Effects of metal surface morphology on deposition behavior of microstickies from papermaking white water

YUTING XU, XIAOLI CAO, CARL HOUTMAN, BO LI, WENXUAN MO, AND XIAOHONG LI

ABSTRACT: Deposition of small adhesive particles, called microstickies, onto pulp processing equipment and paper machines causes quality and operational problems for recycling mills. The factors that control deposition of microstickies onto surfaces of metal parts remain unclear.

In this work, aluminum surfaces with a range of surface roughness were exposed to slurries containing microstickies. The deposition results showed that flat surfaces promote the aggregation and deposition of microstickies particles. Uneven surfaces tended to favor deposition of smaller microstickies, 0.2–1 μm, which may be related to greater contact area presented by the rougher surface. This work provides insights into the deposition of microstickies.

Application: The deposition behavior of microstickies on metal surfaces is related to their surface properties, and it is helpful for mills to understand this as a way to avoid sticky deposition on devices.

Recovered paper has become an essential raw material for the paper industry. It can provide advantages in terms of saving resources, reducing environmental pollution, and lowering energy consumption [1-3]. However, this paper carries contaminants, particularly sticky substances, which unavoidably cause problems during paper recycling, along with a reduction in quality of paper products [4-7]. Due to their adhesive properties, stickies are not only retained in the white water, but are also deposited on metal surfaces of the paper machine, such as machine chest walls, degassing units, runner walls, forming fabrics, felts, and driers [8,9]. The deposition of stickies can lead to reductions in machine runnability and increases in web breaks, which ultimately reduce economic efficiency [10,11].

Stickies can be divided into microstickies that are smaller than 100 μm and macrostickies that are larger than 100 μm. Most macrostickies can be efficiently removed by slotted screening, but microstickies pass through the slots and are carried with the pulp and white water. As a result, microstickies accumulate in mill water cycles to a high level. In addition, the reduction in recovered paper quality due to China's waste import ban has brought an even more complex mixtures of stickies to mills. Therefore, removal of stickies is an urgent problem in the paper industry, especially in China.

Stickies in recovered paper contain various components, such as adhesives, coating binders, ink residues, deinking chemicals, and wood derivatives. These materials share many properties, e.g., low surface energy, deformability, hydrophobicity, etc. These complex characteristics likely play a role in controlling stickie deposition on equipment during pulp and papermaking processes. Doshi et al. [12] studied the deposition of microstickies in pulp on the surface of different materials, such as polypropylene (PP) microfoam, low-density polyethylene film (LDPE), high-density polyethylene flat piece (HDPE), and polyester wire. The study indicated both identity and morphology of the surfaces affect deposition rate. Ruben et al. [13] investigated the deposition on other parts of a paper machine, including the wire, press, and drying sections, in a paper mill using 100% recovered newspapers. They observed that deposition on drying cylinder surfaces was particularly serious. Doshi et al. [14] also concluded that colloidally dispersed stickies tend to precipitate on drying cylinder surfaces. Chen [15] observed that surface roughness of drying cylinder surfaces affects deposition of stickies. However, the factors controlling deposition of microstickies remain unclear.

In this study, we investigated deposition of microstickies in white water onto metal surfaces. Aluminum foil was chosen as the experimental material. A commercial adhesive was coated on paper and processed under laboratory conditions to obtain the microstickies slurry. The deposition was measured by determining the quantity of microstickies remaining in the white water after the test. The major effect studied was surface. The measure of surface roughness indicates the degree of unevenness.

EXPERIMENTAL

Materials and process

Materials

The solvent-based acrylic adhesive with 40% solids content was supplied by Changxing Chemical Industry Co. Ltd. in Guangzhou, Guangdong, China. The A4 copy paper with a basis weight of 80 g/m² was supplied by Asia Pacific Senbo Paper Industry Co. Ltd. in Rizhao, Shandong, China. Flat aluminum foil with a thickness of 70 µm was the representative metal surface. It was MONA, a brand obtained at the supermarket.

Coating

The copy paper was kept at ISO Standard constant temperature and relative humidity (RH) laboratory conditions (23±1°C, 50±2% RH) for at least 48 h to balance moisture, and then its weight, m_1, was measured. The adhesive was uniformly coated on the surface of the copy paper by an automatic coater (ZAA2300, Zehntner GmbH Testing Instruments; Sissach, Switzerland). The coated paper was kept at ISO Standard constant temperature and RH laboratory conditions for at least 48 h to balance moisture, and then the weight of the coated paper, m_2, was measured. The weight of adhesive coated on each paper was obtained by subtracting m_1 from m_2.

Pulping

The balanced coated paper was torn into small pieces and pulped at 50±1°C, 300 rpm, and 15% consistency for 10 min in a high consistency repulper (Formax 450H, Adirondack Machine Corp.; Hudson Falls, NY, USA). Next, the pulp was put into the Messmer Somerville sieve (S401700001, Testing Machines Inc.; New Castle, DE, USA). The accepts were collected.

White water preparation

The accepts were diluted to 2% consistency and agitated at 50±1°C and 350 rpm for 1 h. Then, they were filtered through a dynamic drainage jar (DDJ, MT2110-086CF, Cleveland Motion Controls; Ladson, SC, USA) with a standard screen of 200 mesh. The material that passed through the mesh—water, microstickies, fines, etc.—was collected and used for the subsequent experiments.

Design and measure of interface morphology

The flat aluminum foil was roughened by pressing specially-made needles with tip diameters of 0.4 mm, 0.6 mm, and 0.8 mm to obtain the surfaces with hemispherical bulges at an interval of about 1 mm. In order to facilitate analysis, rough surfaces were prepared in a regular arrangement on aluminum foils of the same size.

The surface roughness (*Ra*) of metal materials with different interface morphology was measured by first imaging with a metallographic microscope (DMI3000M, Leica Microsystems; Wetzlar, Germany) and then calculating *Ra* with the GWYDDION modular program [16]. The value of *Ra* (µm) can be expressed by the equation:

$$Ra = \frac{1}{l}\int_0^l |Y(x)|\, dx \quad (1)$$

where l and $Y(x)$ in µm represent the sampling length and the distance from each point on the contour to the center line, respectively.

The surface tension surface contact angle tester (OCA40 Micro, DataPhysics Instruments; Filderstadt, Germany) was used to measure the contact angle of the aluminum foil and the surface of the adhesive material by using water and diiodomethane. Angles were measured by dropping 2 µL of liquid from a syringe onto a surface and taking photos immediately. The surface energy (γ, mJ/m²) of the material was calculated from the following equation [17]:

$$\gamma = \gamma^p + \gamma^d \quad (2)$$

where the superscripts in γ^p (mJ/m²) and γ^d (mJ/m²) represent the dipole-hydrogen bonding and dispersion force components, respectively.

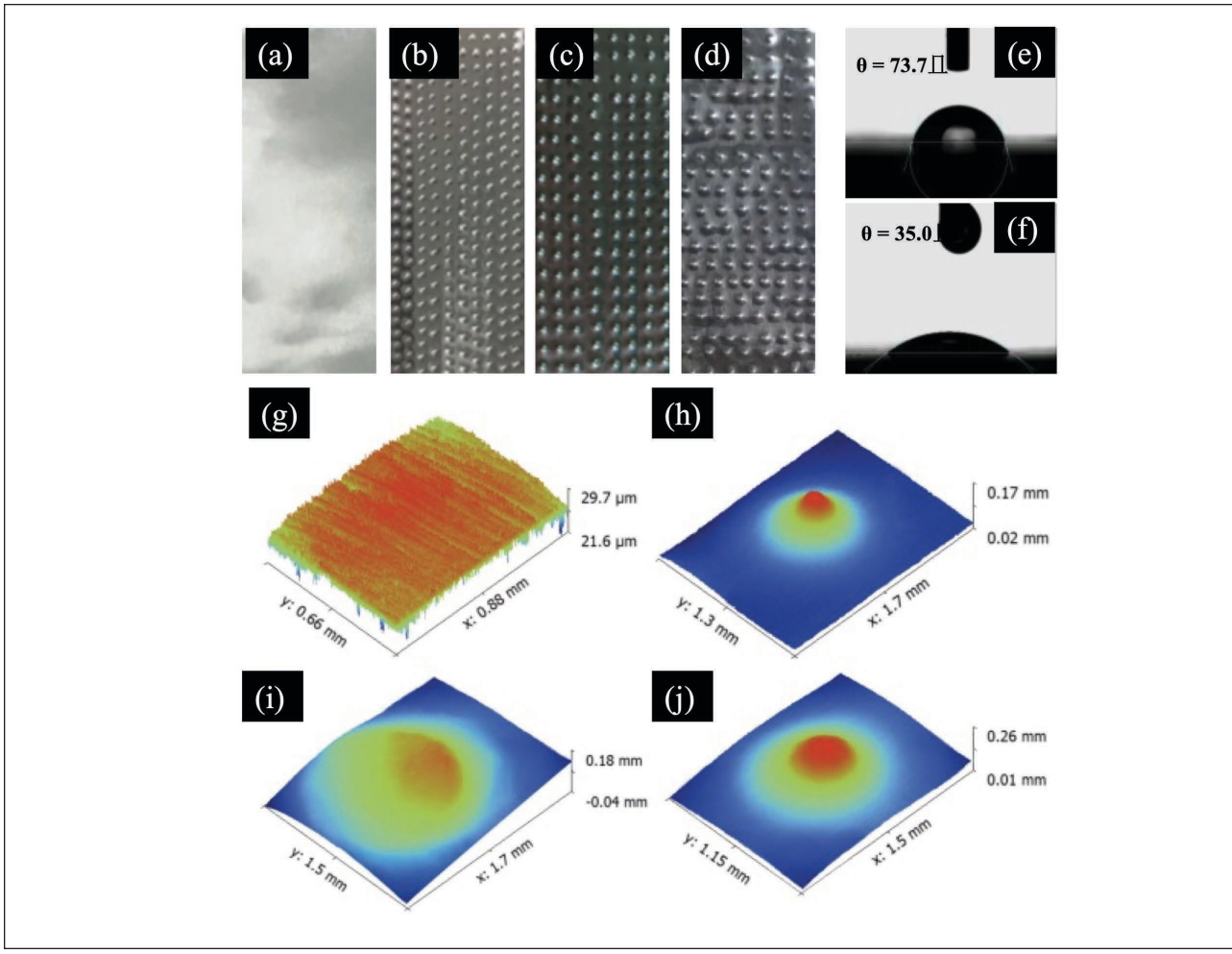
Characterization of adhesive

Fourier transform infrared (FTIR) spectrometry measurements were carried out on a Bruker Tensor 27 FTIR spectrometer (Bruker; Billerica, MA, USA) at frequencies from 400 to 4000 cm^{-1} in the ART mode. Thermogravimetric analysis (TGA) was performed in a thermogravimetric analyzer (Q500, TA Instruments; New Castle, DE, USA). The temperature range was 25°C to 700°C with a heating rate of 20°C/min under a 40 mL/min flow of nitrogen (N_2). The data on weight loss rate was analyzed using TA Instruments Universal Analysis software. The glass transition temperature (Tg) of the samples was estimated from differential scanning calorimeter heat signature (DSC214, Netzsch Taurus Instruments; Weimar, Germany). The temperature range was -60°C to 90°C with a heating rate of 10°C /min under a 40 mL/min flow of N_2.

Deposition of microstickies from white water

A measured volume of prepared white water was placed in a beaker and kept at 50°C. The aluminum foils were wrapped on the surface of a stirring paddle. Then, the paddle covered with aluminum foil was placed in white water for the deposition test. The length and height of the paddle placed in white water were kept at 5 cm. The rotation speed of the stirring paddle was maintained at 80 rpm. The white water was sampled at 0 h, 2 h, and 4 h. The residual microstickies in white water after deposition were determined by flow cytometry, which has been previously described by our laboratory [18-22].

The particle size of microstickies was expressed as cube



1. (a) Flat aluminum foil, and (b-d) the modified ones with different morphology interfaces. Surface wettability of the flat aluminum foil: (e) water, and (f) diiodomethane. Modified aluminum foil with different surface roughness: (g) Ra_1 = 0.752 µm; (h) Ra_2 = 16.7 µm; (i) Ra_3 = 29.9 µm; and (j) Ra_4 = 36.2 µm.

root diameter d (µm), total volume v (µm³), and total number n of particles. They are defined by the equations:

$$d = \sqrt[3]{\frac{\sum n_i d_i^3}{\sum n_i}} \quad (3)$$

$$v = \sum_{i=1}^{n} v_i \quad (4)$$

$$n = \sum_{i=1}^{m} n_i \quad (5)$$

where d_i, n_i, and v_i (i=1,2,3…) refer to the equivalent volume diameter of a single particle, the total number of particles with a diameter of d_i, and the equivalent volume of a single particle, respectively, and m refers to the total number of d_i.

The particle size distribution of the microstickies was represented by the particle number frequency distribution g (%). The g can be expressed by Eq. 6:

$$g = \frac{n_i}{n} \times 100\% \quad (6)$$

The deposition of microstickies on the surface was expressed by the deposition efficiency η (%). The η can be expressed by Eq. 7:

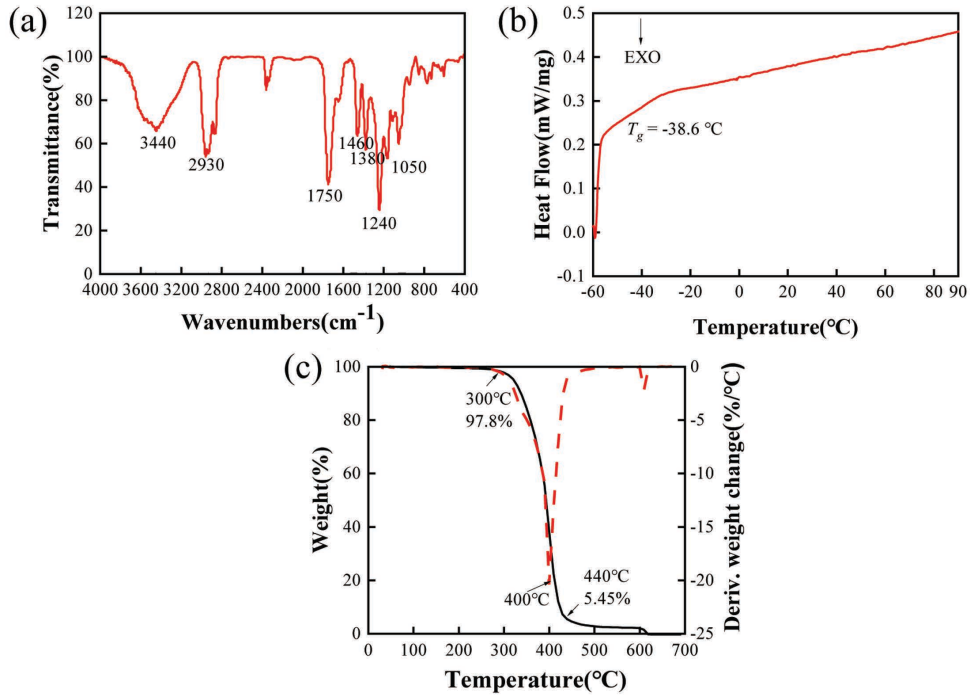
$$\eta = \frac{v_0 - v_1}{v_0} \times 100\% \quad (7)$$

where v_0 (µm³) and v_1 (µm³) are the total volume of particles in white water before and after deposition, respectively.

RESULTS AND DISCUSSION

Characteristics of the metal and adhesive surfaces

Figures 1a–1d show photos of the aluminum foil with different surface morphologies. The static contact angles of the surface of the original aluminum foil and the adhesive were tested and then used to predict their wetting



2. (a) Fourier transform infrared (FTIR) spectra of adhesive; (b) differential scanning calorimeter (DSC) heat signature curves of adhesive; and (c) mass loss of organic matter with temperature (TG) and decomposition of organic matter constituents with temperature (DTG) curves of adhesive.

characteristics and surface energy. Figure 1e and Fig. 1f showed images of droplets of water and diiodomethane on aluminum foil, with static contact angles of 73.7° and 35.0°. The same method was used to test the adhesive. The surface energies of the aluminum foil and the adhesive were 42.88 mJ/m² and 40.83 mJ/m², respectively. The difference in surface energy between the foil and the adhesive can be used to predict the compatibility of the two surfaces. This means that the smaller the surface energy difference between the two surfaces is, the more stable the association [23,24]. Therefore, the stickies are compatible with the aluminum foil and should show deposition.

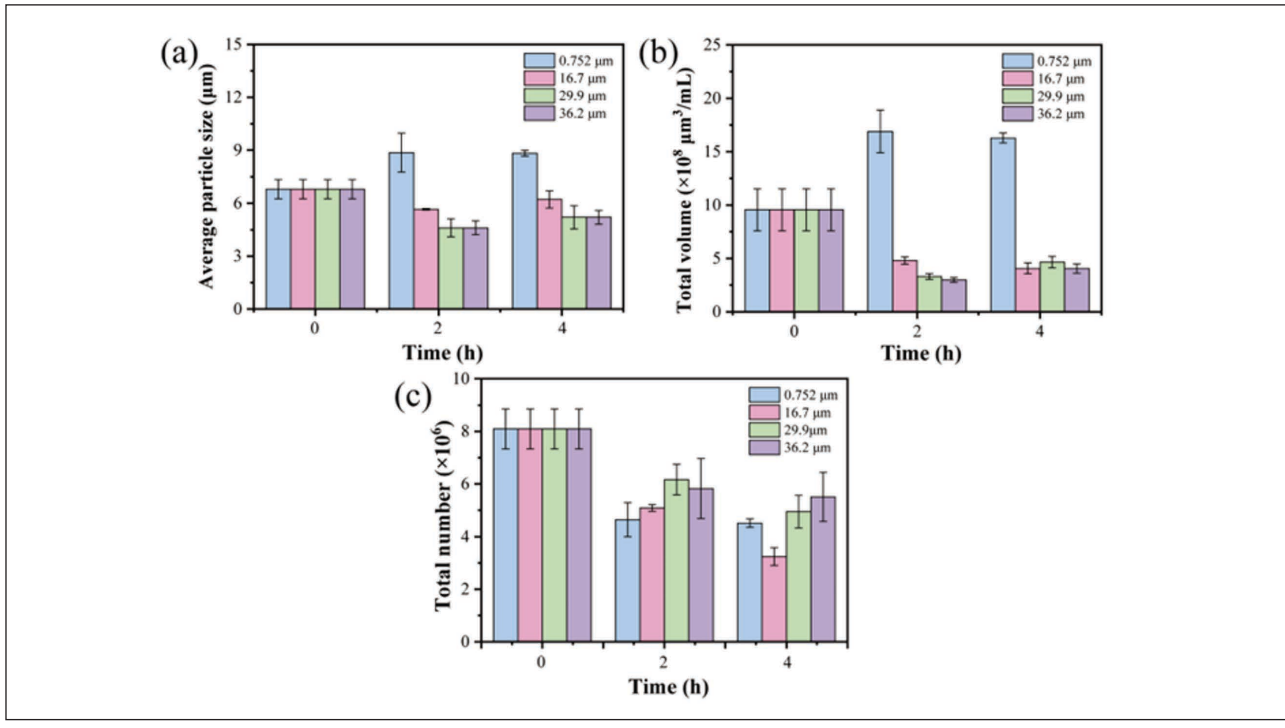
Surface roughness was measured to indicate the degree of surface unevenness. The surface roughness of the modified surface of the aluminum foil was characterized by the metallographic microscope. As shown in Fig. 1g, it can be found that the unprocessed aluminum foil had a relatively flat surface. Figures 1h–1j showed that the surface of aluminum foil had a convex structure after physical treatment. According to the international comparison table of surface roughness, a surface with Ra<0.8 µm was generally called a mirror surface, which was relatively flat. A surface with Ra_1>12.5 µm was called a rough surface. Therefore, the surface of aluminum foil with Ra_1 = 0.752 µm can be considered a flat surface, and the surfaces of aluminum foil with Ra_2 = 16.7 µm, Ra_3 = 29.9 µm, and Ra_4 = 36.2 µm can be called rough surfaces.

Basic properties of the adhesive

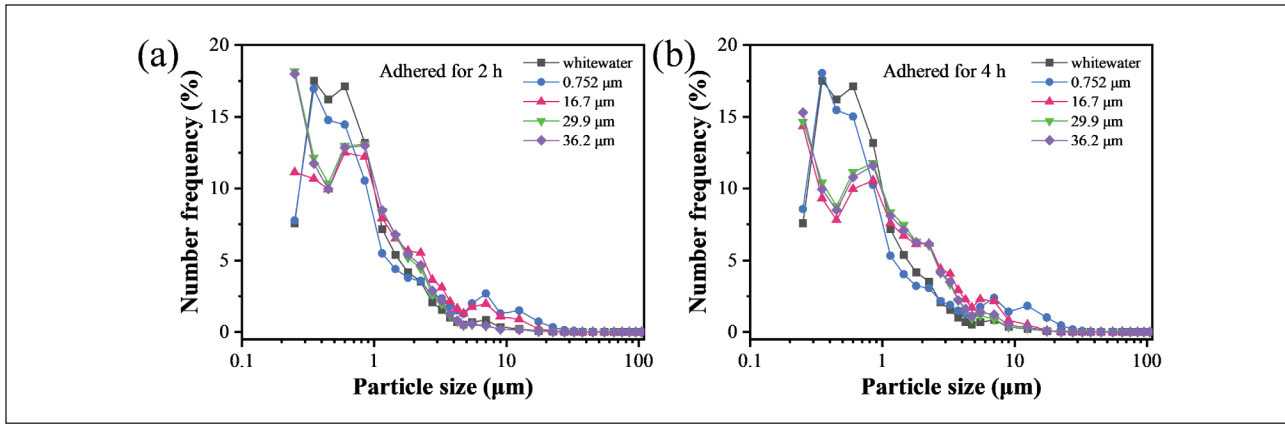
The functional groups of the adhesive were identified with FTIR, as shown in **Fig. 2a**. The characteristic peaks of FTIR spectra and corresponding structures of the adhesive are shown in **Table I**. The FTIR spectra of the adhesive showed a stretching vibration peak of OH at 3440 cm^{-1}, a C=O stretching vibration peak, a C-O-C asymmetric stretching vibration peak, and a C-O stretching vibration peak in O-CH at 1750 cm^{-1}, 1240 cm^{-1}, and 1050 cm^{-1}, respectively. These peaks indicated the adhesive might contain unsatu-

Wavenumbers, cm^{-1}	Corresponding Structures
3440	O-H stretching vibration
2930	-CH_3 stretching vibration
1750	C=O stretching vibration
1460	-CH_2 bending vibration
1380	C=O enhanced -CH_3 bending vibration
1240	C-O stretching vibration of the ester group
1050	C-O stretching vibration of the aldehyde group

I. The characteristic peaks of Fourier transform infrared (FTIR) spectra and corresponding structures of the adhesives.



3. The effect of the unevenness of the aluminum foil (Ra$_1$ =0.752 μm, Ra$_2$ = 16.7 μm, Ra$_3$ = 29.9 μm, and Ra$_4$ = 36.2 μm) on the: (a) average particle size of microstickies; (b) total volume of microstickies; and (c) total number of microstickies before and after deposition.



4. Effect of aluminum foil with different surfaces (Ra$_1$ = 0.752 μm, Ra$_2$ = 16.7 μm, Ra$_3$ = 29.9 μm, and Ra$_4$ = 36.2 μm) on the size distribution of microstickies in white water after deposition on aluminum foil.

rated fatty acid esters. In addition, the FTIR spectra also showed the -CH_2 asymmetric stretching vibration peak, the -CH_2 bending vibration peak, and the -CH_3 bending vibration peak at 1930 cm^{-1}, 1460 cm^{-1}, and 1380 cm^{-1}, respectively. Therefore, the stickies were presumed to contain a large number of alkanes. Based on the primary type of the adhesive [22], it can be assumed that the main component of the adhesive was ethylene vinyl acetate copolymer.

Inspection of Fig. 2b shows that the glass transition temperature of the adhesive was -38.6°C. The samples continued to absorb heat as the temperature rose, which suggests the samples are a partial solid solution. Previous studies [25] have shown that when the temperature of the polymer exceeds its Tg, it becomes elastic and tacky. The range of 40°C to 70°C was a typical temperature range in papermaking processes. In this range, the adhesive is in a highly elastic state, so the adhesive is likely tacky at typical papermaking temperatures, which supports the observation that stickies deposit on equipment and aggregate with each other.

The result of Tg showed the basic features of the organic components, as shown in Fig. 2c. The pyrolysis range of the adhesive was narrow, and the pyrolysis rate was relatively fast. The main pyrolysis reactions began at 300°C and decomposed at 440°C. Below 300°C was the evaporation process of moisture in the sample, and the weight loss rate of the sample was about 2%. The range of 300°C to 400°C

was the main area of weight loss, and the weight loss rate of the sample was as high as 92%. The peak temperature corresponding to the highest peak of the weight loss rate was 400°C. This stage was mainly chain breakage of the adhesive and the pyrolysis process of the carbonaceous material. Combined with the analysis results from differential scanning calorimeter (DSC) heat signature, one can conclude that the adhesive was far from its decomposition temperature. It was still in the solid state. However, the adhesive exists in a high elastic state, which had a certain degree of tackiness.

Effects of deposition on size distribution of microstickies

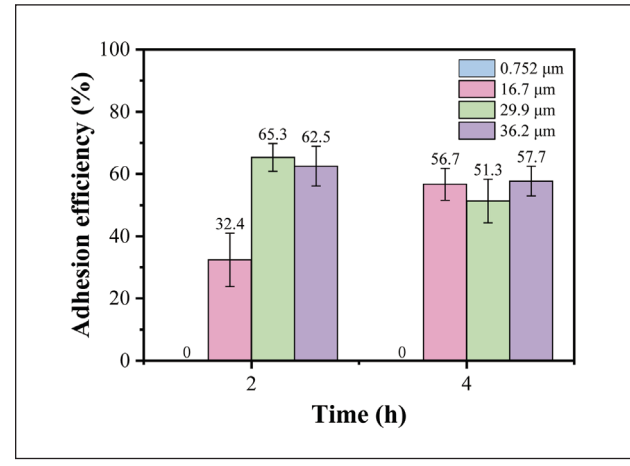
The variation of the size distribution for residual microstickies illustrated the effect of deposition and aggregation on the microstickies in white water. The size and number distributions of microstickies before and after the deposition on aluminum foil with different surfaces are shown in **Fig. 3** and **Fig. 4**.

Stirring with a rotor covered with a flat (Ra_1 = 0.752 µm) aluminum foil surface appears to favor aggregation. The average particle size and total volume increases after mixing for 2 h or 4 h (Fig. 3a and Fig. 3b). Increases in total volume indicate that the aggregates are not dense and appear larger than the constituent particles. This observation of aggregation is in contrast to results with uneven surfaces (Ra_2 = 16.7 µm, Ra_3 = 29.9 µm, and Ra_4 = 36.2 µm). The unevenness of surface appears to promote the dispersion of microstickies. The average particle size of the residual microstickies in white water and the total volume decreased. A reduction in the number of particles, Fig. 3c, was observed for all aluminum surfaces. The change in the number of particles can be due to both aggregation and deposition.

Inspection of Fig. 4 shows that the size distribution of the original white water was wide and dominated by small particles, with the largest number in the 0.2–1 µm range. Upon stirring with a flat aluminum foil rotor for 2 h or 4 h, there are not significant changes in the small ranges, but one can observe an increase in the 10–20 µm range. This result agrees with the observation that flat aluminum foils promote aggregation of microstickies. In comparison, the uneven surfaces promote a dramatic loss of particles in the 0.2–1 µm range. This indicated that the smaller microstickies deposited on the uneven aluminum foils. After 4 h of exposure, the number of residual microstickies in the white water increased in the range of 1–3 µm. The increase in number may be due to agglomeration of small sized particles or detachment from the aluminum foil surface.

Effect of interface morphology on deposition

The deposition efficiency of aluminum foils with uneven surfaces was higher than that of flat aluminum foil, as shown in **Fig. 5**. As noted previously, stirring with the flat aluminum foil promoted aggregation, and total volume and size of the microstickies increased. One would expect shear forces on microstickies with a larger radius to be higher so there will be less deposition. Microstickies of small particle size appeared to be well removed by uneven surfaces, as shown in Fig. 4. The uneven surface may increase the contact area between the aluminum foil and the microstickies, promoting deposition.



5. Effect of aluminum foil surface unevenness on deposition of microstickies.

The metal surface unevenness affects the deposition efficiency of microstickies from white water. An uneven metal surface increases the contact area of the microstickies, which in turn changes its deposition efficiency.

CONCLUSIONS

In this work, deposition of microstickies from papermaking white water onto aluminum metal surfaces with varying unevenness was investigated. The results showed that flat surfaces promote the aggregation of microstickies. The average particle size increased from 7 to 9 µm. Uneven aluminum foils appeared to collect microstickies with smaller particle sizes (0.2–1 µm). However, as the deposition proceeds, aggregates appear to be released. One explanation is that uneven surfaces have higher contact areas and promote more deposition of microstickies onto the aluminum foil. These conclusions provide a possible explanation for the accumulation of microstickies at particular locations on paper machines and suggest that surface morphology is an important consideration. **TJ**

ACKNOWLEDGEMENTS

The authors are grateful for the National Natural Science Foundation of China (31470607); R&D projects in key areas of Guangdong Province (2022B1111080004); Natural Science Foundation of Guangdong Province (2023A1515011941); and the Foundation of Hubei Key Laboratory of Pollutant Analysis & Reuse Technology, Hubei Normal University, China (PA200101).

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ABOUT THIS PAPER

Cite this article as:
Xu, Y., Cao, X., Houtman, C., et al., *TAPPI J.* 22(7): 466(2023). https://doi.org/10.32964/TJ22.7.466

DOI: https://doi.org/10.32964/TJ22.7.466

ISSN: 0734-1415

Publisher: TAPPI Press

About this journal

ABOUT THE AUTHORS

We chose this topic of study to better understand why microadhesives deposit on the surface of paper machines. This work provides insights into how the deposition behavior of microstickies on metal surfaces is related to surface uneveness.

The most difficult part of the research was the characterization of the uneven surface; roughness was chosen as its index. We ultimately discovered that the uneveness of metal surfaces has an effect on the deposition of microstickies.

Based on this research, mills can change the properties of metal surfaces to reduce the deposition of microstickies and to promote quality production. The next step is to study the influence of roughness of different materials on microstickies to further explore the deposition mechanism of microstickies.

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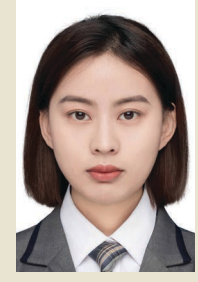

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