



## Preparation and characterization of a novel multi-component composite biological filler

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### ABSTRACT

A novel multi-component biological filler, consisting of residual activated sludge, clay, humus, silicon compounds, fly ash, and humic acid organic fertilizer, is described here. The aim of the study was to enhance and improve the activity of activated sludge in a biological wastewater treatment process. Following the addition of the filler to a sequencing batch reactor, significant effects on sludge settling properties, sludge yield, sludge dewatering performance, and sludge odor were observed. To further optimize the components of the novel multi-component composite bioactive filler and to improve the activity of the sludge, its composition and microstructure were studied. The biological filler was characterized by different techniques, i.e. field emission scanning electron microscope (FESEM), energy dispersive spectrometer (EDS), and Fourier transform infrared spectroscopy (FTIR). Through measurements of the composition of organic matter and humus, it was found that the content of each component remained unchanged over a six-month period. The FESEM results showed that the surface of the filler was rough and porous, while the EDS results revealed that the main elements in the filler were O, Si, and C, and the FTIR results indicated that the main functional groups of the absorption peak were  $-\text{COOH}$ ,  $\text{CO}_3^{2-}$ ,  $\text{SiO}_2$ , and  $-\text{C}=\text{O}$ . The elements contained within these functional groups (O, Si, and C) were the same as those identified by the EDS.

*Keywords:* Multi-component composite; Sludge settling; Sludge yield; Sludge dewatering performance; Organic matter; Humus; Components

### 1. Introduction

Improvements in the activity of activated sludge are essential to the development of biological sewage treatment systems. To effectively use the characteristics of microorganisms attached to a solid surface, a series of biological fillers were recently developed to enhance and improve the biological treatment of sewage [1–6].

Sui and Shi invented an active filler for wastewater treatment, which contains an active substance [7]. The filler can promote the growth of the microorganisms on the active surface, ensure the rapid formation of a biofilm rapidly, and increase the useful area, thereby increasing the microbial biomass and improving the biological activity of the surface. Long tongrui developed a filler, which can easily culture a biofilm and stimulate microbial activity [8,9]. The filler not only

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provides appropriate nutrition for microorganisms, but also provides a location for physical attachment, and is hydrophilic which promotes microbial growth, accelerates biofilm culturing, and improves the treatment efficiency. Zhang et al. developed a new filler, which combines biological affinity and hydrophilic modification [10]. Calcium alginate, starch, polyvinyl alcohol, and stearic, magnetic, and activated carbon were introduced into an ordinary polymer bioactive filler. The filler can promote biofilm culturing, metabolism, and biological contact oxidation. Hideo invented a filler that can be added to the aeration tank, enriching the microbial population in the sewage treatment process, and strengthening the effectiveness of sewage treatment [11]. Qiyuan Feng developed a lightweight sludge ceramic filler, which has a large surface area and pore size that is suitable for microbial attachment and growth [12]. These bioactive fillers have mainly been used in biological contact oxidation sewage treatment or in hybrid biological treatment technologies. Their main role is to provide a good location for microbial growth, but they also enhance and improve the activity of sludge in the sewage treatment.

The activated sludge process is the most widely used sewage treatment process worldwide, but its effectiveness is limited by several problems, including its propensity for sludge bulking, high sludge yields, poor sludge separation, inconsistent removal of nitrogen and phosphorus, and odor pollution. Therefore, a novel multi-component composite biological filler (bio-filler) was developed, which can enhance and improve the characteristics of activated sludge through internal or external processes. By incorporating this biological filler into a sequencing batch reactor (SBR), its effect on sludge settling, sludge yield, and sludge dewatering performance was determined. The composition and microstructure of the biological filler were characterized using a field emission scanning electron microscope (FESEM), energy dispersive spectrometer (EDS), and Fourier transform infrared spectroscopy (FTIR). A potassium dichromate oxidation–external heating method and a sodium pyrophosphate extraction–potassium dichromate oxidation method were used to analyze the organic matter and humus, respectively.

## 2. Experimental

### 2.1. Biological filler preparation

To improve the sludge activity, residual activated sludge, clay, humus, silicon compounds, sawdust, fly ash, iron magnesium compound, and humic acid organic fertilizer were used as biofiller materials. Following a single-component test and multiple-

component ratio test, the materials that were finally selected were residual activated sludge, clay, humus, silicon compounds, fly ash, and humic acid organic fertilizer. The materials were mixed in a certain proportion and cured in ambient air to produce a cylindrical biofiller (Fig. 1). The average dimensions and related parameters were as follows: diameter = 15 mm, length = 35 mm, volume = 6 cm<sup>3</sup>, weight = 12 g, density = 2 g cm<sup>-3</sup>, surface area = 9.89 m<sup>2</sup> g<sup>-1</sup>, total pore volume = 0.04 cc g<sup>-1</sup>, and pore size = 18.8 Å.

### 2.2. Experimental conditions

A simulated domestic wastewater was used as the raw water, by adding C<sub>6</sub>H<sub>12</sub>O<sub>6</sub>, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, and KH<sub>2</sub>PO<sub>4</sub> as a source of nitrogen and phosphorus, and adding NaHCO<sub>3</sub> to adjust the pH of the water. The pH was controlled to be between 6.5 and 7.5, the COD was 400–450 mg L<sup>-1</sup>, total nitrogen was 45–50 mg L<sup>-1</sup>, total phosphorus was 7–8 mg L<sup>-1</sup>, TOC was 180–190 mg L<sup>-1</sup>, mixed liquid suspended solids (MLSS) was 6,500 mg L<sup>-1</sup>, and temperature was about 25°C. The experimental sludge was taken from a sewage treatment plant in the city of Changchun. Under laboratory conditions, three experimental devices were seeded for cultivation and domestication at the same time. The aeration and running time of the three experimental devices were identical.

### 2.3. Experimental device

Three different experimental devices were tested. The first device (Fig. 2) was a modified SBR. It was constructed as a ring sandwich inside the reactor, with the biological filler and small stones placed into the sandwich. The top and bottom surface of the sandwich were densely covered with small ventilation



Fig. 1. Novel multi-component composite biological filler.

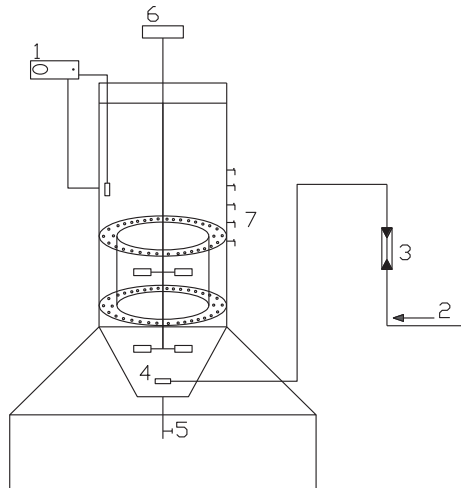


Fig. 2. SBR reactor with internal biological filler.  
Notes: 1—temperature controller, 2—compressed air, 3—gas rotameter, 4—aerator, 5—mud pipe, 6—mechanical stirrer and 7—taps.

holes, with the blender placed into a large hole in the middle of the sandwich. The second device (Fig. 3) was an SBR connected to a biological filler reactor. The biological filler reactor was made of organic glass, with a diameter of 10 cm, height of 40 cm, and effective volume of 3.1 L. At the bottom of this reactor, a sludge vent pipe and a return pipe connected with the SBR reactor using a Lange peristaltic pump. The third experimental device (Fig. 4) was an ordinary SBR as a comparison.

The SBR reactor was made of organic glass, with a truncated cone-shaped mud bucket in the bottom of the device. The diameter was 19 cm, height was

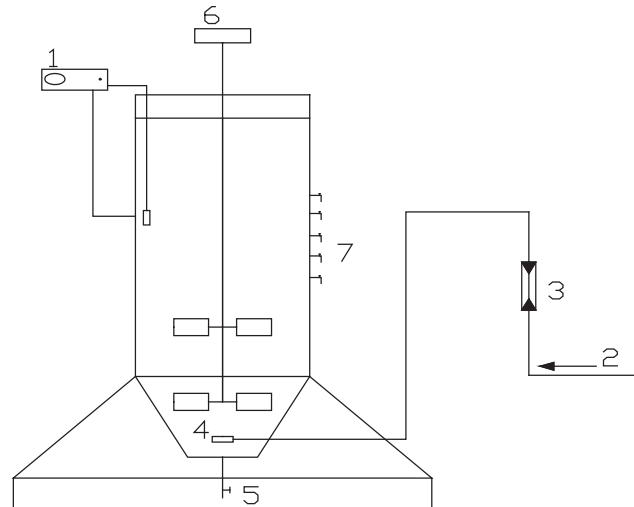


Fig. 4. Ordinary SBR reactor.  
Notes: 1—temperature controller, 2—compressed air, 3—gas rotameter, 4—aerator, 5—mud pipe, 6—mechanical stirrer and 7—taps.

60 cm, and the effective volume was 15 L. Sampling ports were established vertically in the reactor wall at 10-cm intervals. At the bottom of the reactor, a sludge vent pipe was established. The SBR reactor used blast aeration, with an air compressor providing blasts of compressed air through porous stone.

#### 2.4. Organic matter and humus measurement

The potassium dichromate oxidation–external heating method [13] and sodium pyrophosphate extraction–potassium dichromate oxidation [14] were used

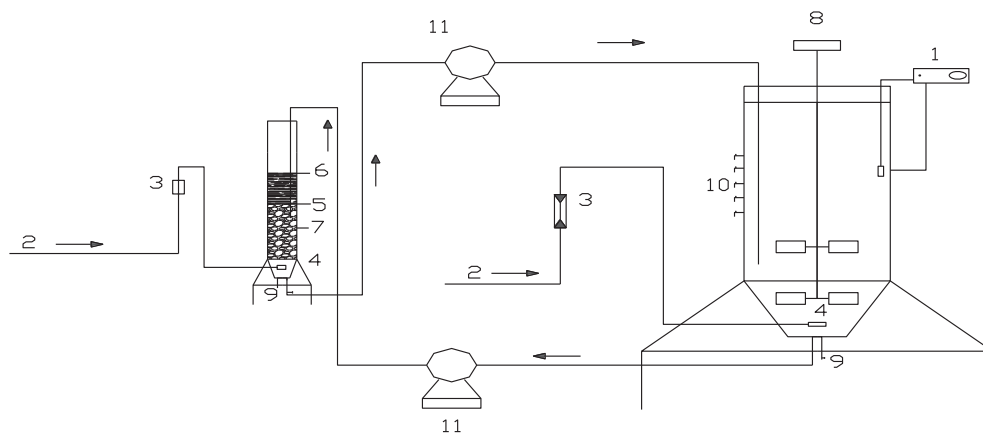


Fig. 3. SBR reactor with external biological filler.  
Notes: 1—temperature controller, 2—compressed air, 3—gas rotameter, 4—aerator, 5—perforated plate, 6—biological filler, 7—light stone, 8—mechanical stirrer, 9—mud pipe, 10—taps and 11—peristaltic pump.

for the analysis of organic matter and humus, respectively.

### 2.5. Characterization

A capillary suction timer (304 BC, Triton) was used to determine the sludge capillary suction time (CST). A FESEM (Quanta-200; FEI, Hillsboro, OR, USA) was used to detect the morphology and composition biological filler. of the surface of the composite. FTIR spectra were recorded using a spectrometer (AVATAR360; Nicolet/Thermo Fisher Scientific, Waltham, MA, USA).

## 3. Results and discussion

### 3.1. The effect of the biological filler on the removal of organic matter and nitrogen

The removal of COD and  $\text{NH}_3\text{-N}$  in the three reactors was compared to investigate the effect of the biological filler.

Fig. 5 shows the removal of COD in the three reactors. The inlet water COD was  $432.3 \text{ mg L}^{-1}$ , and the average removal of COD by the three reactors was 91.64, 91.62, and 90.69%, with final effluent water concentrations of 36.17, 36.27, and  $40.23 \text{ mg L}^{-1}$ . The COD removal efficiency was therefore high, with the addition of the filler producing a slightly better result than in the control group. Fig. 6 shows the removal of  $\text{NH}_3\text{-N}$  in the three reactors. The  $\text{NH}_3\text{-N}$  concentration in the inlet water of  $33.61 \text{ mg L}^{-1}$ , and the average removal of  $\text{NH}_3\text{-N}$  in the three reactors was 83.08, 82.97, and 81.67%, with final effluent water concentrations of

5.69, 5.72, and  $6.16 \text{ mg L}^{-1}$ . The addition of the filler resulted in a better  $\text{NH}_3\text{-N}$  removal than in the control group. The addition of a filler can therefore enhance the removal efficiency of COD and  $\text{NH}_3\text{-N}$ . It is likely that the filler releases humic substances and silicates, which could enrich the microbial community, changing its structure, and improving the sewage treatment [11,15].

### 3.2. The effects of the biological filler on sludge settling

The sludge settling properties have a crucial effect on the activated sludge treatment system. If the sludge is loose or settles poorly, the quality of the outlet water will be seriously affected. This results in not only a loss of sludge, but also reduces the pollutant removal efficiency. The sludge settling properties were measured in the three experimental devices for the same level of MLSS and under the same operating conditions, with stable aerobic and anoxic phases. With different idling times, the sludge settling properties can change, and therefore, sludge settling was measured as the idling time was varied from 12 to 48 h.

In both the aerobic and anoxic phases, the sludge properties of the three reactors were very different (Fig. 7).

The fitting equation of the settling curve was as follows [16]:

$$V = \frac{H \times 3600}{t} \quad (1)$$

where  $V$  is the rate of sludge settling in  $\text{m h}^{-1}$ ,  $H$  is the distance of sludge settling in  $\text{m}$ , and  $t$  is the time

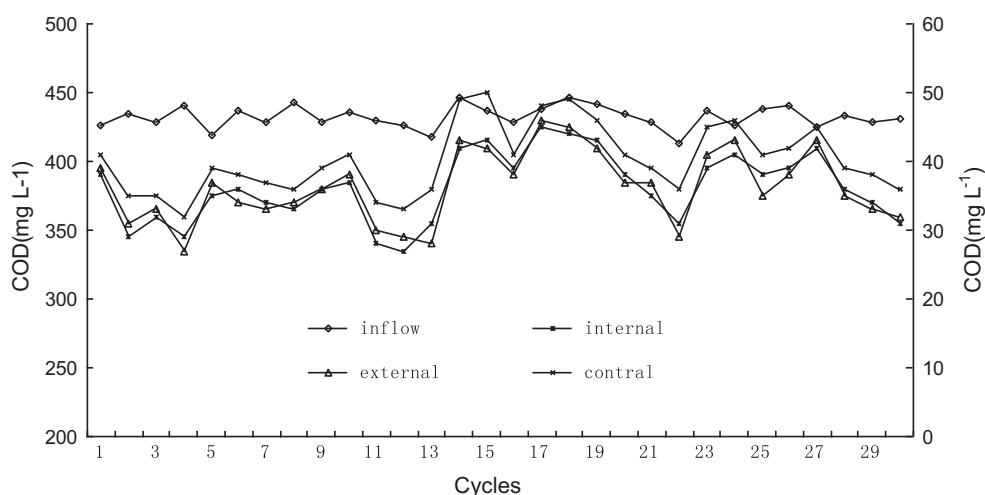


Fig. 5. The removal of COD in the three reactors.

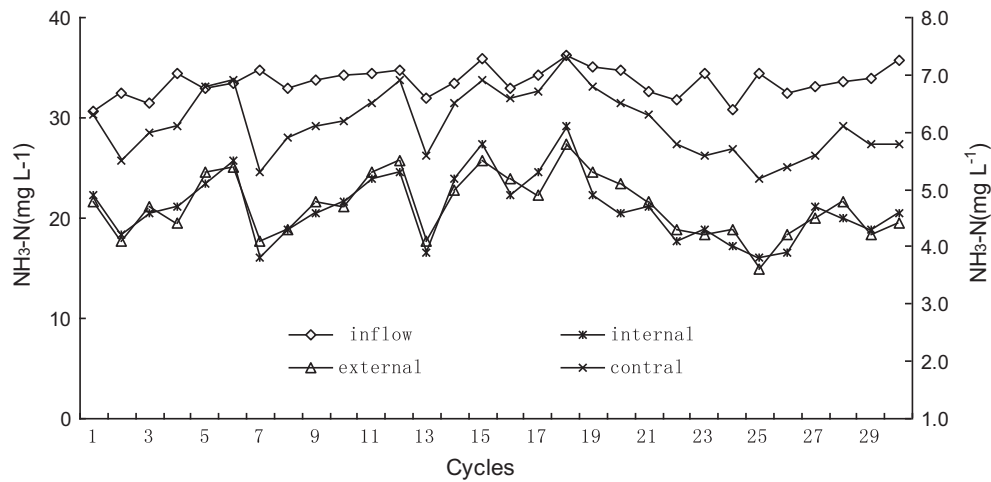


Fig. 6. The removal of NH<sub>3</sub>-N in the three reactors.

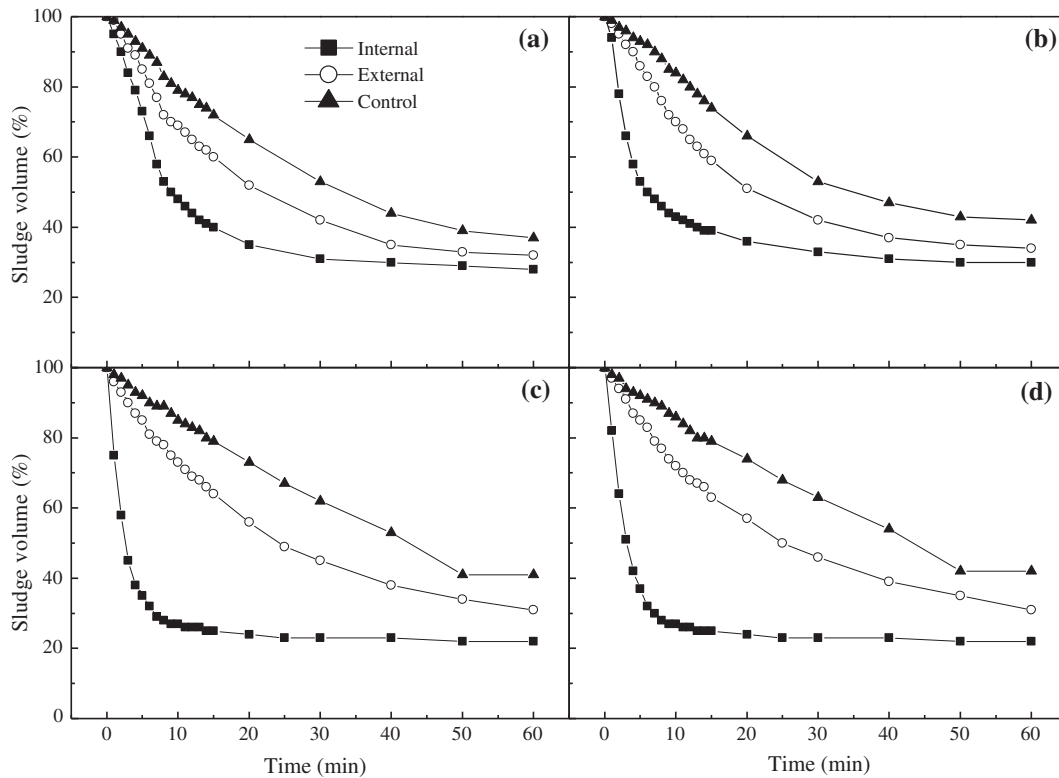


Fig. 7. Sludge settling of aerobic phase (a) idle for 12 h, (b) idle for 48 h, sludge settling of anaerobic phase, (c) idle for 12 h and (d) idle for 48 h.

of sludge settling in *s*. A successful result was obtained when the experimental data were in accordance with Eq. (1): in the aerobic phase,  $V_{\text{internal}}$  ( $V_I$ ) = 0.234 m h<sup>-1</sup>,  $V_{\text{external}}$  ( $V_E$ ) = 0.194 m h<sup>-1</sup>, and  $V_{\text{control}}$  ( $V_C$ ) = 0.158 m h<sup>-1</sup>, ( $V_I > V_E > V_C$ ), and in the anoxic phase,  $V_I = 0.226$  m h<sup>-1</sup>,  $V_E = 0.194$  m h<sup>-1</sup>, and

$V_C = 0.158$  m h<sup>-1</sup>, ( $V_I > V_E > V_C$ ). Thus, the sludge settling properties of the internal biological filler were much better than the other two groups, although the sludge settling properties of the external biological filler was also superior to the control group. The sludge volume reduced sharply during the initial settlement

in the aerobic phase, as shown in Fig. 7(a) and (c); after 10 min, it had fallen to about 40% and in the anoxic stage was down to about 30%. In contrast, the ordinary activated sludge reduced to about 85% after 10 min, and about 40% after 60 min. The sludge settling properties with an idling time of 48 h were the same as with the normal running time, as shown in Fig. 7(b) and (d), with the addition of the filler producing a slightly better result than in the control group. These results demonstrate that the sludge settling properties can be significantly improved by adding biofiller to the SBR. The EDS results indicated that the filler contained compounds of Al, Ca, Mg, and other elements, which were likely to improve sludge settling by adsorption bridging.

The formula used to determine the area of the secondary sedimentation tank surface was as follows [17]:

$$A = \frac{Q}{q} = \frac{Q}{3.6u} \quad (2)$$

where  $A$  is the surface area in  $\text{m}^2$ ,  $Q$  is the maximum water flow rate in  $\text{m}^3 \text{h}^{-1}$ ,  $q$  is the surface load in  $\text{m}^3 (\text{m}^2 \text{h})^{-1}$ , and  $u$  is the sedimentation rate of sludge settling in  $\text{mm s}^{-1}$ . Thus, the surface area of the secondary settling tank was closely related to the sludge sedimentation rate. The sludge settling properties of the internal biological filler were much better than other two groups, so the surface area of the second settling tank could be reduced, saving floor space, and reducing costs.

### 3.3. The effect of the biological filler on sludge yield

The activated sludge process is a widely used sewage treatment process, although it generates a large amount of excess sludge [18]. The excess sludge is a pollution problem because it contains harmful organic matter. Therefore, sludge reduction and recycling technologies have received much attention. To determine the apparent sludge yield coefficient, for the same sludge concentration and stable operating conditions, with each mg reduction of the COD and suspended solids (SS) [19], the following formula was determined:

$$Y = \frac{Q_w X_w + (Q - Q_w) X_e}{Q(S_0 - S_e)} \quad (3)$$

where  $Q$  is the water inflow in  $\text{L d}^{-1}$ ,  $Q_w$  is the residual sludge volume in  $\text{L d}^{-1}$ ,  $X_w$  is the concentration of excess sludge in  $\text{mg L}^{-1}$ ,  $X_e$  is the concentration of effluent SS in  $\text{mg L}^{-1}$ ,  $S_0$  is the COD concentration

of influent water in  $\text{mg L}^{-1}$ ,  $S_e$  is the COD concentration of outlet water in  $\text{mg L}^{-1}$ . A successful result was obtained when the experimental data were in accordance with Eq. (3),  $Y_I = 0.23 \text{ mg MLSS mg COD}_{\text{cr}}^{-1}$ ,  $Y_E = 0.33 \text{ mg MLSS mg COD}_{\text{cr}}^{-1}$ ,  $Y_C = 0.49 \text{ mg MLSS mg COD}_{\text{cr}}^{-1}$ , ( $Y_C > Y_E > Y_I$ ). Thus, adding a biological filler to an SBR reactor can reduce both the sludge yield and pollution of the environment.

### 3.4. The effect of the biological filler on sludge dewatering performance

The CST refers to the time required for a filtrate to penetrate a certain distance on the filter and is used to characterize sludge dewatering [20]. To investigate the effects of the biological filler on the sludge dewatering performance, the CST of the three experimental devices was measured. The CST values of the internal, external, and control groups were 16.7, 18.5, and 22.9 s, respectively. This shows that a biological filler can play a role in improving sludge dewatering, which is mainly due to the physical structure and chemical composition of the biological filler. A biological filler has a porous and loose physical structure, and these structural characteristics can greatly improve the sludge dewatering performance [21]. In addition, the  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  content in a biological filler is relatively high, and both can affect the sludge dewatering performance [22–24]. Therefore, the use of a biological filler can improve the sludge dewatering performance.

### 3.5. Organic matter and humus composition of the biological filler

As a consumable in the sewage treatment process, a biological filler could release organic functional

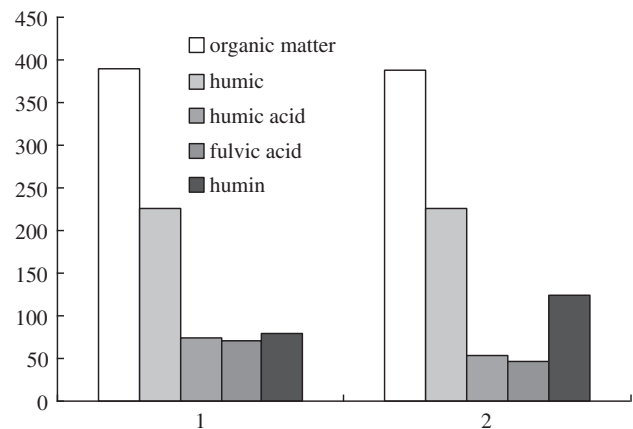


Fig. 8. Content of organic matter with biological filler.

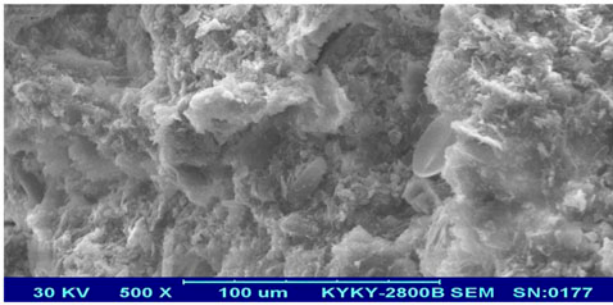


Fig. 9. 500× FESEM image of biological filler.

groups that contact each other during aeration over time. This ensures that the biological filler material continuously enters the sludge. To determine the stability of the organic matter and humus in the biological filler, a six-month analysis of the used and unused organic matter and humus was undertaken. The surface of the filler was cleaned using a soft brush and then washed three times with distilled water. The results are shown in Fig. 8.

The organic matter and humus content of the biological filler were 390.0 and 226.0 g kg<sup>-1</sup>, respectively. The organic matter and humus content of a biological filler used for six months were 389 and 226 g kg<sup>-1</sup>, respectively, i.e. a difference of only 1 g kg<sup>-1</sup> in the organic matter while the humus content was unchanged. This suggests that the biological filler was stable during the sewage treatment process. The humic and fulvic acid concentrations were reduced by 20.8 and 24.2 g kg<sup>-1</sup>, respectively. The humin concentration increased from

79.9 to 125.0 g kg<sup>-1</sup>, which may be because humin was in a transitional position between humic acid and carbides, or because humic acid could become humin due to freezing and drying processes, which are irreversible [25,26]. Humin has a critical role in the migration, transformation, and bioavailability of hydrophobic organic pollutants in the environment [27].

### 3.6. Surface morphology of biological filler

Fig. 9 is a 500× SEM image of biological filler. It shows that the surface of the biological filler had a rough and loose structure, which provides a large surface and ensures a large adsorption capacity [28].

### 3.7. Components of the biological filler determined by spectrum analysis (FESEM–EDS)

Fig. 10 shows the EDS results for the biological filler, and Table 1 shows the elemental composition and content of three parallel samples. It can be seen from Fig. 8 and Table 1 that the main elements within the biological filler were C, N, O, Al, Si, and Fe. The element with the highest content was O, with a weight percentage (Wt) of 41.37%, and average percentage of atomicity (At) of 42.36%. The next most abundant element was Si, with a Wt of 23.50% and an At of 15.11%. The third most abundant element was C, with a Wt of 19.01% and an At of 28.38%. The percentage composition of the three elements was 83.88%, with an At of 86.13%, indicating that the main elements within the biological filler were O, Si, and C.

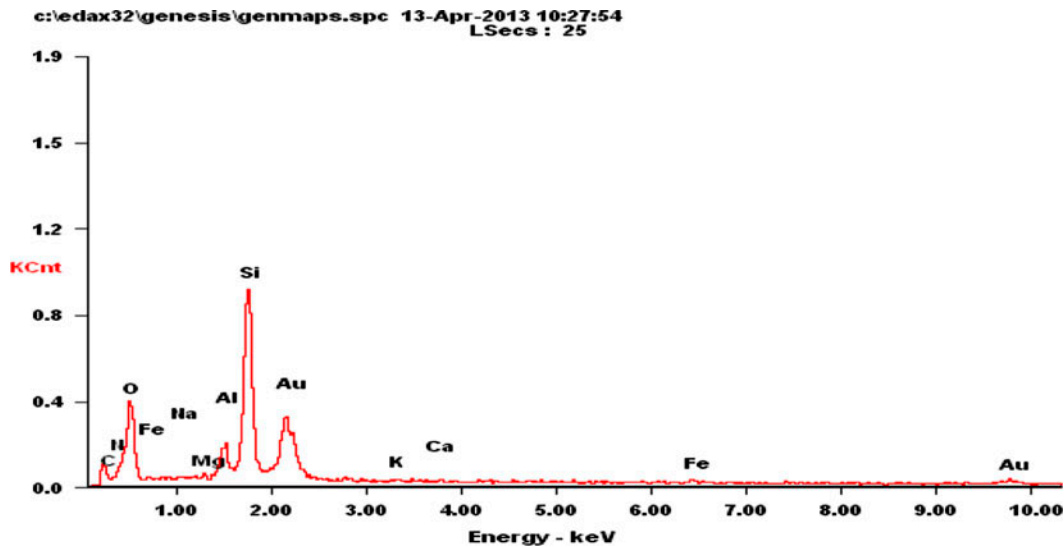


Fig. 10. EDS spectra of biological filler.

### 3.8 Fourier transform infrared spectra of the biological filler

Fig. 11 shows that the biological filler contained several functional groups, such as  $-\text{COOH}$ ,  $-\text{OH}$ ,  $-\text{NH}_2$  and  $-\text{C}=\text{O}$ , which could coordinate, complexate, and chelate with heavy metals, thus reducing heavy metal pollution. The absorption peaks of the biological filler appeared at 3,425.57, 2,931.80, 2,856.57, 2,025, 1,627.92, 1,577.77, 1,435.03, 1,033.85, 777.31, and 470.63  $\text{cm}^{-1}$ . The absorption peaks at 3,425.57 and 1,627.92  $\text{cm}^{-1}$  may be the functional group  $-\text{OH}$  of water, organic acids ( $-\text{COOH}$ ,  $-\text{COO}$ ), and phenol, or the  $-\text{NH}$  of amines. Absorption peaks of 2,931.80, 2,856.57, and 1,435.03 may be the functional group  $-\text{NH}_2$  of methylene. The absorption region of 1,640–1,650  $\text{cm}^{-1}$  may be the functional groups  $\text{C}=\text{C}$ ,  $-\text{COO}$ ,  $\text{C}=\text{O}$ , and  $-\text{NH}$  of water and ammonia. Absorption peaks of 1,577.77  $\text{cm}^{-1}$  may be the functional groups  $-\text{NO}_3$  and  $-\text{NO}_2$ . The absorption region of 1,410–1,450  $\text{cm}^{-1}$  may

be the functional group  $-\text{CO}_3^{2-}$ . The absorption region of 860–880  $\text{cm}^{-1}$  was also a significant region, which indicates the presence of  $-\text{CO}_3^{2-}$ . The absorption peaks of 1,033.85, 777.31, 526.57, and 470.63  $\text{cm}^{-1}$  may be the functional group  $\text{SiO}_2$ . The elements contained within these functional groups (O, Si, and C) were the same as those identified by the EDS.

### 4. Conclusion

Residual activated sludge, clay, humus, silicon compounds, fly ash, and humic acid organic fertilizer were used as biological filler materials. The material in a certain proportion was used to construct a cylindrical biofiller. The biological filler significantly improved the sludge settling properties, reduced the sludge yield, and improved the sludge dewatering performance and sewage treatment efficiency. It was found that after six month operation, the organic matter content had only declined by 1  $\text{g kg}^{-1}$  and the humus content was unchanged, indicating that the biological filler was is stable in the sewage treatment process. Analysis of the microstructure of this filler by FESEM, indicated that the surface of the filler, was rough and porous. The EDS results revealed that the main elements contained within the filler were O, Si, and C. The FTIR analysis indicated that the main functional groups were  $-\text{COOH}$ ,  $\text{CO}_3^{2-}$ ,  $\text{SiO}_2$ , and  $-\text{C}=\text{O}$ . The elements contained within these functional groups (O, Si, and C) were the same as those identified by the EDS.

Table 1  
Elemental composition and content of three parallel samples with biological filler

Elements	1		2		3	
	Wt %	At %	Wt %	At %	Wt %	At %
C	17.43	25.39	24.75	34.97	14.84	24.78
N	5.11	8.88	5.06	8.24	5.21	7.65
O	41.66	44.62	36.05	37.64	46.42	45.64
Na	0.25	0.19	0.44	0.32	0.52	0.45
Mg	0.24	0.17	0.26	0.19	0.13	0.11
Al	3.79	2.46	4.02	2.53	5.28	3.93
Si	26.69	16.63	23.35	14.11	20.45	14.6
K	0.59	0.27	0.75	0.33	1.57	0.8
Ca	0.56	0.24	0.51	0.21	0.51	0.22
Fe	3.68	1.15	4.81	1.46	5.07	1.82
Total	100	100	100	100	100	100

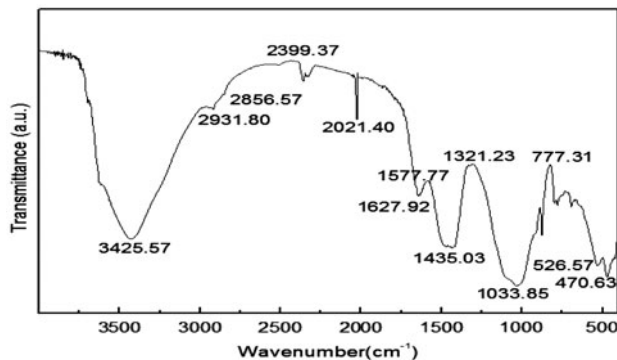


Fig. 11. FTIR spectra of biological filler.

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