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Effect of Bulking Agents on Dewatered Sludge Biodrying Followed by Thermal Drying

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Abstract: As an exogenous substance, bulking agent plays a considerable role in dewatered sludge (DS) biodrying by affecting the organics degradation and water migration. Meanwhile, biodried product needs to be further dried because its final moisture content (MC) is usually too high for direct incineration. In this study, the influence of adding different bulking agents on organics decomposition and extracellular polymeric substances (EPS) release during DS biodrying, and thermal drying characteristics of the biodrying mixtures was investigated. Results showed that spent coffee ground (SCG), corncob (CC) and wheat husk (WH) improved water removal ratios of DS biodrying. SCG contributed various types of organic matter to the biodrying matrix, thus attaining the longest high-temperature period (4.5 days). Meanwhile, SCG contributed lipids, which have high calorific value, to the biodrying pile, leading to the highest biodrying index (3.70) among all treatments. SCG, CC and WH also accelerated EPS delamination, which was beneficial for microbial activity and bound water release. Besides, the biodried product with SCG was endowed with the highest C content and calorific value. Furthermore, bulking agents improved the thermal-drying rates of the feedstocks, and intensified the positive impacts of biodrying on the thermal-drying characteristics of the mixtures. In particular, the biodried product with SCG had a high drying rate even with low residual water content in the mixture.

Keywords: dewatered sludge; biodrying; bulking agent; organics degradation; thermal drying characteristics

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1. Introduction

Nowadays, biodrying derived from composting has been developed as an environmentally friendly treatment method for maximizing the energy recovery of organic wastes with high moisture content (MC), including dewatered sludge (DS) [1]. During biodrying, water is removed by bio-heat generated from microbial degradation of various types of organic matter in aid of forced aeration. The biodried product can be used as refuse-derived fuel (RDF), which is a carbon-neutral and renewable energy alternative to fossil fuel source [2]. Thus, biodrying is considered to be a prospective DS treatment method for RDF production aiming for a circular economy with minimal energy consumption.

DS was characterized by rich microbes, mainly composed of proteins, resulting in a monotonous biochemical composition and limited biodegradable organic matter content [3]. Besides, due to the poor gas permeability caused by its high MC (80–85%) and low porosity, DS is commonly biodried after undergoing some pretreatment [4,5] or mixing with bulking agents [6,7]. When a bulking agent was added to DS, its physicochemical properties could play important roles and affect the organic matter degradation for bio-heat generation during biodrying [3,8,9]. What is more, the slurry-type DS is flocculated by multilayered extracellular polymeric substances (EPS), which are secreted by microbes in consumption of organic matter and considered crucial for dewaterability of sludge [10]. Besides, EPS can be used to serve as carbon or energy sources for the microbes in conditions of short nutrient supply [11]. Hence, investigating the transformation of EPS is crucial for understanding the effects of bulking agents on the organic matter biodegradation patterns during the biodrying process. Cai et al. [10] investigated the EPS variation during different phases of DS biodrying with sawdust (SD) as a bulking agent. However, few studies have made a thorough inquiry into multi-layered EPS profiles of DS biodrying processes with different bulking agents.

The final MC of the biodried product is important because it influences the low heating value (LHV), which represents the energy content and quality of the RDF [12]. However, the final MC of the biodried product strongly depends on biodrying modality (e.g., ambient temperature, turning frequency, residence time, etc.) and characteristics of raw materials (e.g., initial MC, organic matter content and biodegradability, porosity, etc.) [5,13]. As reported, the MC of biodrying material composed of DS and spent coffee ground (SCG) decreased from 63.27% to 31.20% during 8-day biodrying at an average ambient temperature of 25 °C [8]. The final MC of biodried product from 20-day DS biodrying with corncob (CC) as a bulking agent conducted at around 28 °C was 32.53% [13]. A lower final MC of less than 25% could be achieved by biodrying of municipal solid waste from an initial MC of 40% [14]. Although the MC for feedstock of DS biodrying could be amended by pretreatment or adding bulking agents, it should not be too low to avoid excess energy consumption on pretreatment or a high proportion of bulking agent in biodrying pile. On the other hand, matrix MC lower than 50% might inhibit microbial activity, which is crucial for effective bio-heat generation and water removal [15]. In practice, the initial MCs of biodrying matrixes were usually adjusted to 50–70% [5,13], and the final MCs of DS biodrying products were higher than 45% in spite of the long biodrying period (2–3 weeks) [5,9,16]. It was reported that the MC of organic wastes for sustainable combustion and RDF production were 25–30%. Therefore, MCs of the DS biodrying outputs were too high for subsequent utilization in most cases.

In order to improve the quality of the biodried product, Ma et al. [15] proposed an externally thermal assistant strategy in the cooling stage of biodrying to stimulate microbial activity and increase water removal. The MC decreased further from 45.15–49.42% to 15.20–25.85% after a 6-day thermal assistance period. Thermal drying has been well developed and widely accepted as a fast and controllable method, but this process consumes much energy when it is directly used to remove moisture from DS on account of its structural features and high bound water content [5]. It was verified that SD addition improved the thermal drying characteristics of DS because of higher total exchange surface areas [17,18], and SD was also commonly used as a bulking agent in organic waste biodrying [9]. Besides, Cai et al. [10] proposed that biodrying resulted in a multi-porous micro-structure and the transformation from bound water to free water of biodrying samples, which implied that the thermal drying characteristic of the biodried product might be improved. Up to now, however, little research has focused on the effect of bulking agent addition and the subsequent biodrying process on thermal drying characteristics of the matrix and the feasibility of thermal drying as a further drying strategy for biodried products.

In light of the above, the main objectives of this study were to (1) systematically investigate the influence of frequently-used bulking agents, SCG, CC, bagasse (BA), wheat husk (WH) and SD on DS biodrying performance, (2) reveal the biodegradation patterns of organic matter and the changes of multi-layered EPS component distribution during the biodrying process, (3) evaluate the quality of the biodried products, and (4) study the drying characteristics of biodrying feedstocks and biodried products amended with various bulking agents. Results from this study could provide important insights into organic matter decomposition during DS biodrying with various bulking agents and useful information for DS biodrying-thermal drying treatment aiming at energy recovery.

2. Materials and Methods

2.1. Materials

The DS was collected from the wastewater treatment process of Liwayway Co., Ltd. (Shanghai, China) and stored in the refrigerator at 4 °C before use. In order to unify the initial MCs and particle sizes of the biodrying feedstocks, all of the bulking agents were thermally dried, crushed, and screened to the size of 0.15–0.2 mm. The characteristics of raw materials used in this study are presented in Table 1.

Table 1. Characteristics of raw materials used in this study.

	DS	**SCG**	**CC**	**BA**	**WH**	**SD**
MC (%)	91.26	0	0	0	0	0
VS (%, dry basis)	78.04	97.54	96.12	94.70	89.80	95.17
Organic components (mg·g^{-1} TS)						
Hemicellulose	104.51	122	190.56	159.17	127.85	141.49
Cellulose	18.01	121.23	300.54	467.16	324.34	352.65
Lignin	27.97	348.83	153.42	362.22	155.33	171.94
Proteins	290.65	138.00	95.06	26.32	143.02	125.93
Lipids	24.27	135.14	13.55	10.56	26.95	16.20
Amylums	115.09	153.57	140.08	151.56	177.66	165.33
Dissolved organic matter (DOM) (mg·g^{-1} TS)						
Proteins	26.58	16.36	11.63	11.01	5.46	7.98
Polysaccharides	7.76	12.17	20.07	12.03	6.89	4.76
Reducing sugars	1.73	6.43	8.42	3.30	0.77	0.82
Others						
Electric conductivity (EC)	89.3	84	170.0	84	130.4	135.9
pH	6.81	6.53	5.21	5.74	6.69	4.01
Dissolved chemical oxygen demand (DCOD) (mg·g^{-1} TS)	77.56	88.78	91.93	17.49	186.12	22.39
NH_3-N (mg·g^{-1} TS)	3.57	3.34	3.70	0.39	19.34	12.09
Elemental content (%, dry basis)						
C	34.77	50.00	41.60	44.10	39.00	42.53
N	5.47	2.49	0.94	0.40	1.63	5.97
H	5.12	6.27	5.41	5.15	5.42	5.30
S	0.82	0.16	0.07	-	0.16	-
O	31.90	38.60	48.10	45.00	43.60	41.37

2.2. Experimental Equipment and Operation

The lab-scale biodrying reactor was a 4.375 L styrofoam box (L × W × H: 300 × 225 × 275 mm) with a wall thickness of 50 mm proposed by Yang et al. [19]. The temperature of the biodrying pile was detected using a digital thermocouple installed in the middle of the reactor. Room air was pumped into the reactor by using an air pump (S-60B, Saier Co., China), and the air was diffused into the biodrying pile through aeration stone balls loaded at the bottom of the reactor (Figure 1). In this study, the DS and bulking agents were crushed into fine particles ($\leq$0.2 mm) to ensure homogeneous feedstock properties and sampling. This strategy was proved to be effective by previous research [5], and no replicates were conducted for each experimental condition.

Biodrying feedstock was prepared by manually mixing DS and each bulking agent with a mixing ratio of 3:1 (*w*/*w*, wet basis). The MC of the feedstock was about 70%. As a control check (trial CK), DS was air-dried at room temperature for several days to reach MC of ~75% for biodrying directly without bulking agent. In each trial, 1.2 kg of mixture was added to the above-mentioned reactor to conduct the biodrying process. The operating period for biodrying was 12 days, during which the air flow rate was controlled by a gas-flow meter (LZB-3, 0.1–1.0 L·min^{-1}) at the rate of 0.4 L·min^{-1} (0.06 m^3 kg·TS^{-1}·h^{-1}). The biodrying piles were weighed and manually turned every three days to regulate temperature and moisture uniformity of the materials, after which samples were collected and stored in a 4 °C refrigerator for analysis.

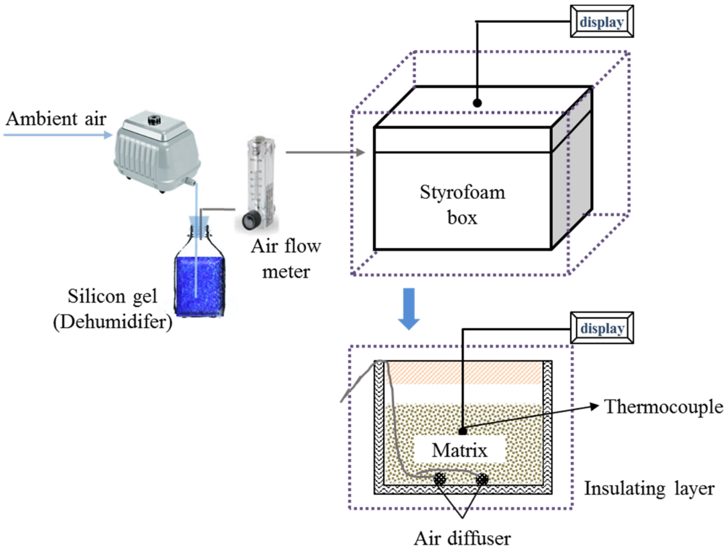


Figure 1. Experimental setup for biodrying.

2.3. Sample Analysis

2.3.1. General Indicator Analysis

The MC was measured by drying the sample in an oven (DHG-9145A, Yiheng Scientific Instrument Co., Shanghai, China) at 105 °C for 24 h. The VS content was determined by burning the dried sample at 550 °C for 6 h in a muffle furnace (KSL-1100X, Kejing Material Technology Co., Beijing, China) [20]. A fresh sample (1 g) was diluted with deionized water (1:10, *w*/*v*) and vortexed for 30 s. The pH and EC values were measured in the supernatant suspension by using a digital pH meter (ST2100, Changzhou Ohaus Instrument Co., Changzhou, China) and a conductivity meter (DDS-307, Inesa Scientific Instrument Co., Shanghai, China), respectively. The free air space (FAS) was calculated by the reported method [5]. The elemental compositions of C, N, H, and S were analyzed using an elemental analyzer (Unicube, Elementar, Germany), and the percentage content of O was calculated by subtracting the sum of C, N, H, and S from VS/TS [21]. The high heating value (HHV) was measured using an oxygen bomb calorimeter (SDC712, Sundy Science and Technology Co., Changsha, China), and LHV was calculated according to He [22]. All measurements were conducted in triplicate.

2.3.2. Organic Composition Analysis

For determination of organics compositions of biodrying materials, the samples were dried at 60 °C and pulverized to less than 0.2 mm. The proteins were estimated from the Kjeldahl nitrogen content multiplied by 6.25 [3]. The lipid content was measured gravimetrically after the lipids were extracted with the mixture of chloroform and methanol (2:1, *v*/*v*) from the dried sample, and the solvent was removed using a rotary evaporator (W2-100SP, SENCO Technology Co., Shanghai, China) [23]. The amylum content was determined using the enzymatic hydrolysis method described by Hao and Jahng (2019) [8]. The lignocellulose (hemicellulose, cellulose, and lignin) was analyzed according to the standard method [24] and Goering et al. [25].

2.3.3. EPS Extraction and Three-Dimensional Excitation Emission Matrix (3D-EEM) Analysis

The extraction of soluble EPS (S-EPS), loosely bound EPS (LB-EPS), and tightly bound EPS (TB-EPS) were performed as described by the previous literature [26] with a slight modification. A 1-g fresh sample was mixed with 25 mL of deionized water and vortexed for 15 s. Then the mixture was stirred magnetically at 150 rpm for 10 min and centrifuged (4000× *g*, 15 min) at 4 °C to separate S-EPS in the sludge supernatant. Next, the pellet left in the centrifuge tube was re-suspended in 25 mL of 0.05% (*w*/*w*) NaCl solution, which was pre-heated to 70 °C. Then the suspension was thoroughly mixed with a vortex mixer for 1 min without any delay, followed by centrifugation at 4000× *g* at 4 °C for 10 min. The organics in the supernatant were regarded as LB-EPS. Afterwards, the pellet left in the centrifuge tube was re-suspended in 0.05% NaCl solution to a volume of 25 mL. The mixture was heated in a water bath at 60 °C for 30 min, and was then centrifuged at 4 °C at 4000× *g* for 15 min. The supernatant was collected, which was regarded as TB-EPS. The S-EPS, LB-EPS, and TB-EPS extractions were filtered through 0.45 μm nylon membranes.

3D-EEM was performed using a fluorescence spectrophotometer (F-4700 FL spectrophotometer, Hitachi, Japan). In view of the high organic content, the EPS samples were diluted with deionized water to keep the DCOD below 30 mg·L^{-1}. The EEM spectra were obtained with scanning emission (Em) wavelength from 200 to 600 nm at 10 nm intervals by changing the excitation (Ex) wavelength from 200 to 500 nm at 10 nm intervals [27]. The slit widths were fixed at 10 nm for both excitation and emission monochromators, and the scan speed was set to 1200 nm·min^{-1}. The fluorescence response of deionized water was recorded as the blank [28]. The software Origin 2018 (OriginLab, Northampton, MA, USA) was employed to handle the EEM data.

2.3.4. Thermal Drying Characteristics Analysis

The influences of different bulking agents and biodrying on the isothermal drying characteristics of the mixtures were evaluated. The weight loss profile of fresh samples (10 g) before and after biodrying was recorded by a halogen moisture analyzer (HC103, Mettler Toledo Inc., Columbus, OH, USA) at 60 °C. The drying process was continued until the MC did not decrease significantly with increasing drying time. Then, the water loss and water removal ratio at different time intervals were calculated according to Equations (1) and (2), respectively. The water desorption rate (WDR, mg H_2O min^{-1}) closely related with the drying characteristics of the biodrying sample was determined by Equation (3) [3]. The WDR curve was fitted versus the residual water in the mixture (g water 10 g^{-1} wet sample) using the software Origin 2018 (OriginLab, Northampton, MA, USA).

$$m_i = M_{i-1} - M_i \tag{1}$$

$$mr_i = \frac{M_{i-1} - M_i}{m} \tag{2}$$

$$WDR = \frac{1000 \times (M_{i-1} - M_i)}{t_i - t_{i-1}} \tag{3}$$

where, m_i and mr_i are the water loss (g) and water removal ratio, respectively, between time t_i and time t_{i-1} (min). M_i and M_{i-1} are the weight recorded at time t_i and time t_{i-1}, respectively. m is the total weight loss (g) during the drying process, which was regarded as the water content of the sample.

3. Results and Discussion

3.1. Biodrying Performance

Figure 2a shows the temperature profile of the matrix with different bulking agents. As for trial CK, the temperature rapidly increased without a lag phase and reached the first peak value of 47.1 °C on day 0.5. In contrast, in the biodrying pile with bulking agents, lag phases of 0.5 to 2 days were observed, among which SD was the longest. This is consistent with observations of Yang et al. [5] for DS biodrying. These results suggested that the exogenous bulking agents aroused microbial activity to different extents. Each temperature peak was followed by a declining trend except that of trial CK. This was thought to be because the organics in the DS were accommodative with its microbes, resulting in a sustainable release of metabolic heat. After the first turning on day 3, the matrix temperature rebounded for all treatments. In the case of trials SCG and WH, plateaus with high temperatures were observed from day 3 to day 6. However, the second peak values of CK and SD were lower than the first ones, indicating insufficiency of biodegradable organics. It seems that the second turning had no noticeable influence on the bulk temperature of WH, while those of SCG and CC were promoted. The temperatures of SCG, CC, and WH reached the highest values of 67.6 °C (day 6.5), 68.1 °C (day 3.5), and 65.6 °C (day 4.1), respectively. Although similar peak temperatures were obtained in trials SCG and CC, a longer high-temperature period (≥45 °C) was observed in trial SCG (4.5 days). It was thought that biodrying of all the treatments ceased on day 9 because the temperature no longer increased even after the third turning.

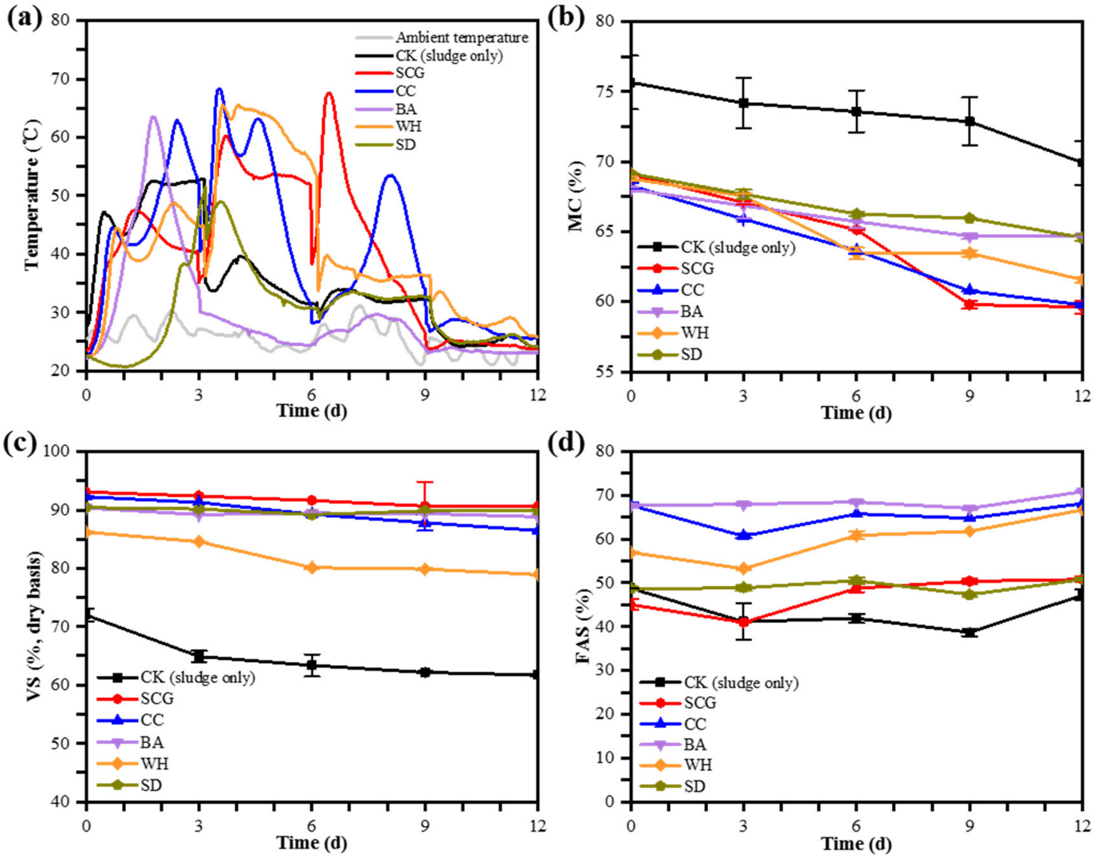


Figure 2. Changes in (**a**) matrix temperature (**b**) MC, (**c**) VS, and (**d**) FAS during biodrying with different bulking agents.

The bulking agents also strongly influenced MC variations during the biodrying process (Figure 2b). After 12-day biodrying, MCs of trials CK, SCG, CC, BA, WH, and SD decreased from 75.65%, 69.09%, 68.29%, 68.02%, 68.78%, and 69.15% to 69.93%, 59.62%, 59.75%, 64.69%, 61.57%, and 64.58%, respectively. In the case of trial SCG, the matrix MC sharply decreased from 65.14% to 59.79%, along with the highest value of matrix temperature from day 6 to day 9. Similar results were also observed in trial WH, whose MC decreased from 67.54% (day 3) to 63.44% (day 6) with a high-temperature plateau (Figure 1a). The water removal ratios of SCG (57.50%), CC (66.39%), and WH (67.03%) were higher than CK (56.94%), indicating that these bulking agents were favorable for biodrying of DS. The MCs of the biodried products in this study were higher than those of many other reports [6,29,30], which might be due to the higher initial MCs, lower air flow rate for ventilation, and/or poor heat preservation of such a small size pile. Nonetheless, the MCs of the final product in those reports (≥45%) were also beyond the limit value (25–30%) for spontaneous combustion and efficient energy recovery [15]. These results suggested that biodrying was efficient for water removal without external thermal energy consumption, but further drying is essential because the final MC was still too high for utilizing the product as RDF.

The VS consumption ratios of trials SCG, CC and WH were 37.35%, 54.30%, and 58.51%, respectively, during the biodrying process (Figure 2c). In case of trial CK, the VS content decreased faster (especially in the initial 3 days) because the feedstock had a low VS content compared to other trials. As water in the matrix is evaporated by metabolic heat produced from VS degradation, biodrying index expressing the mass of water removed per unit of VS (g) was calculated to quantitatively evaluate the biodrying efficiency. Trial CK showed the highest biodrying index (4.85) because the initial MC and free water content in the matrix was higher, which was easily carried by the forced air. Nonetheless, the water removal ratio of trial CK was lower than those of trials SCG, CC, and WH. Although trial SCG showed a lower water removal ratio than trials WH and CC during 12-day biodrying, its biodrying index was obviously higher (3.70, 2.93, and 2.85 for trials SCG, WH, and CC, respectively), indicating more water was removed with less combustible fractions consumed and more energy was retained in the biodried product of trial SCG. Meanwhile, the biodrying material of trial SCG on day 9 was observed with higher VS content (90.67%) and lower MC (59.79%), which is beneficial for further utilization and energy recovery. SCG is also reported to be a special material for the synthesis of advanced materials for environmental sustainability [31–34]. Biodrying indices in this study were relatively lower than in other studies, which might be due to the differences in feedstock properties and biodrying operation parameters. Nonetheless, this study demonstrated that SCG was an excellent bulking agent for DS biodrying in terms of both water removal and VS conservation.

It was also noticed that the initial FAS (Figure 2d) of trial SCG (45.07%) was lower than trial CK (48.75%) while trial SCG showed better performances in DS biodrying. Meanwhile, the fluffier matrix of trials WH and CC had higher FAS, while the biodrying indices were lower compared to trial SCG. The reason might be that excess pore volume resulted in longer air-flow channels in the materials, which is unfavorable for water transfer. These results indicated that, in addition to biodegradable organics, adequate FAS is also important for an efficient biodrying process.

3.2. Biodegradation Patterns of Organic Matter

In order to investigate VS consumption in detail, organic matter including proteins, lipids, amylums, hemicellulose, cellulose, and lignin in the biodrying materials were determined. As Figure 3 shows, the biodegradation pattern of organic matter during the 12-day biodrying process was obviously influenced by the bulking agents. In the initial three days, proteins decreased much (18.80 g) in trial CK because it was the main organic matter occupying 42.05% (dry basis) of the feedstock. Besides, carbohydrates including amylums and hemicellulose were also degraded by 56.60% (16.96 g) and 49.59% (16.20 g), accounting for 13.52% and 12.90% of the VS losses, respectively. With respect to trial SCG, however, various types of organic matter including hemicellulose (11.65 g), lignin (9.15 g), proteins (5.02 g), lipids (5.04 g), and amylums (6.05 g) were consumed for activation of the microbes at the beginning of biodrying. Hemicellulose was dominantly consumed in trials CC (24.83 g), BA (16.22 g), WH (12.29 g) and SD (13.48 g) in the initial three days, indicating that hemicellulose was more easily biodegraded compared to other lignocellulose components. It was observed that in trial WH, proteins (9.75 g) and amylums (7.40 g) also decreased obviously before the first turning because wheat bran is rich in amylums and proteins (Table 1).

Proteins decreased by 11.84 g in trial CK from day 3 to day 6 and were dominantly consumed until day 12. The total degraded mass of proteins in trial CK was 41.10 g, accounting for 38.53% of the consumed organics during the whole biodrying process. In trial WH, the degradation performance of proteins (15.75 g) was superior to other trials from day 3 to day 6. Meanwhile, the degraded masses of amylums (19.20 g), hemicellulose (16.18 g), and cellulose (22.05 g) in this period were also the highest during the whole biodrying process, which was in agreement with the temperature evolution (Figure 1a). After the second turning, amylums, and hemicellulose were the main substrates for maintaining moderate matrix temperature from day 6 to day 9. In the case of trial SCG, all the organic matter degraded simultaneously, and the degradation capacity of lignin was the highest, with a degraded mass of 38.97 g from day 3 to day 9. Over the entire biodrying process, lignin was consumed by 48.93 g, accounting for 37.96% of the VS loss, and was the main component consumed in trial SCG. Although lignin is considered to be recalcitrant to biodegradation, the possibility cannot be excluded that lignin contents in the raw materials affect the degradation ratio during biodrying [35]. Similar to trial SCG, various types of organic matter including amylums, lignocellulose, and proteins were biodegraded (Figure 3), and a temperature peak was observed after the second turning in trial CC (Figure 2a). However, the biodegraded mass of lipids in trial CC (0.71 g) was negligible compared to that in trial SCG (4.52 g) from day 6 to day 9. In trials BA and SD, the cooling phase and high-temperature period were observed, respectively, from day 3 to day 6, and lignocellulose was the main component biodegraded. Meanwhile, 7.69 g amylums and 7.36 g proteins were consumed in trials BA and SD, respectively, during this period. After the second turning on day 6, the biodegradation capacity of organic matter became weaker, and their matrix temperature level maintained moderate accordingly. For all treatments, the third turning exerted no influence on the organic matter degradation, which was also reflected in matrix temperature profile. These results suggested that bulking agents regulated the nutrient compositions of biodrying materials and affected the sequence and extent of organic matter biodegradation.

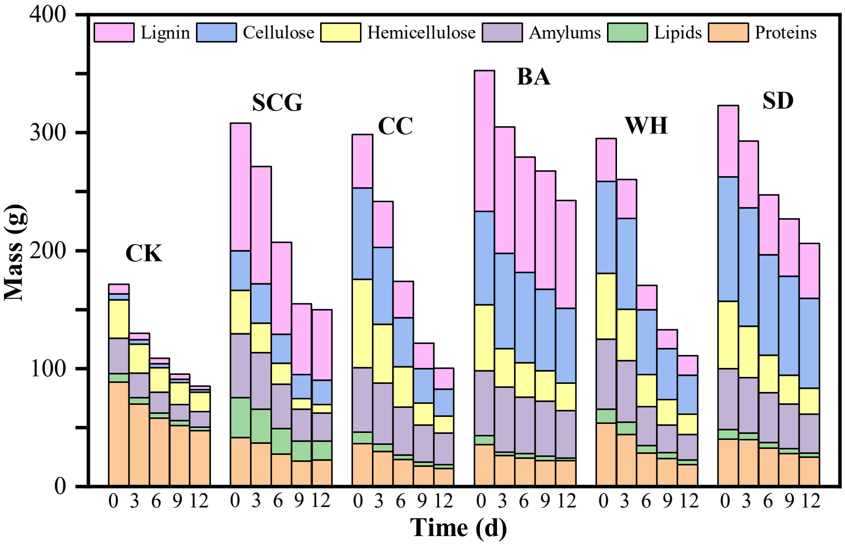


Figure 3. Weight changes of organic matter contained in biodrying piles with various bulking agents.

On the whole, the masses of total organic matter decreased from 171.63 to 84.88 g, 307.95 to 149.75 g, 298.28 to 100.33 g, 359.14 to 220.97 g, 294.95 to 110.96 g, and 323.07 to 206.18 g for trials CK, SCG, CC, BA, WH, and SD, respectively. The main contributors to the VS loss and bio-heat for trials CK and SCG were proteins and lignin, respectively, while those for other trials were hemicellulose and/or cellulose. It is worth noting that lipids were barely biodegraded in all biodrying piles except trial SCG (17.91 g) because the contents of lipids in those feedstocks were low. Moreover, the degradation order and patterns of various types of organic matter were also different in biodrying with various bulking agents. Generally, microbes require an energy source (biodegradable organic-C) and N for their development and activity in aerobic treatment of solid wastes, thus nutritional balance defined by the C/N ratio is important for biodrying [36]. In this case, the types and biodegradation potentials of C and N are also key factors influencing biodrying performances. It can be inferred that the proteins (N source) contained in DS were readily biodegradable because biodrying was successfully conducted using DS only in trial CK. Among the bulking agents used in this study, SCG rich in biodegradable lipids

(135.14 mg·g^{-1} DS, Table 1) regulated C/N ratio by improving lipid content, which was low in DS, of the biodrying feedstock. Consequently, trial SCG showed a more proportional biodegradation ratio for every nutrient component, especially for lipids, compared to other treatments. It is well-known that the calorific value for lipids (39.3 MJ·kg^{-1}) is higher than those of carbohydrates (17.4 MJ·kg^{-1}) and proteins (23.4 MJ·kg^{-1}) [37]. Therefore, the biodrying index of trial SCG was higher than those of trials CC and WH, which showed good biodrying performance at the expense of higher VS losses. These results suggested that the bulking agents affected the biodrying process through their different biochemical components and biodegradability.

3.3. EPS Release and Transformation

EPS are a complex high-molecular-weight mixture of polymers excreted by microbes during the biodrying process. Figure 4 displays the EEM spectra changes of the S-EPS, LB-EPS, and TB-EPS samples collected before and after biodrying. According to Guo et al. [38], the EEM spectra were divided into five regions depending on the Ex/Em wavelengths of the EPS. Peaks at Ex/Em wavelengths of (200–250)/(300–330) and (200–250)/(330–380) are related to aromatic proteins I (tyrosine-like materials, region I) and aromatic proteins II (tryptophan-like materials, region II), respectively. Peaks at Ex/Em wavelengths of (250–400)/(300–380) are related to soluble microbial byproduct-like materials (region IV). Peaks at Ex/Em wavelengths of (200–250)/(380–500) and (250–400)/(380–500) are related to fulvic acid-like (region III) and humic acid-like (region V) substances, respectively.

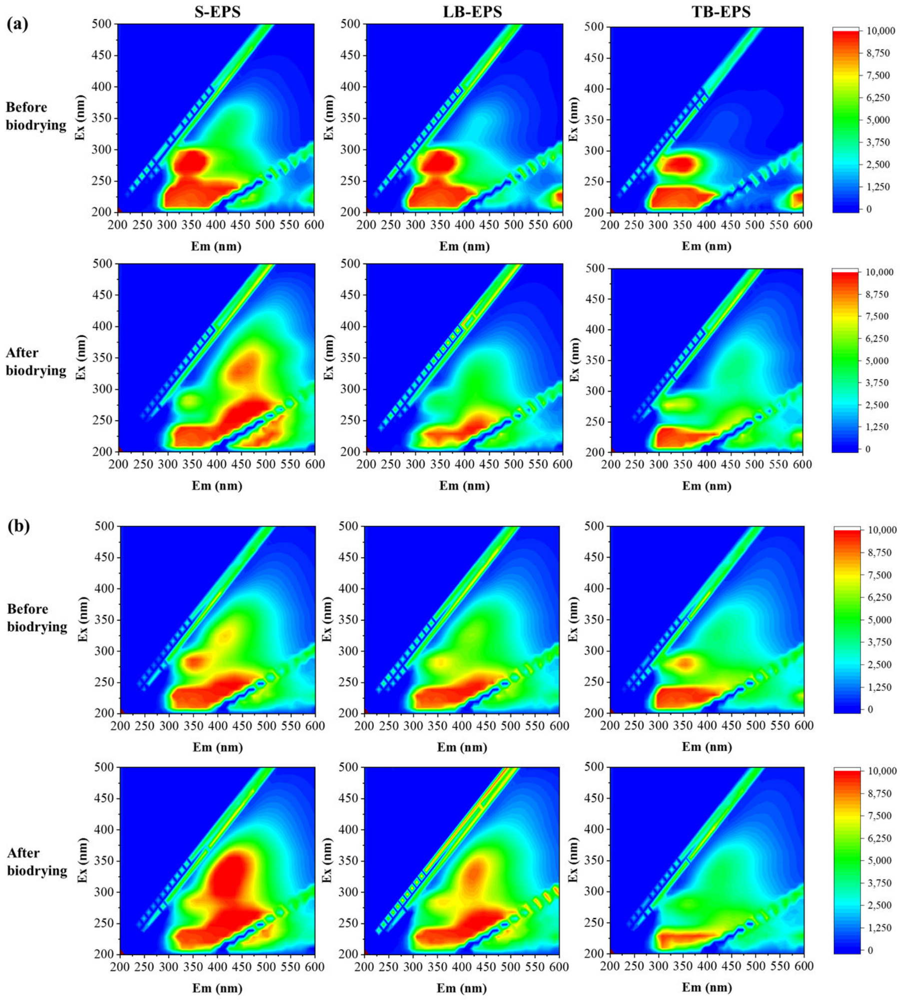
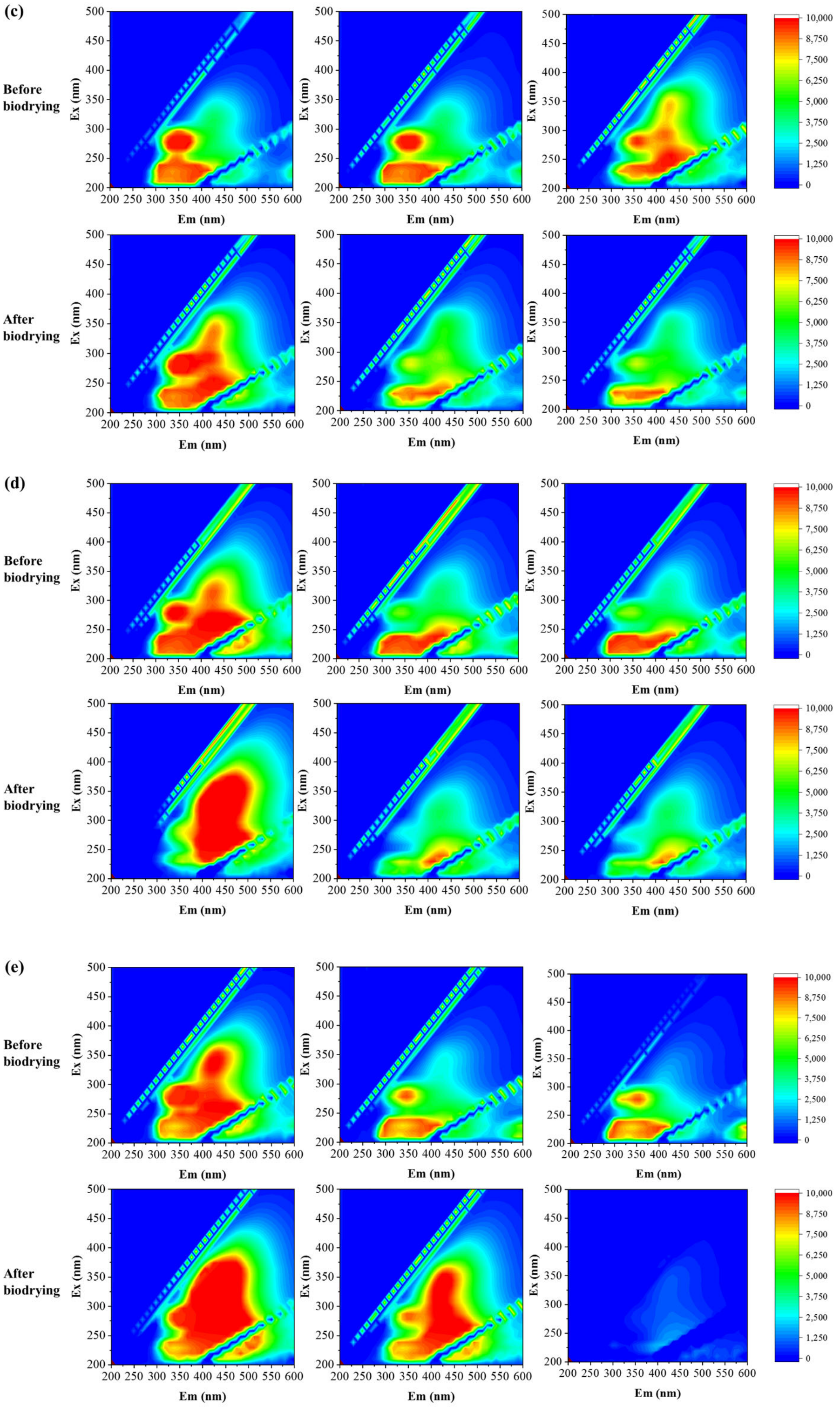


Figure 4. *Cont.*



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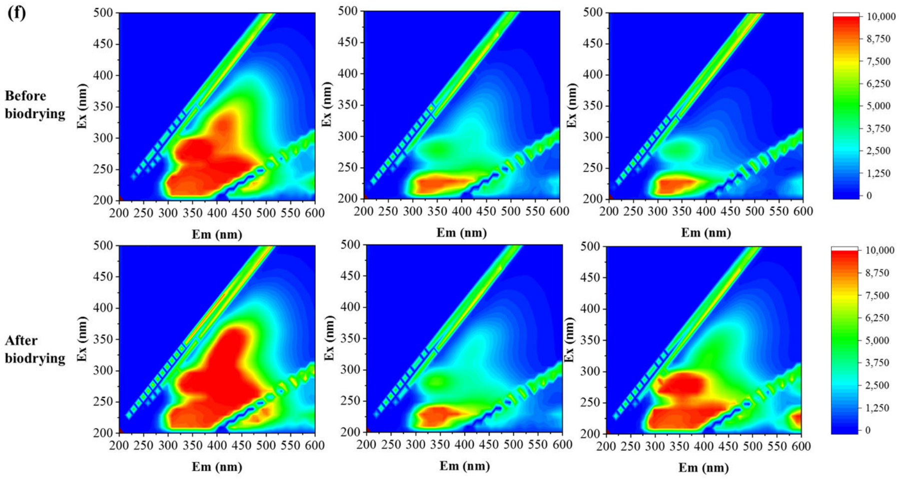

Figure 4. Excitation emission matrix (EEM) profile of EPS fractions in samples before and after biodrying. (**a**) Trial CK, (**b**) trial SCG, (**c**) trial CC, (**d**) trial BA, (**e**) trial WH, (**f**) trial SD.

For TB-EPS, it was observed that the fluorescence signals of regions I, II, and IV decreased in all biodrying piles but that of trial SD, while those of regions III and V were enhanced in all biodrying piles. This result implies that biodrying could promote the transformation of macromolecular organic matter (i.e., proteins and soluble microbial byproduct-like materials) into stable organic matter (i.e., fulvic and humic acid-like substances). However, for trial SD, although the solid organic matter was decomposed and soluble biopolymers were released into TB-EPS, it seemed that these biopolymers were neither able to be utilized as carbon and energy source by the microbes nor released to LB or S-EPS, resulting in the accumulation of organic matter in TB-EPS. The fluorescent spectra of TB-EPS of trial SD revealed the reason for inferior biodrying performance with SD.

For LB-EPS, the fluorescence intensities of regions I and II in biodrying pile of trial SD had no obvious changes, while those in other biodrying piles diminished to a different extent after biodrying. Meanwhile, fluorescence signal peaks appeared in regions III and V of all samples after biodrying, and the fluorescence intensities in regions III and V of samples from trials SCG, CC, and BA were much higher than in other trials. Besides, the fluorescence intensities of region IV decreased obviously, which indicates the good biodegradability of soluble microbial byproduct-like materials in those trials. These results suggested that biodegradation or release of aromatic proteins from LB-EPS facilitated the biodrying process in all trials but trial SD, and the good biodrying performances in trials SCG, CC, and WH resulted in an effective transformation of biodegradable EPS (i.e., proteins and soluble microbial byproduct-like materials) to stable humic acid- and fulvic acid-like substances.

S-EPS is an important source of dissolved organic matter (DOM) which is the most active organic fraction easily utilized by microbes [39]. In trials SCG, CC and WH, the fluorescence intensity of proteins and soluble microbial byproduct -like materials in S-EPS increased obviously. Meanwhile, the fluorescent groups belonging to biorefractory compounds (i.e., fulvic acid and humic acid-like substances) were also enhanced after biodrying. However, in trial CK, the fluorescence intensity of protein-like materials was barely changed, while that of soluble microbial byproduct-like materials decreased obviously after biodrying. Besides, the peak intensities of the fulvic acid and humic acid-like substances were significantly enhanced. Similar results were observed in the S-EPS of trials SD and BA.

Zhang et al. [11], who characterized EPS during anaerobic digestion of heating pretreated sludge, found that heat pretreatment (80 °C, 30 min) could release EPS and intracellular biopolymers, such as protein and carbohydrates, from the sludge flocs into the DOM. In addition, during the anaerobic digestion process, EPS would be hydrolyzed to serve as carbon and energy source for microbial activities. In this study, the high temperature of biodrying piles (especially for trials SCG, CC, and WH) could destroy the sludge flocs and rupture the microbial cell walls, resulting in the release of EPS and intracellular materials into DOM. Furthermore, the microbes use soluble matter to maintain the metabolism and compose themselves, and it led to the hydrolysis of solid particles occurring throughout the biodrying process [8]. The thermophiles enriched during the high-temperature period of biodrying could secrete thermostable hydrolytic enzymes, which could decompose the sludge flocs and disintegrate the EPS more efficiently [38].

For trial SD, the low microbial activity manifested by moderate biodrying pile temperature caused the accumulation of biodegradable organic matter (proteins and soluble microbial byproduct-like materials) in TB-EPS, and vice versa. According to Guo et al. [38], the fluorescence intensity of humic acid-like organics in EPS decreased while that in DOM increased after sludge hydrolysis using thermophilic bacteria, implying the accumulation of non-biodegradable organics in DOM. In this study, however, the fluorescence intensities of humic and fulvic acid-like organics increased in almost all EPS layers of all biodrying piles, perhaps because the biodrying piles had experienced a long period of aerobic bio-stabilization.

Furthermore, a large amount of bound water, which could not be easily removed during dewatering or drying process, was trapped in EPS [40]. Therefore, the destruction of EPS could lead to the release of bound water [41], which is beneficial for the water mass transfer during biodrying. Cai et al. [10] reported that when the biodrying pile temperature exceeded 40 °C, a large amount of bound water was transformed into free water. In trial SD, it was speculated that the bound water could not be released due to the low biodrying pile temperature and inefficient conversion of TB-EPS.

3.4. Elemental Compositions and Calorific Values

Table 2 shows the elemental compositions and calorific values of the biodrying feedstocks and biodried products. Due to the amendment of the bulking agents, the C contents of the mixtures were higher than that of DS (feedstock for trial CK), and the C content of the feedstock for trial SCG was the highest (46.83%). Through the biodrying process, the C content for the mixtures did not show remarkable changes, while that of trial CK decreased sharply from 34.77% to 25.96%. It was also observed that the N content for DS decreased from 5.47% to 3.59% because proteins were dominantly biodegraded and transformed into free NH_3, which is stripped by airflow during the biodrying process. These results suggested that bulking agents with higher carbon contents modified the elemental composition of the mixtures and buffered against the elemental content changes during the biodrying process.

The HHV decreased after biodrying because the organic matter was consumed by microbes, and the VS contents of the biodried products decreased. LHVs reflecting the heating values of the wet materials were calculated to evaluate the effect of biodrying on the energy content of the matrix. Generally, water removal could increase the matrix LHV, while organic matter consumption could reduce its calorific value [35]. As shown in Table 2, LHVs of all trials increased after biodrying, indicating that the positive effect of water evaporation was greater than the energy loss caused by VS consumption. Especially, the LHV of trial SCG increased by 1234.21 $kJ \cdot kg^{-1}$, which was the maximum LHV enhancement among these trials because the water removal ratio during biodrying was high. As the C content, which is positively correlated with the HHV [42], of the matrix of trial SCG was the highest, the LHV of the biodried product of trial SCG was also the highest (4984.30 $kJ \cdot kg^{-1}$). However, due to the higher MC of the biodried product, this value was much lower than the LHV reported by Hao et al. [7], who also conducted DS biodrying using SCG as a bulking agent. Another possibility that could not be excluded is that the calorific values of the feedstocks used in this study were much lower than the reported values. Nonetheless, this value was close to the lowest LHV threshold (5000 $kJ \cdot kg^{-1}$) for direct use as RDF [35]. These results suggested that the addition of bulking agents improved the calorific value of the mixture and protected the energy content of the biodried product. However, in consideration of the variations in physicochemical characteristics of the raw materials and biodrying efficiency, additional thermal drying might be necessary for efficient utilization of the biodried products with relatively low LHV resulting from high MC.

Table 2. Elemental compositions and calorific values of biodrying feedstocks and biodried products.

Parameters	Samples	CK	SCG	CC	BA	WH	SD
C	Biodrying feedstock	34.77	46.83	40.17	42.17	38.16	40.91
(%, dry basis)	Biodried product	25.96	46.44	40.04	42.36	34.09	40.09
N	Biodrying feedstock	5.47	3.11	1.88	1.45	2.42	5.87
(%,dry basis)	Biodried product	3.59	4.00	2.96	1.11	3.00	5.77
H	Biodrying feedstock	5.12	6.03	5.35	5.14	5.35	5.27
(%, dry basis)	Biodried product	4.47	5.81	5.29	5.25	4.77	5.31
S	Biodrying feedstock	0.82	0.30	0.22	0.17	0.30	0.17
(%, dry basis)	Biodried product	0.97	0.38	0.44	0.20	0.68	0.27
O	Biodrying feedstock	31.90	37.23	44.74	42.31	41.12	39.40
(%, dry basis)	Biodried product	26.72	33.99	37.75	39.94	36.38	38.46
HHV (kJ/kg)	Biodrying feedstock	14,975.75	17,720.28	14,810.22	14,834.17	15,241.10	14,957.79
	Biodried product	14,387.81	16,034.68	12,121.21	13,653.36	13,800.84	14,596.64
LHV (kJ/kg)	Biodrying feedstock	1755.35	3750.09	2989.07	3043.47	3038.77	2885.73
	Biodried product	2578.16	4984.30	3385.04	3203.75	3764.41	3555.63

3.5. Thermal Drying Characteristics

Figure 5a shows the water removal of the feedstocks and biodried products during different periods (i.e., 0–10 min, 10–20 min, 20–30 min, 30–60 min, and 60–120 min) of the isothermal drying process at 60 °C. The water removal in the initial 10 min for the biodrying feedstocks of trials SCG, CC, BA, WH, and SD was much higher than that of CK (1.53 g), with values of 1.94, 2.07, 2.15, 2.17, and 2.03 g, respectively. What is more, 1.12 g water was removed from 60 to 120 min for the feedstock of trial CK, while the drying processes for feedstocks of other trials were almost completed within 60 min. In consideration of the different MCs of the biodrying feedstocks and biodried products for the isothermal drying test, the water removal ratio, which was defined as the mass percentage of water removed in a specific period compared to the water removed during the whole drying process, was calculated for the comparison of drying performance. As Figure 5b shows, the water removal ratios of the initial 10 min for the feedstocks with SCG, CC, BA, WH, and SD were 29.04%, 28.16%, 30.51%, and 28.69%, respectively, and they were also obviously higher than that of CK (20.50%). These results suggested that adding bulking agents improved the drying characteristics of the biodrying feedstocks.

In the initial 10 min of the isothermal drying process, less water (1.31 g) was removed from the biodried product of trial CK, compared to its biodrying feedstock (1.53 g). Meanwhile, the water removal ratio for biodried product (18.25%) in this stage was also lower than that of biodrying feedstock (20.52%), indicating that biodrying without a bulking agent deteriorated the drying characteristics of the sludge. The reason might be that high matrix temperature and efficient water removal swiftly promoted sludge aggregation when DS was biodried without a bulking agent [5,13]. As a result, the thermal drying of the biodried sludge became more difficult because the remaining water was trapped in the sludge particles.

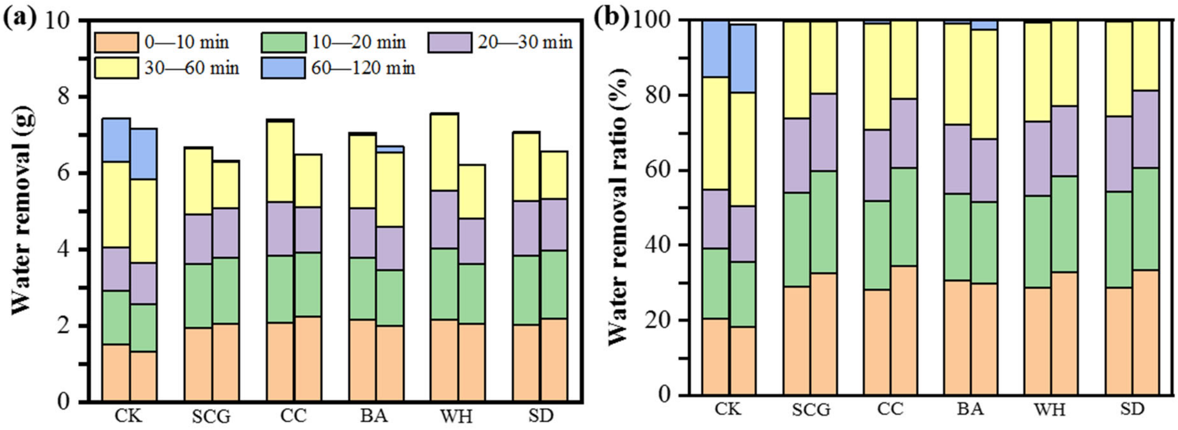

Figure 5. Drying characteristics of biodrying feedstocks and biodried products during thermal drying at 60 °C: (**a**) Water removal and (**b**) water removal ratio at different time intervals.

The initial 10-min water removal ratio of the biodried product for trial BA was identical to that of the biodrying feedstock, indicating that biodrying with BA as a bulking agent barely influenced the drying characteristics of the mixture although BA improved the drying characteristics of the biodrying feedstock. With respect to trial SCG, 2.06 g water was evaporated from the biodried product in the initial 10 min of the isothermal drying process, accounting for 32.62% of the total water loss, and both water removal (1.94 g) and water removal ratio (29.04%) of the biodrying feedstock was lower than those of the biodried product. Similar results were observed in the biodrying materials for trials CC and SD. In the case of trial WH, although the biodried product showed a slightly lower water removal (2.04 g) compared to the biodrying feedstock (2.17 g), its water removal ratio (32.78%) was higher than that of the latter (28.62%), which implied the drying performance of the biodried product was better.

During the drying period of 10 to 20 min, the water removal for biodried product (1.72 g) of trial SCG was still higher than that of biodrying feedstock (1.67 g). For biodried products of other trials, the water removals were lower than those of feedstocks, while the water removal ratios for biodried products of trials CC, WH, and SD were higher than those of biodrying feedstocks. After that, the biodried products showed lower water mass losses and water removal ratios than the biodrying feedstocks due to lower water contents for biodried products. The water removal profile results indicated that DS biodrying with bulking agents improved the drying characteristics of the mixture, and SCG was a more desirable bulking agent compared to other materials.

To compare the drying kinetics of the matrix, the WDR was calculated and fitted versus the residual water in the mixture (Figure 6). Since the drying process mainly occurred in the first failing rate period [43], the values of the preheating period and the second failing rate period were excluded. All samples showed linear trends with high coefficients ($R^2 \geq 0.98$), while the slopes and intercepts of these trends varied. The slopes of the fitted lines of the biodrying feedstocks and biodried products were in the order of BA > SD > CC > SCG > WH > CK and CC > SD > WH > BA > SCG > CK, respectively. The WDR of the feedstocks mixed with bulking agents was much higher than that of sludge (trial CK) when the residual water in the matrix was the same, indicating that the water in the matrix conditioned using bulking agents could be more easily evaporated than that in the sludge. For trial CK, the WDR of the biodried product was lower than that of the feedstock when they were with the same residual water. On the contrary, all the biodried products with bulking agents, except that with BA, showed higher WDR values than their feedstocks. These results agreed well with the water removal profiles shown in Figure 5 and suggested that bulking agents not only improved the drying characteristics of the biodrying feedstocks, but also enhanced the positive effects of biodrying on the water removal profile of the matrix. The linear regression of the biodried product with SCG showed a gentler slope compared to those with CC and WH, while its intercept was relatively higher, indicating that its drying rate was more reluctantly affected by the residual water content.

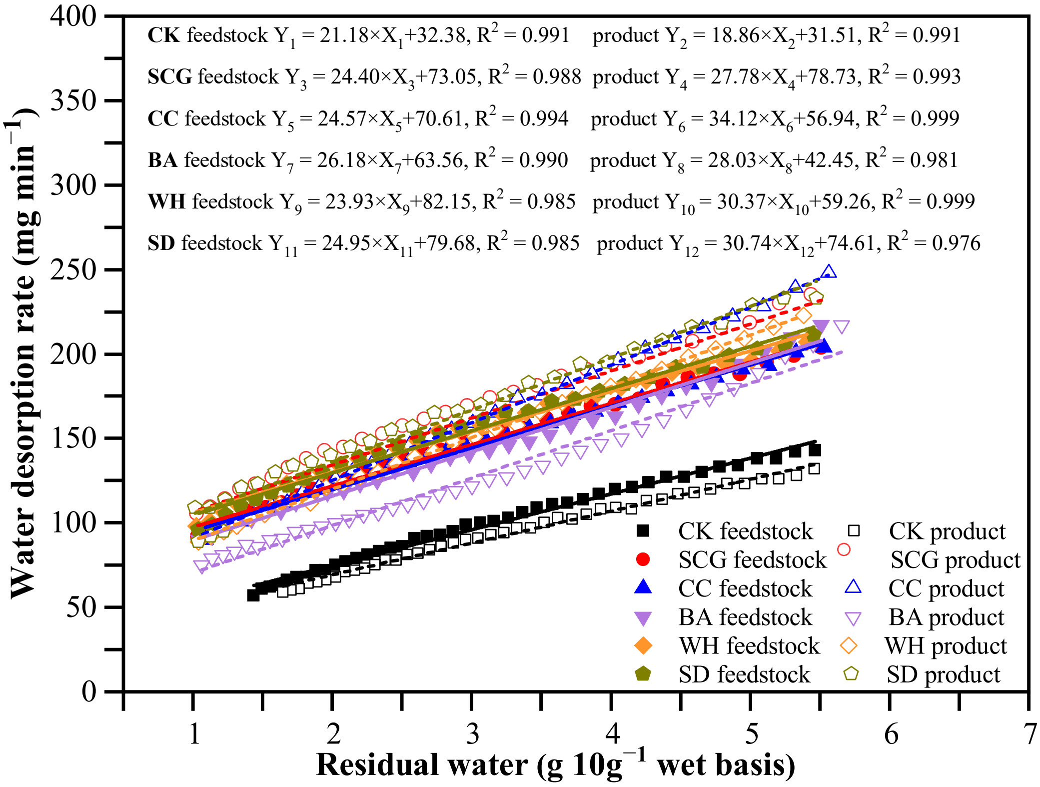

Figure 6. Water desorption rate of biodrying feedstocks and biodried products with different bulking agents.

In summary, bulking agent addition to DS led to higher drying rates and shorter drying times of the mixture. Meanwhile, bulking agents also reinforced the matrix structure during biodrying, resulting in excellent drying characteristics of the biodried products with bulking agents, especially those with SCG.

4. Conclusions

Results of this study indicated that the bulking agents greatly affected the DS biodrying performance, organics degradation, EPS release and transformation, and thermal drying characteristics of the biodrying mixtures. Abundant biochemical compositions in SCG facilitated the bio-heat generation and accelerated the water removal during biodrying. Besides, SCG rich in biodegradable lipids regulated C/N ratio of biodrying pile by improving lipid content, resulting in the highest biodrying index in trial SCG. The high matrix temperature reflecting vigorous microbial activity in trials SCG, CC, and WH facilitated the release and conversion of EPS. The biodried product with SCG was endowed with higher C content and calorific values (both HHV and LHV), which was beneficial for RDF production and energy recovery. In addition, adding bulking agents in DS not only improved the drying rates of the mixtures but also enhanced the positive effect of biodrying on thermal drying characteristics of the biodried products. The biodried product of trial SCG showed remarkable drying characteristics with a high drying rate, which was reluctantly affected by the residual water content in the matrix. These findings could serve as the references for selecting bulking agents for engineering the DS biodrying combined with further thermal drying.

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Abbreviations

BA	bagasse	LHV	low heating value
CC	corncob	MC	moisture content
CK	control check	RDF	refuse-derived fuel
DOM	dissolved organic matter	SCG	spent coffee ground
DS	dewatered sludge	SD	sawdust
EPS	extracellular polymeric substance	3D-EEM	three-dimensional excitation emission matrix
S-EPS	soluble EPS	VS	volatile solid
LB-EPS	loosely bound EPS	WDR	water desorption rate
TB-EPS	tightly bound EPS	WH	wheat husk
HHV	high heating value		

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