value shown for each processing time is an average of four AOR measurements obtained by a digital camera. While the humidity at which the tests were conducted was not controlled, the experiments were all performed on the same day and under the same conditions (all samples were dried in an oven before the experiments), therefore minimizing the error associated with changes in humidity. The AOR for untreated cornstarch is approximately 59°, as shown in the figure. The AOR decreases as processing time increases for both magnetic particles to powder mass ratio, indicating improvement in the flow of cornstarch due to a surface coating of silica. For example, it is 54° after 5 min for a magnetic particle to powder mass ratio of 1, further decreasing to about 47° after 20 min. When the magnetic particle to powder ratio increases, the collision frequency of the magnets and the powders also increases. Thus, the same coating conditions are achieved at shorter processing times as seen in Fig. 4. This is evidenced by the lower AOR at 5 min for the larger magnetic particle mass ratio of 2.

Craik [9] tested several materials, including silica, as a flow aid by mixing them with cornstarch. However, the addition of silica did not improve the flowability. In the study done by Craik [9], a fixed base AOR method similar to that used in this investigation was utilized to measure the AOR. In that study, large silica aggregates were observed. The presence of the large aggregates indicates that simple mixing cannot break the agglomerates due to the large attractive forces between the individual silica particles as discussed above. In contrast, in the MAIC, it is possible to coat fine silica particles onto the cornstarch surface by first de-agglomerating the silica. The difference between these studies shows the importance of the size of guest particles in improving flowability.

The results for the AOR of the coated cellulose products as a function of processing time are shown in Fig. 5. As was found with cornstarch, a surface coating of silica increased the flowability of the cellulose for all processing times investigated. However, a processing time of 10 min yielded the best flowing cellulose. Unlike the coated cornstarch product, further increases in the processing time did not improve the flowability. In fact, the flowability of the cellulose decreased after a processing time of 10 min, suggesting that longer processing times may actually degrade the surface coating or the cellulose fiber host particles, perhaps because of their relatively high aspect ratio. While we could not determine whether the surface coating degrades, Fig. 6 shows that for all of the processing times examined, there was a reduction in the mean particle size of the fibers. This indicated that the MAIC process is causing significant attrition of the high aspect ratio cellulose fibers. Each value given in the figure is the average of five measurements. Thus, it appears that attrition causes a decrease in flowability for a processing time greater than 10 min.

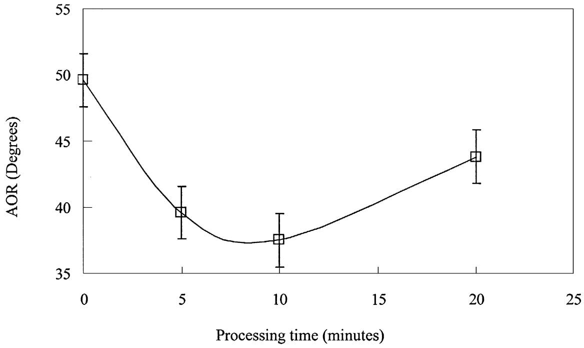

Fig. 5. Flowability of cellulose as a function of processing time for a magnetic particle to powder mass ratio of 2.

The ability to improve the flowability of a material by coating with fine particles has been discussed by Mei et al. [17]. The authors used a discrete element simulation of powder flows between a moving and stationary plate to examine the effect of fine coatings on the surface of a larger substrate. They also developed an extended JKR (Johnson–Kendall–Roberts) particle contact model to include the effect of particle coating on the force–displacement relationship due to surface energy and elastic deformation. Their results indicated that the cohesion force

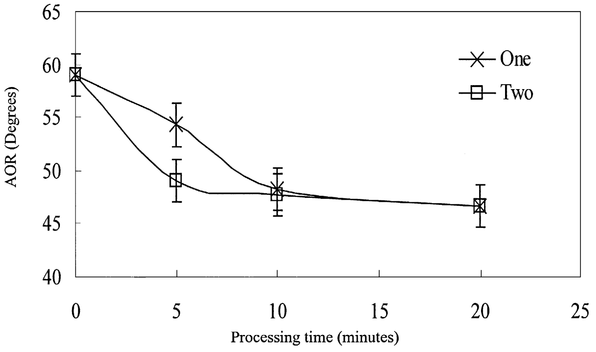

Fig. 4. Flowability of cornstarch as a function of processing time for two different magnetic particle to powder mass ratios.

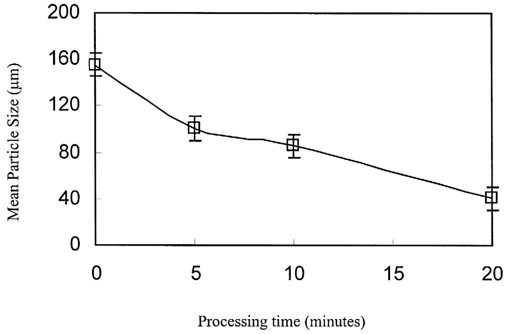

Fig. 6. Mean particle size of cellulose as a function of processing time.

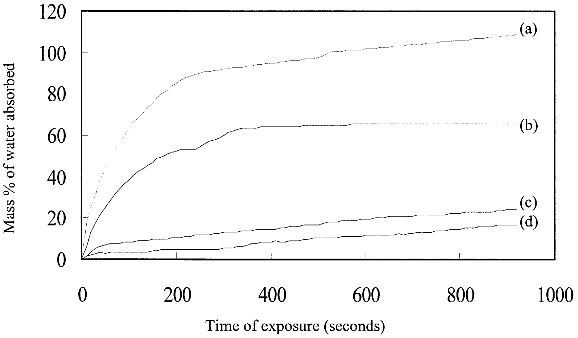

Fig. 7. Wettability study of (a) silica, (b) uncoated cornstarch and cornstarch processed for (c) 5 and (d) 20 min.

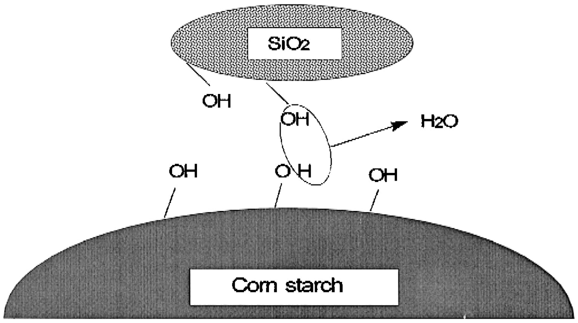

Fig. 9. Mechanochemistry effect occurring on the surface of cornstarch to form hydrophobic groups.

between two primary particles in the presence of a fine coating is directly proportional to the size ratio of the coating particles to the host powder particle and results in drastic reduction in the cohesion forces. This argument has been adapted to explain the improve flowability of cornstarch and cellulose in the presence of a discrete coating of silica as obtained by MAIC. The fine silica particles reduce the Van der Waals attraction force between the host particles, making them flow more easily. The number of guest particles on the surface of the host particles have only a minor effect on the flowability once the cohesion force is reduced by one or more coating particles. Hence, even with a very discrete coating on the surface of the host particle, there is a significant improvement in the flowability of the material.

3.1.3. Hydrophilicity

To measure the changes in the hydrophilicity of the surface of cornstarch and cellulose, wettability tests of the coated product were conducted. This was done using a rate penetration method, whereby a column was filled with the powder and a load applied to compact and control the voidage of the powder. The voidage was set at a value of 0.35 for both materials. Then, to investigate the mass percentage of water being absorbed, the column was gently submerged into a dish of water. The weight change of the water in the dish (that absorbed by the sample) was measured as a function of time.

The water absorption results for the coated cornstarch products (products from the magnetic particle to powder mass ratio of 2 were used for this test), together with silica alone and cornstarch alone, are shown in Fig. 7. Silica is very hydrophilic in nature and absorbed approximately 110% of its weight in water, during an exposure time of 5 min as shown in Fig. 7. Unmodified cornstarch is also hydrophilic and absorbed about 60% of its weight in water for the same 5-min period of exposure. For cornstarch processed in the MAIC for a time of 5 min, it is observed that the water absorption is reduced to about 28% of the weight of cornstarch using the rate penetration test. A larger processing time of 20 min further reduced the mass percentage of water absorbed by the cornstarch to about 18%.

For the coated cellulose products, water absorption capacities of unmodified cellulose and coated products at 5, 10 and 20 min processing times are shown in Fig. 8. The results were also obtained by the rate penetration method. The water absorption capacities decreased for all the investigated processing times. The absorption capacity at 10 min, however, was lower than that at 20 min, again indicating deterioration of the surface coating or attrition of the host particles with increased processing time as was observed in the flowability study. This behavior was also observed in a previous study [23] for PMMA coated with

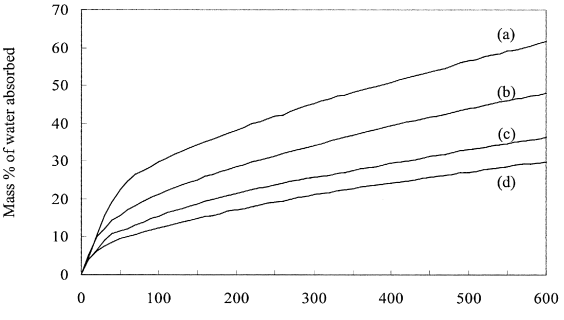

Fig. 8. Wettability study of (a) uncoated cellulose and cellulose processed for (b) 20, (c) 5 and (d) 10 min.

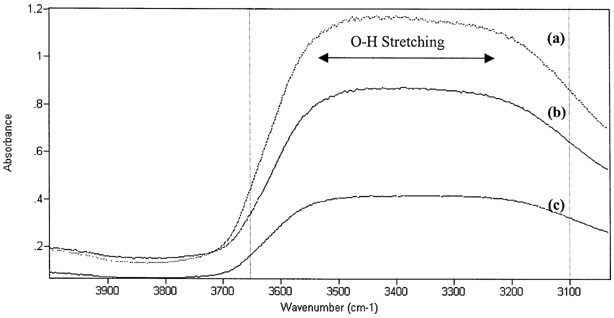

Fig. 10. IR absorption caused by O–H stretching vibrations for (a) uncoated cornstarch, (b) cornstarch coated for 10 min and (c) cornstarch coated for 20 min.

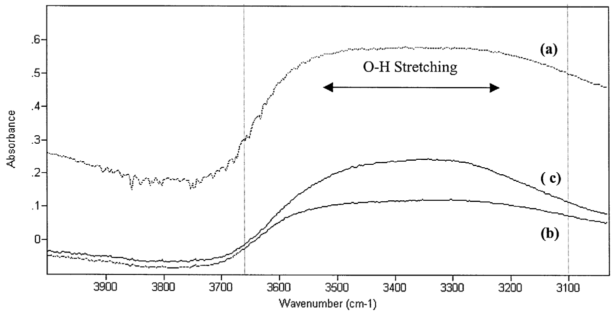

Fig. 11. IR absorption caused by O–H stretching vibrations for (a) uncoated cellulose, (b) cellulose coated for 10 min and (c) cellulose coated for 20 min.

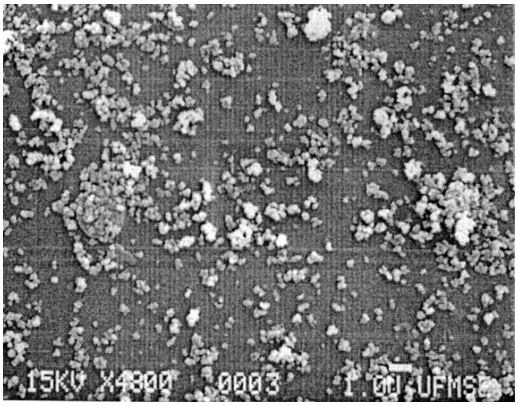

Fig. 13. SEM micrograph of PMMA coated with alumina for a processing time of 1 min.

alumina in the MAIC process. The surface coating condition of PMMA deteriorated with increased processing times. It is conjectured that the continuous impaction and collisions of the magnets onto the host particles can destroy the coating integrity as well as the size and shape of the material, after optimum processing conditions are reached.

A reduction in hydrophilicity is believed to result from the reaction of the acidic silanol groups (–Si(OH)–) on the silica surface and the almost neutral hydroxyl groups (–(OH)–) on the cornstarch/cellulose surfaces, to form hydrophobic groups (Fig. 9) by releasing water molecules. The high mechanical forces arising from the particle collisions during ‘‘fluidization’’ enhances the reaction by mechanochemistry [24]. In order to confirm the hypothesis of a mechanochemical mechanism for the reduction in water absorption as described above, we used FTIR to measure the changes in OH groups before and after coating.

Some preliminary FTIR results are presented in Figs. 10 and 11, for cornstarch and cellulose, respectively. In Fig. 10, there is a reduction in the absorption caused by O–H stretching vibrations between the wavenumber of 3100 and 3650 cm^{-1} for the coated samples as compared to the uncoated cornstarch, indicating a reduction in the O–H groups on the surface of the samples. There is also a significant reduction in absorption due to O–H stretching vibrations with an increase in processing time from 10 to 20 min, again within the above mentioned wavenumber range. Fig. 11 (for cellulose) shows that the least absorption caused by O–H stretching vibrations is obtained for the 10-min processed sample. This appears to confirm the hydrophilicity studies for cellulose presented in Fig. 8. This increase in absorption of the 20-min coated sample can be attributed to the significant reduction in the particle size of the cellulose fibers, making more O–H sites available for absorption due to increased surface area. The FTIR results lend credence to the hypothesis that MAIC processing caused a mechanochemical surface reaction between the corn starch/cellulose host particles and the silica guest particles. This represents a new contribution, as for the first time some evidence is presented that MAIC is capable of causing mechanochemical effects.

3.2. Study II: optimization of the critical parameters affecting the coating performance of MAIC

For the optimization study, each system and operating parameter was individually varied using the model system PMMA (host particle) and alumina (guest particle). Sur-

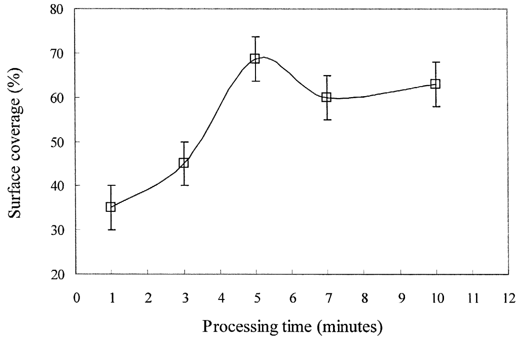

Fig. 12. Surface coverage as a function of processing time (PMMA/$\mathrm{Al_2O_3}$).

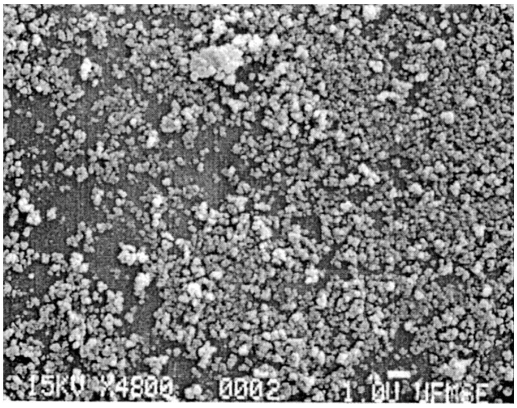

Fig. 14. SEM micrograph of PMMA coated with alumina for a processing time of 5 min.

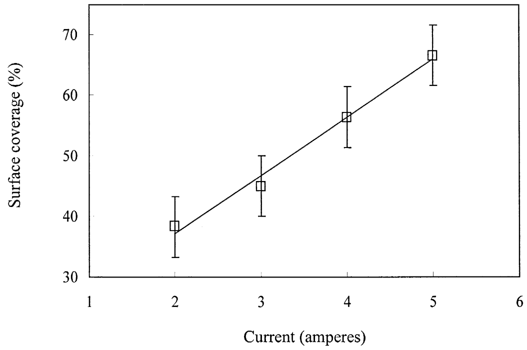

Fig. 15. Surface coverage of the coated products as a function of current at a fixed frequency of 45 Hz.

face morphology micrographs for all the coated products were obtained using a SEM. The micrographs were quantitatively analyzed by a statistical technique done by individually counting the number of guest particles on the surface of the host particles to obtain the percentage of surface covered. The results for each parameter studied are presented in the following sections.

3.2.1. Processing time

The processing time was investigated by conducting experiments at 1, 3, 5, 7 and 10 min, respectively. The size of alumina used for this study was 0.2 μm. The mass percentages of alumina guest particles used were based on the assumption of 100% surface coverage of the host particles with a monolayer of guest particles. The surface coverage of the coated products as a function of processing time is shown in Fig. 12. An increase in processing time from 1 to 5 min showed a corresponding increase in the percentage of surface area covered. After 5 min, the amount of surface coverage achieved fluctuated slightly. A previous study [23] has shown that it is possible for the magnetic particles to detach and reattach the guest particles after an optimum processing time is reached. The small fluctuations after 5 min can be due to small differences between detachment and reattachment, where an ‘‘equilibrium’’ between both is reached, or to experimental error, which is estimated at about ±5%. The surface morphology of PMMA coated with alumina for processing times of 1 and 5 min are shown in Figs. 13 and 14, respectively. In Fig. 13, for a processing time of 1 min, a very discrete and relatively small amount of surface coating of alumina is seen as compared to a processing time of 5 min (Fig. 14), where the coating appears thicker and much more uniform.

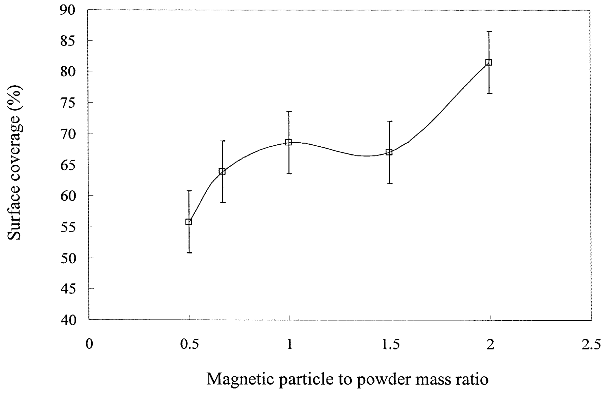

Fig. 17. Surface coverage of coated products as a function of magnetic particle to powder mass ratio.

3.2.2. Current and frequency

The effect of current on the coating efficiency was examined by varying the current at a fixed frequency of 45 Hz. The current was varied from 1 to 5 A and each experiment was conducted for a processing time of 5 min, with a guest particle size of 0.2 μm. It was found that the surface coverage increased linearly with increased current and the results are shown in Fig. 15. The maximum current that could be attained by the Triathalon Power Control, which we used in our experiments, was 5.0 A.

Using an optimum current of 5.0 A and a processing time of 5 min, the frequency of the system was varied from 45 to 100 Hz. An unusual behavior was observed as shown in Fig. 16. At 45 Hz, the surface coverage obtained was about 66%; then from 45 to 70 Hz, the surface coverage decreased with increasing frequency. After 70

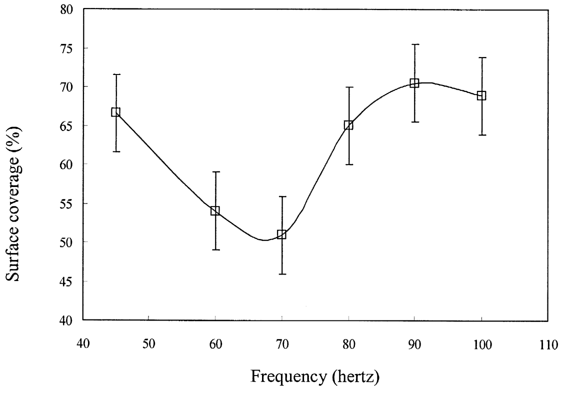

Fig. 16. Surface coverage of coated products as a function of frequency at a fixed current of 5 A.

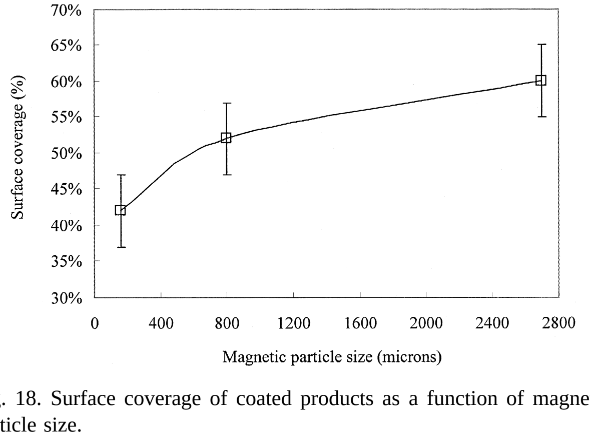

Fig. 18. Surface coverage of coated products as a function of magnetic particle size.

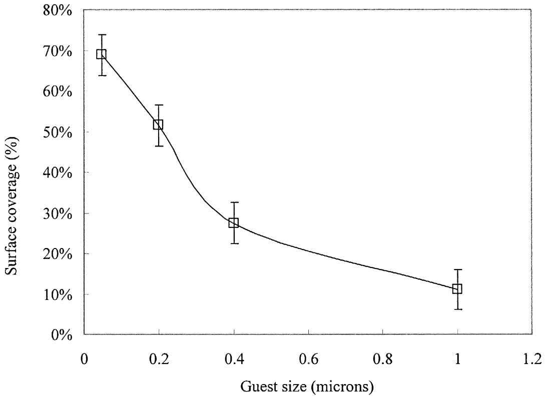

Fig. 19. Surface coverage of coated products as a function of guest particle size.

Hz, the surface coverage of the coated product increased gradually with increasing frequency, where it again peaked in surface coverage at a frequency of 90 Hz. This behavior suggests that the variations in the frequency cause a periodic fluctuating behavior in the amount of surface coverage obtained.

3.2.3. Magnet to powder mass ratio

The motion of the magnetic particles is responsible for the coating of guest particles onto the host particles by a vigorous ''fluidized'' type motion causing collisions between host particles and the host and guest particles. Therefore, the mass of magnetic particles used in the system significantly affects the surface coverage obtained. The magnetic particle to powder mass ratio was varied to determine the optimum mass of magnetic particles needed. Several ratios were investigated and the results obtained are shown in Fig. 17. It can clearly be seen that as the magnetic particle to powder mass ratio is increased, the percentage of surface area covered also increased. For ratios larger than 2, it has been shown that there is not much change in the coating efficiency, and in some cases coating is even poorer than at lower ratios [23].

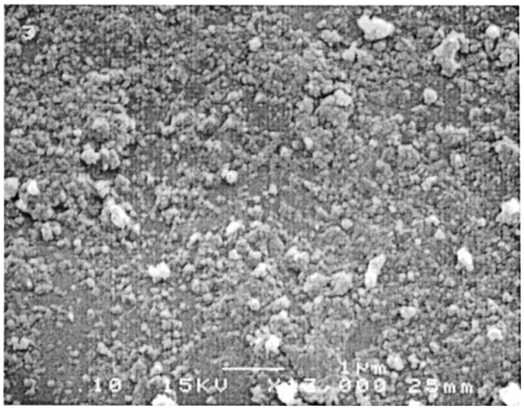

Fig. 20. Surface morphology of PMMA coated with 0.5 μm Al_2O_3 guest particles.

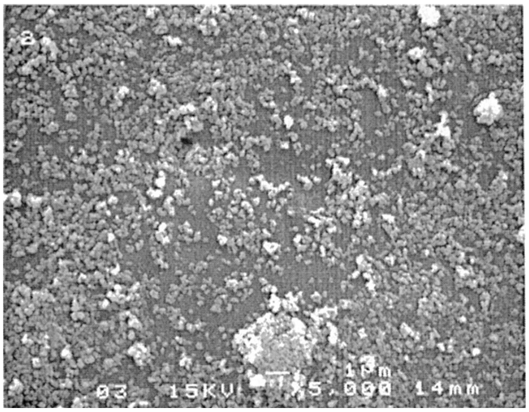

Fig. 21. Surface morphology of PMMA coated with 0.2 μm Al_2O_3 guest particles.

3.2.4. Magnetic to host size ratio

Three sizes of magnetic particles were used to investigate the effect of size on coating in the MAIC system. The percentage of surface coverage achieved for each size is shown in Fig. 18. As seen in the figure, the largest magnets with a mean size of 2.7 mm gave the best surface coverage results.

3.2.5. Guest to host size ratio

The size effect of guest particles on the coating efficiency was investigated using four sizes of alumina guest particles: 0.05, 0.2, 0.4 and 1 μm. Based on complete coverage with a monolayer of alumina particles, the percentage of alumina by weight for each of these 4 sizes was chosen as 0.25, 1.0, 2.0 and 5.0 respectively. As the guest particles size increased, the area of coverage decreased, as shown in Fig. 19. The two smallest sizes gave the best coating results. However, careful examination of the surface morphology of the products coated with 0.05 and 0.2 μm alumina, Figs. 20 and 21, respectively, showed that the PMMA particles coated with 0.2 μm alumina were more uniformly coated than the PMMA particles coated with 0.05 μm alumina. Many more agglomerates of alumina were observed on the surface of PMMA for a guest

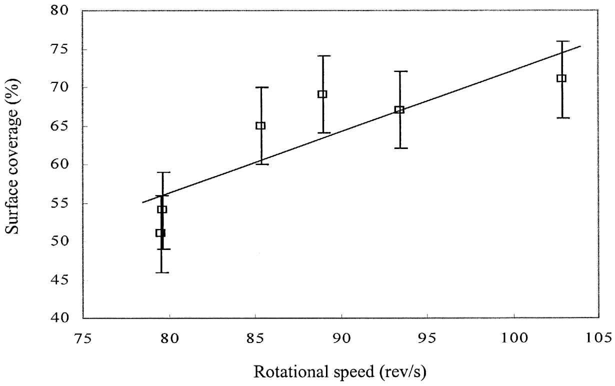

Fig. 22. Surface coverage of the coated products as a function of the rotational speed of the magnetic particles.

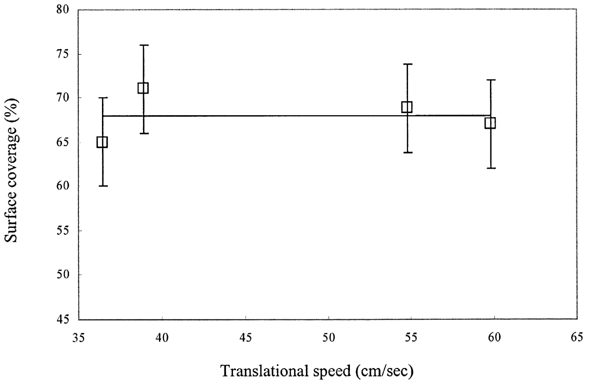

Fig. 23. Surface coverage of the coated products as a function of the translational speed of the magnetic particles.

size of 0.05 μm. This is due to the inability of the MAIC device to efficiently de-agglomerate the 0.05-μm size guest particles. This conclusion was based on the study of several SEM micrographs (not included in the paper). Therefore, from these observations, the 0.2 μm-sized guest particles were considered to be the better guest size for obtaining a more uniform surface coverage.

3.2.6. Magnetic particle speed and behavior studies

A small experimental system consisting of magnets, host and guest particles was assembled similar to that used for the parameter studies. Using a Kodak camera, the movement of the magnetic particles was observed at different frequencies at a fixed current of 5 A. The first important observation made was that in addition to the magnetic particle moving haphazardly in all directions (translation), they were also spinning furiously (rotation). The movement of the magnetic particles at different frequencies was measured and recorded. These recorded images were further analyzed to obtain approximate values for the translational and rotational motions. The translational and rotational speeds as a function of frequency were then related to the previous study of surface coverage as a function of frequency. Thus, the relationships of the surface coverage as a function of translational and rotational speed were obtained (Figs. 22 and 23). The second important observation (obtained from the figures) showed that the rotational speed of the magnetic particles influenced the coating efficiency more significantly than the translational speed.

The combination of parameter and magnetic particle motion studies suggests that the primary motion due to the magnetic field is the spinning of the magnetic particles, promoting de-agglomeration of the guest particles as well as the spreading and shearing of the guest particles onto the surface of the host particles. However, the effect of the translational speed is also significant as it allows for the impaction of one particle onto another, promoting coating. Further studies are clearly needed in this area to clarify these phenomena. A schematic diagram of a proposed mechanism of coating in the MAIC derived from this study is shown in Fig. 24.

We are presently using discrete element simulation to develop a model to help us understand the behavior of the magnetic particles and its influence on the overall coating performance in the MAIC system. Preliminary results show that the observed motion of the magnetic particles matches reasonably well with the high-speed video images of an actual experiment. Quantitative results of the simulation study will be presented in a forthcoming paper.

4. Conclusions

In this work, the feasibility of using the MAIC device for the surface modification of cornstarch and cellulose fibers was demonstrated. Both materials flowed better after a discrete coating of silica was added to the surface. In

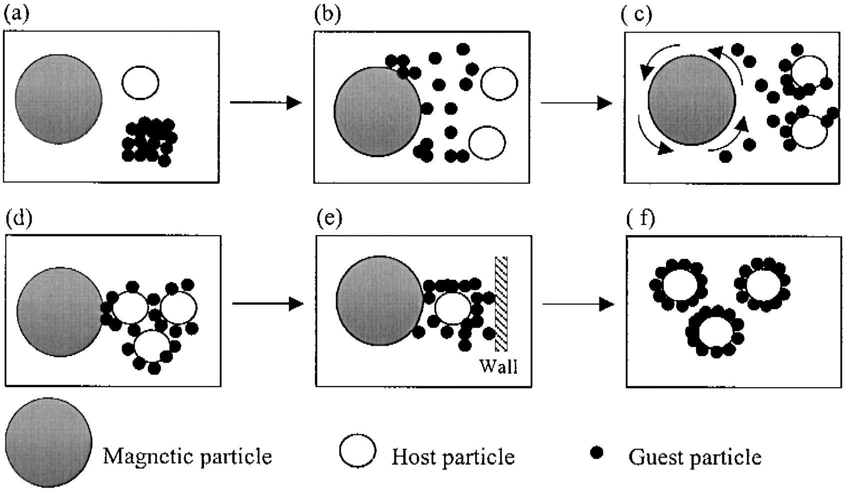

Fig. 24. Mechanism of coating in the MAIC process: (a) excitation of magnetic particle, (b) de-agglomeration of guest particles, (c) shearing and spreading of guest particles on the surface of the host particles, (d) magnetic–host–host particle interaction, (e) Magnetic–host–wall interaction and (f) coated products.

addition, wettability tests revealed that coated products absorbed less water after coating, indicating a reduction in hydrophilicity. Preliminary FTIR studies also showed that there was a reduction in O–H groups for the coated as compared to the uncoated samples, lending credence to the hypothesis that a mechanochemical reaction occurred.

The optimum parameter studies showed that there is an optimum processing time for the MAIC device, dependent on the materials being coated. An increase in magnetic particle size increased the coating efficiency, with an optimum magnetic to host size ratio of approximately 10. The coating efficiency increased with decreasing guest size. However, with very small guest particles the uniformity of coating is poor due to severe guest particle agglomeration.

With the use of high-speed imaging, it was found that the magnetic particles in the MAIC system spin furiously during the coating process. The rotational speed (spinning motion) is more dominant than the translational speed in its influence in particle coating. We propose that the mechanism of coating is the de-agglomeration of the guest particles, followed by the shearing and spreading of the guest particles onto the surface of the host particle, coupled with host–magnet–wall collisions and interactions.

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