Coating

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Research on brightening modification of molecular sieves coated fly ash based on alkaline melting hydrothermal method

<https://doi.org/10.1515/npprj-2021-0007>

Received January 23, 2021; accepted March 31, 2021; previously published online April 16, 2021

Abstract: Improving the utilization rate of fly ash (FA) and realizing its resource utilization has always been a popular research topic in the application field of solid waste recycling. This paper aims to improve the basic brightness of fly ash (FA) before calcium carbonate coating, and realize the filling application of fly ash in medium and high-end paper. Taking advantage of the high brightness of molecular sieves (MS) (brightness $\geq 90\%$ ISO), the alkali melting hydrothermal synthesis (AM-HS) method was used to generate high brightness MS on the surface of fly ash. The results showed that the optimum conditions of alkali melting hydrothermal synthesis (AM-HS) modification were as follows: the mass ratio of sodium hydroxide to fly ash was 1:1, the time of hydrothermal modification was 6 h, the temperature of hydrothermal modification was 80 °C, and the brightness reached to 64.54 %ISO; The results of SEM, particle size analysis and ultrasonic vibration showed that the surface of fly ash is covered by MS and has strong binding effect.

Keywords: alkali melting; brightness; fly ash; hydrothermal synthesis; molecular sieves.

Introduction

Fly ash (FA) is a kind of pozzolanic mixed material formed by high temperature combustion of pulverized coal (par-

ticle size $\leq 100\mu\text{m}$). At present, most of them are disposed by landfilling and stacking, which not only occupies farmland, but also causes serious air pollution due to a large amount of dust (Dias-Ferreira et al. 2016); the toxic substances contained in FA, such as Pb, MO, Ni, V, As and other heavy metals (Murayama et al. 2002), will pollute groundwater, affect the growth of aquatic organisms, and destroy the ecological balance. In the recent years, domestic and foreign scholars on the resource utilization of FA have mainly focused on building materials (such as Portland cement and concrete) (Mcdonald 1994, Hemalatha and Ramaswamy 2017, Joshaghani et al. 2018, Phoungthong et al. 2018), sewage treatment (Visa 2012, Li et al. 2014, Zhang and Yu 2018), mineral extraction (Gong et al. 2016, Dai and Finkelman 2018), soil improvement (Jala and Goyal 2006, Pandey and Singh 2010), etc. However, there are still a series of problems such as low utilization rate of FA, high treatment cost and low added value, the utilization of FA is seriously restricted.

FA and filler kaolin have similar chemical composition, physical property and unique surface structure, which are potentially suitable for paper filling (Cherian and Siddiqua 2019). In 2008, Sinha et al. (Sinha 2008, Sinha et al. 2014) used FA as a filler for paper filling for the first time, but the brightness of the paper was too low after filling. Subsequently, Yumin Fan and Xueren Qian (Fan 2012) of Northeast Forestry University, Ming Wang (Wang 2015), Shuo Wang (Fan et al. 2015), Xueqin Wang (Fan et al. 2017a, Fan et al. 2018) and Yanan Qi et al. (Fan et al. 2017b) from South China University of Technology have successively brightened and modified FA and used them in paper filling research, but the effect is not very satisfactory. The low brightness of FA is still the main reason hindering its use in papermaking.

The brightness is positively correlated with the amount of light diffusely reflected by the light irradiated material. According to the Kubelka-Munk theory, it is deduced that the greater the relative refractive index of a substance, the light reflectivity will also increase (Shi et al. 2007). Due to the relatively high relative refractive index and low light absorption coefficient of molecular sieves

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(MS), the MS has the characteristics of high brightness. When a purer MS is synthesized on the surface of FA, its brightness will also increase.

The most abundant chemical component in FA is acidic oxide, and the glassy phase substance in the shallow layer of FA is tightly structured, making it difficult for acid to enter. Therefore, it is easier to stimulate the activity of FA in an alkaline environment. The chemical bonds of the glass phase are mainly Si–O and Al–O bonds. Based on previous studies, these chemical bonds are susceptible to the influence of OH[−], and Si–O–Al breaks, thereby reducing the degree of polymerization of the glass body network polymer, forming free unsaturated active bonds on the surface, which is easy to chemically react with alkali.

The synthesizing MS technology from FA mainly include alkali melting hydrothermal synthesis (AM-HS) method (Shigemoto et al. 1993, Liu et al. 2019, Yang et al. 2019), one-step synthesis method (Murayama et al. 2002, Cardoso et al. 2015), two-step synthesis method (Querol et al. 1997, Hollman et al. 1999), microwave-assisted synthesis method (Inada et al. 2005, Arroyo-Martinez et al. 2018), seed crystal method (Zhao et al. 2016), inorganic solvent system synthesis method (Hobbs and Thomas 2007, Walsh et al. 2007), etc.

In 1985, Holler et al. (Ruen-ngam et al. 2009) successfully synthesized MS from FA, and the first to propose the HS method: mix the FA with a certain concentration of NaOH or NaHCO₃ or Na₂CO₃ or KOH, the Si and Al in fly ash (FA) dissolve under alkaline conditions to form aluminosilicate gel, and the aluminosilicate gel transforms into MS crystals under certain temperature, alkalinity and pressure conditions, is currently one of the most widely used methods. In the traditional HS method, the crystal activity of quartz and mullite in FA is very low, which makes it difficult to dissolve these crystals in alkali solution (Liu et al. 2019), but the content of these substances in FA is high.

In order to further improve the yield and purity of the products, Shigemoto (Shigemoto et al. 1993), Yang (Yang et al. 2019) and other researchers proposed the AM-HS modification: firstly, mix the raw materials with alkaline substances such as NaOH uniformly, then melting and calcinate at high temperature to obtain the precursor, and then conduct the HS to prepare MS. The main advantage of this method is that the inert Si–Al oxide in the FA can be converted into aluminosilicate with higher activity, and the conversion rate of Si–Al in the FA can be improved. In this paper, FA coated with MS was prepared by AM-HS modification.

In this study, the Si and Al on the surface of FA are used as raw materials to generate MS on the surface of

Table 1: Analysis of different chemical components of original FA.

SiO ₂	Al ₂ O ₃	CO ₃	CaO	Fe ₂ O ₃	TiO ₂
46.32 %	31.60 %	8.54 %	4.74 %	4.47 %	1.29 %
continued:					
K ₂ O	Na ₂ O	SO ₃	MgO	P ₂ O ₅	CeO ₂
0.63 %	0.60 %	0.48 %	0.42 %	0.37 %	0.20 %
continued:					
SrO	ZrO ₂	Cl	MnO	ZnO	
0.15 %	0.06 %	0.05 %	0.03 %	0.02 %	

FA through AM-HS method. The Si content in the original FA used in this experiment is 46.32 %, and the Al content is 31.60 %. After removing iron and carbon from FA, AM is used to destroy the glass phase structure on the surface, and then MS are formed on the surface of FA by HS reaction to coat its surface to improve the brightness of FA.

Materials and methods

Materials

Industrial grade of FA was obtained from GUANGZHOU PAPER GROUP LTD. in Guangzhou. The main chemical components of FA are shown in Table 1. The raw materials for the experiments were thoroughly placed in an oven at 105 °C to dry for 24 hours, then use a flapping vibrating screen to sieve 500–600 mesh FA and save it for use. The initial brightness of 600 mesh fly ash is 18.01 %ISO.

Analytical grade chemicals of hydrochloric acid and sodium hydroxide were purchased from Guangzhou Chemical ReagentCo.Ltd, and directly used without further purification.

Pretreatment

Weigh 3 g of 500–600 mesh FA and place it in a muffle furnace. The maximum temperature is set to 600 °C, the heating rate is 8 °C/min, and the heat preservation is 2 hours. After cooling, take it out for later use.

FA is mixed with a certain concentration of hydrochloric acid solution in a mass ratio of 1:5 (Li et al. 2010, Cao et al. 2010), reacted in a three-necked flask with a stirring speed of 300 rpm/min, filtered and washed until it is neutral, dried, and weighed; The classic 3-factor 3-level response surface test is used for the concentration of hydrochloric acid, acid leaching time and reaction temperature.

Table 2: Process conditions of FA in AM-HS modification reaction.

	Sodium hydroxide : FA	HS temperature/°C	HS time/h
1	0.4:1	80	6
2	0.6:1		
3	0.8:1		
4	1:1		
5	1.2:1		
6	1:1	50	6
7		60	
8		70	
9		90	
10	1:1	80	4
11			5
12			7
13			8

Alkali melting hydrothermal synthesis (AM-HS)

Alkali melting (AM): the FA and sodium hydroxide after carbon removal and iron removal pretreatment are mixed with sodium hydroxide in a certain mass ratio (0.4:1, 0.6:1, 0.8:1, 1:1, 1.2:1), and grinded in a mortar for 5 min. Then, the mixture is placed in a muffle furnace and calcined for 2 h when the temperature is 600 °C. Ensure that the vitreous substance in the FA is fully destroyed and activated, and retain as much inert crystal quartz and mullite as possible (Li et al. 2019). After the mixture has cooled, grind it in a mortar for a few minutes to prepare for the next HS modification.

Hydrothermal synthesis (HS): the ground AM treated FA and deionized water were mixed and stirred according to the solid-liquid ratio of 1:5, the rotating speed was 400 rpm/min, heated for several hours (2 h, 4 h, 6 h, 8 h, 10 h) in a water bath at different temperatures (50 °C, 60 °C, 70 °C, 80 °C, 90 °C), and then placed in a reactor and aging at 100 °C for 12 hours. After that, the product was washed with distilled water and filtered. When the pH value was neutral, it was dried in an oven at 105 °C for 2 h and then put into a drying vessel for further investigation. The reaction conditions of AM-HS modification of FA are shown in Table 2.

Characterization

The modified FA samples were dried under ultraviolet lamp. The wavenumber ranges from 4000–400 cm⁻¹ at a resolution of 4 cm⁻¹ using a Nicolet-600 model FT-IR. All the samples were mixed with KBr by the weight ratio of sample to KBr was 5:1 using a KBr tableting process before analysis.

The modified FA samples were added with some water and placed in the ultrasonic instrument. The ultrasonic power was set at 200 W, 400 W, 600 W and 800 W, and the samples were taken out after 15 min. Then, the average particle size of the samples was measured by laser particle size analyzer (Mastersizer 3000), and the coating bonding strength was analyzed by particle size distribution.

The particle size of the original 600 mesh FA, the FA pretreated by carbon removal and iron removal and the FA modified by AM-HS were tested by laser particle size analyzer (Mastersizer 3000). First, the sample test container was cleaned with alcohol, and ultrapure water was used as the dispersion medium. Then, FA is added and stopped when the refractive index is about 6 %–7 %, and the particle size test is started.

The brightness, a* value, b* value and L* value of FA were tested by L&W brightness meter (Elrepho 070). Place the original FA, the de-carbon and de-iron FA and the AM-HS modified FA in the powder brightness determination container, respectively, then use a glass cover sheet to press out a flat and firm surface, and measure the brightness. Each group was tested five times, and then the average value was given in this work.

Results and discussion

Pretreatment for carbon and iron removal

The color of FA mainly depends on unburned carbon particles and crystal minerals, especially iron bearing minerals, the chemical composition of the original FA is shown in Table 1. The carbon particles in FA are the remaining unburned carbon due to insufficient coal combustion, which is related to the gray and black color of FA, and the specific color depends on its content. Other important low-temperature mineral components, especially iron oxides, also show different colors. For example, trivalent iron is brown, red or yellow, and ferrous iron is light green or gray with a blue tone. Magnetite containing bivalent and trivalent minerals is black, and some finely dispersed particles can also be brown. Removal of these chromogenic substances from FA is a necessary step to improve the brightness of FA (Song et al. 2020). In order to improve the brightness of the original FA, the FA was pretreated with high temperature carbon removal and acidic iron removal.

High temperature carbon removal process: the brightness of FA intercepted on 600 mesh screen reaches 38.27 %ISO from 18.01 %ISO at 600 °C, which is closely related to the oxidation of free carbon particles in FA and the degradation of some minerals (Song et al. 2020). Acid

Table 3: Analysis of chemical components of FA before and after iron removal.

Component	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	TiO ₂	Na ₂ O	MgO	Cl	Others
Before iron removal (%)	49.08	34.6	4.97	4.89	1.47	0.91	0.51	0.04	3.53
After iron removal (%)	55.24	35.71	3.76	1.09	1.45	0.84	0.41	0.24	1.26

Figure 1: Effect of alkali ash ratio on brightness of the modified FA.

iron removal process: the FA was pretreated after high temperature decarbonization, and the process of acid iron removal was optimized by response surface method. The optimal process conditions were as follows: the mass concentration of hydrochloric acid was 15 %, the acid leaching time was 0.75 h, the reaction temperature was 74 °C, and the brightness of FA was finally increased to 43.92 %ISO. Under this condition, the brightness of FA increases obviously, which is not only the embodiment of the advantages of acid iron removal, but also the reason why there are more iron oxides in this batch of FA (Song et al. 2020). The chemical composition changes of FA before and after iron removal are shown in Table 3.

Compared with the increase range of FA brightness by high temperature decarbonization, the increase of FA brightness by acid iron removal is relatively low. However, considering the negative effect of iron oxide on subsequent AM-HS modification of FA, the pretreatment of iron removal from FA becomes more important.

Effect of alkali ash ratio on brightness of modified FA

In the process of AM, all the aluminosilicate minerals (including impurities) in the raw materials can be converted

into soluble aluminosilicates and after adding water, it transforms into amorphous aluminosilicate gel, and finally forms a new zeolite structure through HS reaction. NaOH is the activator for the conversion of natural zeolite into aluminosilicate during the melting process. It also plays the role of adjusting alkalinity and providing Na⁺ during the crystallization process, so the amount of NaOH has a significant impact on the entire modification process (Murayama et al. 2002, Xiong et al. 2019, Yang et al. 2019, Kirdeciler and Akata 2020).

The amount of sodium hydroxide is an important factor affecting the HS modification of FA by AM. It can be seen from Figure 1 that the brightness of modified FA is gradually increased from 47.26 %ISO to 64.43 %ISO with the increase of alkali consumption before the alkali ash ratio (the mass ratio of sodium hydroxide and fly ash) is 1:1. This is because with the increase of sodium hydroxide content, the glass phase structure of FA in AM process is destroyed more seriously, more MS are synthesized on the surface of FA, and the coating of FA is more thorough. During this process, it can be seen from the change of the L* value that the total amount of light reflected by the modified FA is gradually increasing. This is due to the fact that newly formed MS are continuously coated on the surface of FA, which changes the surface structure and shape of FA,

Figure 2: Effect of HS temperature on brightness of modified FA.

and the corresponding high reflectivity characteristics of MS cause more light reflection. However, with the further increase of sodium hydroxide ratio, the brightness and L^* value of modified FA also began to decline, the reason is that sodium hydroxide reacts with some color developing metal ions (Fe^{2+} , Fe^{3+}) and the resulting $\text{Fe}(\text{OH})_2$ and $\text{Fe}(\text{OH})_3$ have high light absorption coefficient, which can absorb visible light in some bands and show other colors, thus affecting the brightness of modified FA.

Effect of HS conditions on the brightness of modified FA

Mix the fly ash after AM with a certain amount of deionized water so that the Si and Al in the FA can be dissolved under alkaline conditions to form aluminosilicate gel, aluminosilicate gel is transformed into MS crystals under certain temperature, alkalinity and pressure conditions. At the same time, the alkaline environment of the solution can promote the dissolution of Si and Al in the fly ash, and the Na^+ in the solution can balance the negative charge on the MS framework and accelerate the synthesis of the MS. The main influencing factors of this process are HS temperature and HS time.

HS temperature

The influence of different HS temperature on the brightness of FA in the process of AM-HS modification is shown in Figure 2. With the increase of HS temperature, the brightness of modified FA is gradually increased. When

the HS temperature increases from 50 °C to 80 °C, the brightness of modified FA increases from 49.58 %ISO to 64.43 %ISO. This is because in the process of HS modification, the increase of temperature reduces the need to overcome the energy barrier of nucleation and crystallization before the formation of MS, so that more high brightness MS are coated on the surface of FA, thus improving the brightness performance of modified FA. At the same time, the surface structure of FA is changed, the diffuse reflection of light is improved, and the brightness of modified FA is improved to a certain extent. In addition, the total light reflection of the modified FA increases with the increase of HS temperature. The optimal HS temperature is obtained at 80 °C. At this time, the coating effect of MS on the surface of FA is better, the surface morphology of fly ash is also rougher, which increases the amount of diffuse reflection light, so that the brightness of modified FA reaches the best. Through the absorption of light, the light scattering is reduced, resulting in red and yellow phase in the white color band of subsequent modified FA, which affects the brightness performance of fly ash.

HS time

It can be seen from Figure 3 that in the HS process, when the reaction time increases from 4 h to 6 h, the brightness and lightness L^* of the modified FA are also continuously improved. This is because if the reaction time is too short, some of the gels can't be fully nucleated and crystallized to form the MS on the surface of the FA. When the HS time is more than 6 h, most of the gelatin is nucleated and crystallized to form a MS on the surface of the FA. However, when

Figure 3: Effect of HS time on the brightness of modified FA.

Table 4: Factors and levels of reaction conditions for orthogonal experiment design.

Level	Value of each factor		
	Sodium hydroxide : FA	HS time/h	HS temperature/°C
1	0.8:1	6	70
2	1:1	7	80
3	1.2:1	8	90

the reaction time is too long, the color matter dissolved in the HS reaction will react with the strong base to form different color precipitates and adsorb on the surface of the MS, which will affect the brightness of the modified FA. Although Fe^{3+} and Fe^{2+} on the surface of fly ash have been removed by acid removal, with the extension of the reaction time, Fe^{3+} and Fe^{2+} in the deep layer of fly ash dissolve and can react with NaOH to form $\text{Fe}(\text{OH})_3$ and $\text{Fe}(\text{OH})_2$. Adsorption on the surface of fly ash causes its brightness to decrease.

Orthogonal analysis of influence of reaction conditions on brightness of modified FA

According to the results of single factor experiment. The mass ratio of FA to sodium hydroxide, HS temperature and HS time, were selected to optimize the AM-HS modification process of FA with three factors and three levels by orthogonal experiment table $L_9(3^4)$ aiming at improving brightness of modified FA. The selection of factors and levels of orthogonal experiment is shown in Table 4. The orthogonal experimental design and experimental results are shown in Table 5, Table 6 and Figure 4 respectively.

Table 5: Orthogonal experimental design and experimental results.

	Sodium hydroxide : FA	HS time/h	HS temperature/°C	Brightness/%ISO
1	0.8:1	6	70	55.22
2	0.8:1	7	80	53.35
3	0.8:1	8	90	49.89
4	1:1	6	80	64.44
5	1:1	7	90	62.32
6	1:1	8	70	58.24
7	1.2:1	6	90	50.47
8	1.2:1	7	70	52.33
9	1.2:1	8	80	51.27

Table 6: Range Analysis of Modified FA brightness.

	Sodium hydroxide : FA	HS time/h	HS temperature/°C
k1	52.82	56.71	55.26
k2	61.67	56.00	56.35
k3	51.36	53.13	54.23
R	10.31	3.58	2.12

According to the analysis of orthogonal experiment results in Table 5 and Table 6, the effect of alkali ash ratio, HS temperature and HS time on the brightness of modified FA is as follows: alkali ash ratio > HS time > HS temperature.

Figure 4 shows the trend chart of range difference between each factor and modified brightness of FA.

It can be seen from the results of orthogonal experiment that the brightness of modified FA first increases and then decreases with the increase of alkali ash ratio and HS temperature, and decreases with the extension of HS time,

Figure 4: The influence of various factors on the brightness of modified FA.

which is consistent with the trend diagram of single factor experiment.

The results of orthogonal experiment show that the increase of alkali ash ratio has the greatest effect on the brightness of modified FA, and the time and temperature of HS modification have smaller effect on the brightness of modified FA. This means that in the process of AM-HS modification of FA, the amount of alkali used is the key factor, which needs to be strictly controlled. The influence of HS time and HS temperature on the brightness value of FA is relatively small. The appropriate conditions can be selected according to the actual needs and energy conservation within the appropriate range.

To sum up, the optimum process conditions of AM-HS modification of FA are as follows: alkali ash ratio is 1:1, HS time is 6 h, HS temperature is 80 °C, under these conditions, the brightness of modified FA is 64.54 %ISO.

Analysis of surface properties of modified FA under optimum conditions

Particle phase composition of AM-HS modified FA

XRD pattern analysis was performed on the pretreated FA after carbon removal and iron removal and the modified FA after AM-HS to determine the phase changes in the AM-HS modification of FA. The XRD pattern results are shown in Figure 5.

It can be seen from the above figure that the broad diffraction peak between 22–35° in the pretreated FA dis-

Figure 5: Phase analysis of thermally modified FA in alkali molten water.

appears, which means that the vitreous phase material is transformed into a new material. The five diffraction peaks of its diffraction angle $2\theta = 7.18^\circ, 10.16^\circ, 23.98^\circ, 27.11^\circ, 29.02^\circ$, can prove that the surface of the FA does have 4A MS after the AM-HS modification. Among them, 4A MS is a kind of crystalline sodium aluminosilicate, the crystal type is mainly $\text{Na}_{12}\text{Al}_{12}\text{Si}_{12}\text{O}_{48}(\text{H}_2\text{O})_{27}$, and the most basic structure is $[\text{SiO}_4]^{4-}$ tetrahedron and $[\text{AlO}_4]^{5-}$ tetrahedron. The diffraction angle $2\theta = 6.13^\circ, 11.74^\circ, 15.45^\circ, 32.03^\circ, 32.64^\circ$, etc. The existence of these characteristic peaks also proves the formation of octahedral MS on the surface of FA.

Figure 6: Infrared analysis spectrum of FA under different treatment levels.

Changes in surface functional groups of FA particles modified by AM-HS

In order to analyze the changes of functional groups on the surface of FA after AM-HS, FT-IR was used to detect the original FA, the FA after carbon removal and iron removal, and the FA after AM-HS modification. The results are shown in Figure 6.

Among them, the original FA has the same performance as the FA after the pretreatment of carbon removal and iron removal, and there is no new absorption peak. The AM-HS modified FA is the hydroxy stretching vibration peak of adsorbed water in the spectral region between $3400\text{--}3500\text{ cm}^{-1}$, and the O–H bending vibration absorption peak of adsorbed water between $1600\text{--}1700\text{ cm}^{-1}$. The intensity of these two peaks is more obvious than that of the former two, which is related to the porous adsorption water characteristics of the formed MS. At the same time, the absorption peaks near 556 cm^{-1} and 770 cm^{-1} are mainly due to the superposition of symmetric stretching vibration of Si–O and Al–O bonds in 4A MS, and the strong absorption peak near 1005 cm^{-1} is the skeleton vibration spectrum of MS, which is mainly caused by the asymmetric stretching vibration of Si–O–Al bond in Si–O–Al tetrahedron. The three absorption peaks are all vibration characteristic peaks of standard 4A MS.

Changes in particle size of AM-HS modified FA

Figure 7 and Table 7 compare and analyze the original particle size of FA under 600 mesh, the particle size of FA

Figure 7: The change in particle size of FA under different treatment levels.

Table 7: Average particle size of original FA, carbon and iron removal FA, AM-HS modified FA.

Sample	Original 600 mesh FA	Carbon and iron removal FA	AM-HS modified FA
Average particle size/ μm	22.2	21.9	22.5

pretreated by carbon removal and iron removal, and the particle size of FA modified by AM-HS. The particle size of the FA after carbon removal and iron removal slightly decreases, because the original FA contains a lot of free carbon particles. The size of these carbon particles is larger than other particles, so the particle size of FA decreases slightly after the oxidation of high temperature carbon particles. But after subsequent HS modification, the particle size of FA increases. This is due to the fact that the surface of the FA is coated with porous MS after AM-HS modification, thereby increasing the particle size of the FA particles.

Analysis of particle size of AM-HS modified FA coating

It can be seen from Figure 8 that the average particle size of the modified FA does not change significantly with the increase of vibration strength after AM-HS modification. This is because the MS formed on the surface of FA and the base of FA not only have physical adsorption due to the porosity of MS, Moreover, there is chemical bond between the gel produced by HS process and the internal material

Figure 8: The change in particle size of modified FA under different oscillation intensities.

of FA, which makes the final formed MS and FA bonded firmly. The particle size of modified FA has little change under ultrasonic vibration.

Changes in the surface structure of AM-HS modified FA

It can be seen from Figure 9 that the original 600 mesh FA is mainly composed of irregular particles and spherical particles with smooth surface. After carbon and iron removal pretreatment, the irregular substances in FA are reduced. Then, after AM-HS modification, the surface of FA formed cubic 4A MS crystal with complete morphology and clear edges and other MS crystal types with high brightness performance. These MS have achieved dense coating on the surface of spherical FA. Compared with the original smooth surface of FA, the surface of FA coated with MS is more rough, irregular and uneven. This surface morphology can not only reduce the specular reflection of light, but also increase the diffuse reflection of light, which is helpful to improve the brightness performance of modified FA, but also greatly improve the adsorption capacity of modified FA due to the porous characteristics of MS on the surface of FA.

Conclusion

In this study, high brightness 4A MS was formed on the surface of FA using AM-HS modification technology to realize the coating of FA and improve the brightness perfor-

Figure 9: Scanning electron micrograph of FA modified by different treatments.

mance of modified FA. Under the optimal process conditions (alkali-ash ratio: 1:1; HS temperature: 80 °C; HS time: 6 h), the brightness of FA after coating with MS increased from 43.92%ISO to 64.44%ISO. Among them, the alkali ash ratio has the greatest influence on the synthesis of 4A MS. XRD, FT-IR and SEM analysis all proved the formation of 4A MS on the surface of FA; ultrasonic vibration and particle size analysis showed that the generated MS and FA had a strong interaction, and the MS would not fall off the surface of fly ash. This method uses the Si and Al of FA itself as raw materials to prepare MS on the surface of FA. Compared with traditional MS preparation methods, this method saves a lot of Si and Al raw materials, and provides

a novel and effective method for improving the brightness of FA. The improvement of the basic brightness of FA also provides the possibility for the CaCO_3 coating to achieve higher brightness after AM-HS modification.

Funding: This work was supported by Natural Science Foundation of Guangdong Province, China (No. 2019A1515012188); Science and Technology Plan Projects of Guangdong Province, China (No. 2016A020221040); and State Key Laboratory of Pulp and Paper Engineering, China (No. 2016C04).

Conflict of interest: The authors declare no conflicts of interest.

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