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Experimental Study on the Effect of Fulvic Acid in Waste Slurry on Flocculation and Zeta Potential

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Abstract: The waste slurry produced by the dredging of urban rivers needs to be dewatered before being reused sustainably. As a large amount of plant debris accumulates in sediment, humus-like substances become one of main components in waste slurry. In light of the lack of research on the effect of fulvic acid (FA) in waste slurry on flocculation and separation, this paper carried out experimental research, including the effect of FA content on flocculation and filtration, as well as flocculation and filtration experiments of eight different sources of waste slurry. The results show that if only the FA content in the slurry is changed, the effect of FA on flocculation and separation is significant when the FA content is 0~3%, but it is not obvious when the FA content exceeds 3%. The flocculation and filtration results of eight different sources of river-dredged waste slurry are obviously different; the D_{10} increment can differ by nearly 10 times, and the specific resistance to filtration (SRF) differs by 2 orders of magnitude. However, FA is not a sensitive factor affecting the flocculation results. FA mainly affects the results by affecting the zeta potential of the slurry. Therefore, in the dewatering design of waste slurry, only the zeta potential needs to be considered.

Keywords: waste slurry; fulvic acid; flocculation; filtration; zeta potential



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1. Introduction

Regular dredging of urban rivers produces a large amount of waste slurry. There is an urgent need to rapidly dewater the waste slurry to facilitate subsequent product resource utilization, such as brick [1,2], ceramsite [3,4], embankment material [5,6] and foamed concrete [7]. In this way, the sustainable recycling process of waste slurry production, treatment and reuse is realized.

Before the waste slurry is converted into economic products, the reduction of waste slurry is the first process to be carried out. In the existing research, flocculation [8] and filtration [9,10] are often used for the dewatering of waste slurry. Song et al. [11] selected two organic polymers to flocculate and filter the dredged slurry and found that flocculation by chitosan achieved more compact floc and better dewatering performance than cationic polyacrylamide (PAM). Liu et al. [12] used the flocculation–vacuum–preloading method for rapid dewatering of river-dredged slurry and tested the optimal dosage and dewatering results of six flocculants. The results showed that adding 0.8% FeCl_3 or 0.08% anionic PAM could quickly accelerate the process of soil–water separation, and the water content of the slurry could be reduced from 140% to 60%. Cui et al. [13] used sulphate aluminum cement instead of lime as an additive for the plate and frame filtration and dewatering of dredged slurry and achieved better separation results.

However, the properties of waste slurry from different sources are diverse, and the same flocculation scheme may not be able to obtain the optimal flocculation and separation effect. It is of great significance to study the influence of the slurry properties on flocculation

and separation. Considering the surrounding environment of urban rivers, a large amount of plant debris is accumulated in sediment [14] and gradually decomposed into humus-like substances over time [15]. Therefore, it is necessary to study the effect of humus in waste slurry on soil–water separation.

Humus not only causes an odor, taste, color and bacterial regrowth in potable water, but it also has the potential to form carcinogenic disinfection byproducts (DBPs) [16]. Therefore, a lot of studies have focused on how to remove water-soluble humus through flocculation in water treatment. Zhang et al. [17] used coagulation and flocculation to treat humus wastewater and found that the removal percentage of humus substances reached 96.0% when the pH was 7.0. Xu et al. [18] found that fulvic acid (FA) and humic acid (HA), among other materials, can improve the stability of colloidal particles in water, resulting in a poor flocculation effect, small floc size and high specific resistance to filtration (SRF). Sillanpää et al. [19] concluded that the flocculation result was affected by the particle size, charge, hydrophobicity, divalent cations and destabilizing anions of humus. Relevant studies show that humus in water has a significant effect on flocculation [17,18]. For waste-dredged slurry, its properties are different from wastewater or raw water. Research on the effect of humus, such as FA on flocculation and separation in waste slurry, is still lacking. Whether FA is a sensitive factor affecting dewatering in waste-dredged slurry needs to be studied.

In relation to the above problems, this paper uses eight types of natural waste-dredged slurry to carry out flocculation and filtration experiments, and the FA content is also determined to explore the effect of FA on flocculation and separation. In addition, additional FA is added to the waste slurry to study the effect of the FA content on flocculation and filtration. Based on these results, the influence mechanism of FA in the waste-dredged slurry on the results of slurry reduction is analyzed and discussed.

2. Materials and Methods

2.1. Waste Slurry

In order to study the effect of FA in the waste slurry of urban rivers on flocculation and separation, eight types of dredged slurry from five rivers were selected as test samples. The five rivers are located in Nanjing, China. The locations are shown in Figure 1a, and the basic information of the rivers is shown in Table 1. The slurry was sampled with a small grab sampler, and the sampling process is shown in Figure 1b. The properties of the eight types of dredged slurry are summarized in Table 2. The humus constituents were determined by the potassium dichromate oxidation-volumetric method [20]. The particle size distribution of the slurry was measured by a Malvern Mastersizer 2000 laser particle size analyzer. D_{10} , D_{50} and D_{90} , obtained by the particle size distribution, were used to characterize the indicative sizes of the waste slurry.

2.2. Fulvic Acid

In order to study the effect of the FA content on flocculation and separation, Honghua River-dredged slurry (H1 and H2) was added with different dosages of FA to conduct flocculation and filtration tests. The FA was produced by Shanghai Yuanye Biological Co., Ltd.; the molecular formula was $C_{14}H_{12}O_8$, and the molecular weight was 308 daltons.

2.3. Flocculant

Two types of the commercial polymer cationic flocculant PAM (Wshinefloc 312VS and 611HN, manufactured by Shanghai Wshine Chemical Co., Ltd., Shanghai, China) were used for flocculation. The flocculant was reconstituted every day at a concentration of 0.1% (*w/w*).

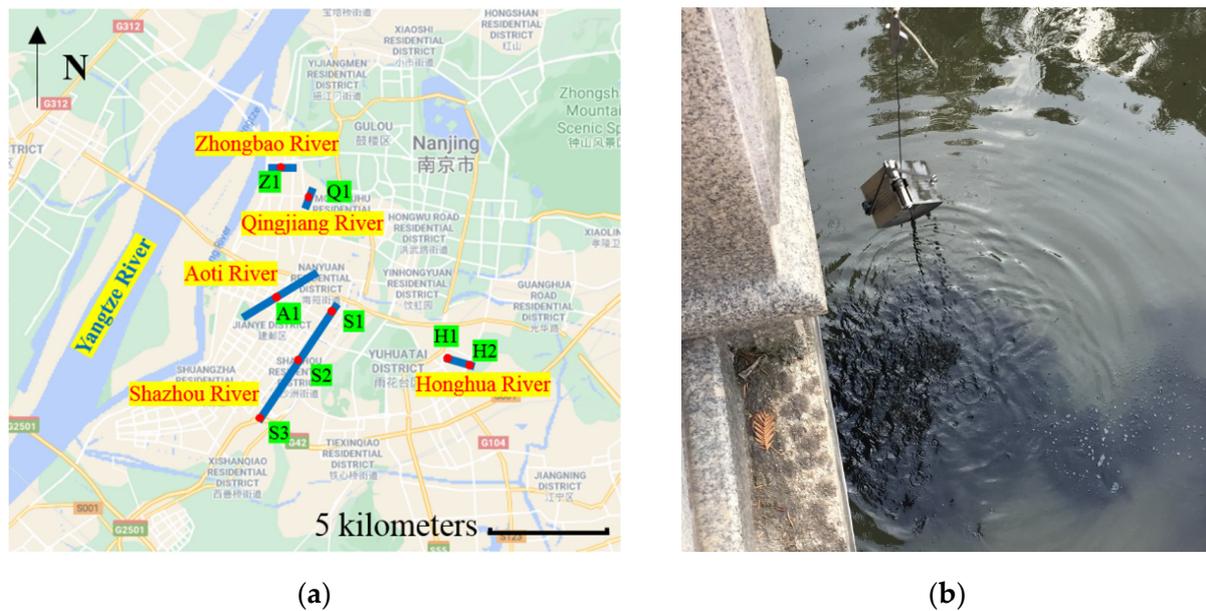


Figure 1. (a) River location and sampling point. (b) Sampling process of the waste slurry.

Table 1. Basic information of the five rivers.

Rivers	Length (km)	Latitude and Longitude of Starting Point	Sampling Point
Zhongbao River	0.72	E118°43'47" N32°3'13"	Z1
Qingjiang River	0.30	E118°44'8" N32°4'13"	Q1
Aoti River	3.00	E118°43'44.7" N32°1'23.2"	A1
Honghua River	0.75	E118°48'22" N32°0'20"	H1, H2
Shazhou River	5.50	E118°42'54.1" N32°58'54.9"	S1, S2, S3

Table 2. Properties of eight types of dredged slurry.

Sampling Point	FA Content ^a (%)	HA Content ^a (%)	Humins Content ^a (%)	Particle Size (μm)		
				D ₁₀	D ₅₀	D ₉₀
Z1	0.35	0.13	1.28	5.5 ^b	36.3	117.6
Q1	0.35	0.13	2.29	3.0	18.4	145.9
A1	0.62	0.62	3.51	8.0	49.6	354.4
H1	0.22	0.14	0.72	6.9	55.2	176.9
H2	0.40	0.25	1.96	3.7	23.3	118.5
S1	0.31	0.09	1.32	4.8	43.3	431.7
S2	0.30	0.00	1.52	2.3	12.7	76.2
S3	0.31	0.06	1.44	3.0	17.9	110.5

^a The humus content (FA, HA and humins) refers to the mass fraction of the carbon of humus in the dry mass of the slurry. ^b In other words, D₁₀ = 5.5 μm means that only 10% of the particles in the slurry are smaller than 5.5 μm .

2.4. Experiment on the Effect of the FA Content on Flocculation and Separation

The effect of the FA content on flocculation and separation was first studied. The slurry (H1 and H2) was first diluted with tap water to a concentration of 5% (*w/w*) to indicate the initial state of the dredged waste slurry. Subsequently, additional FA was added to the slurry, and the FA content of the slurry was adjusted to 0.5%, 1%, 2%, 3%, 4%, 5%, 6% and 7%. After the slurry was uniformly mixed, the slurry of each FA content gradient was added with 611HN or 312VS flocculant with a dosage of 0.025% or 0.05% for flocculation. The PAM dosage was calculated as the ratio of the dry mass of the flocculant to the dry mass of the slurry. Flocculation was conducted in a 500 ml breaker at an agitation speed

of 450 rpm for 2 min. The flocculated slurry was tested for its floc size distribution with a laser particle size analyzer, and the D_{10} increment was used as an index to characterize the flocculation effect. The D_{10} increment was defined as the difference between D_{10} after and before flocculation. The flocculated slurry was subjected to the specific resistance to filtration (SRF). The SRF was used as an index for evaluating the filtration effect. A filtration test was conducted with a Buchner funnel using 0.45 μm filter paper. The effective filtration area of the funnel was 67.17 cm^2 , and the SRF was calculated by measuring the filtrate volume over time as shown in Equation (1):

$$\text{SRF} = \frac{2\Delta PA^2b}{\mu\omega} \quad (1)$$

where ΔP (N/m^2) is the vacuum pressure (set at 60 N/m^2), A (m^2) is the filter area, μ ($\text{N}\cdot\text{s}/\text{m}^2$) is the kinetic viscosity, ω (kg/m^3) denotes the dry solid weight per unit volume cake on the filtrate media and b (s/m^6) is the slope of the curve obtained by plotting the ratio of the time to the filtration volume versus the filtrate volume.

In the field of water treatment, scholars have found that the electrical characteristics of particles affect the removal percentage of humus [21,22]. Therefore, in this study, the zeta potential of slurry with different FA contents was measured. The zeta potential is the electrical potential at the slipping plane. This plane is the interface which separates the mobile fluid from the fluid that remains attached to the surface. The zeta potential was measured through electrophoretic light scattering by using a Zetasizer Nano ZSP (Malvern Instruments, UK). The detailed test scheme for the influence of the FA content on flocculation and separation can be found in Table 3.

Table 3. Test scheme for the influence of the FA content on flocculation and separation.

Slurry	FA Content (%)	Flocculant	Quantitative Indices
H1	0.22, 0.5, 1, 2, 3, 5, 7	312VS: 0.025%, 0.05%	D_{10} increment, SRF, zeta potential
H2	0.40, 0.5, 1, 2, 3, 5, 7	or 611HN: 0.025%, 0.05%	

The floc size tests, SRF tests and zeta potential tests were all conducted parallel to each other three times.

2.5. Flocculation and Separation Tests of Waste-Dredged Slurry

Eight types of waste-dredged slurry from different sources were tested for flocculation and filtration. The slurry was first diluted with tap water to a concentration of 5% to indicate the initial state of the dredged waste slurry. Subsequently, the waste slurry was added to 611HN or 312VS flocculant with a dosage of 0.025% for flocculation. The D_{10} increment, SRF and zeta potential were measured to characterize the effect of flocculation and separation.

3. Results

3.1. The Effect of the FA Content on Flocculation and Filtration

The effect of the FA content on the floc size after flocculation was studied first. Figure 2a,b shows the D_{10} increment of slurry with different FA contents after flocculation. Figure 2a indicates that for the H1 slurry, whether it was added to 611HN flocculant or 312VS flocculant, as the FA content increased from 0.22% to 1.00%, the D_{10} increment decreased rapidly, and the D_{10} increment tended to be stable when the FA content exceeded 1.00%. Figure 2b demonstrates that the D_{10} increment of the H2 slurry had a similar trend to the H1 slurry. The D_{10} increment decreased rapidly when the FA content increased from 0.40% to 3.00%, and the D_{10} increment remained basically unchanged when the FA content was greater than 3.00%.

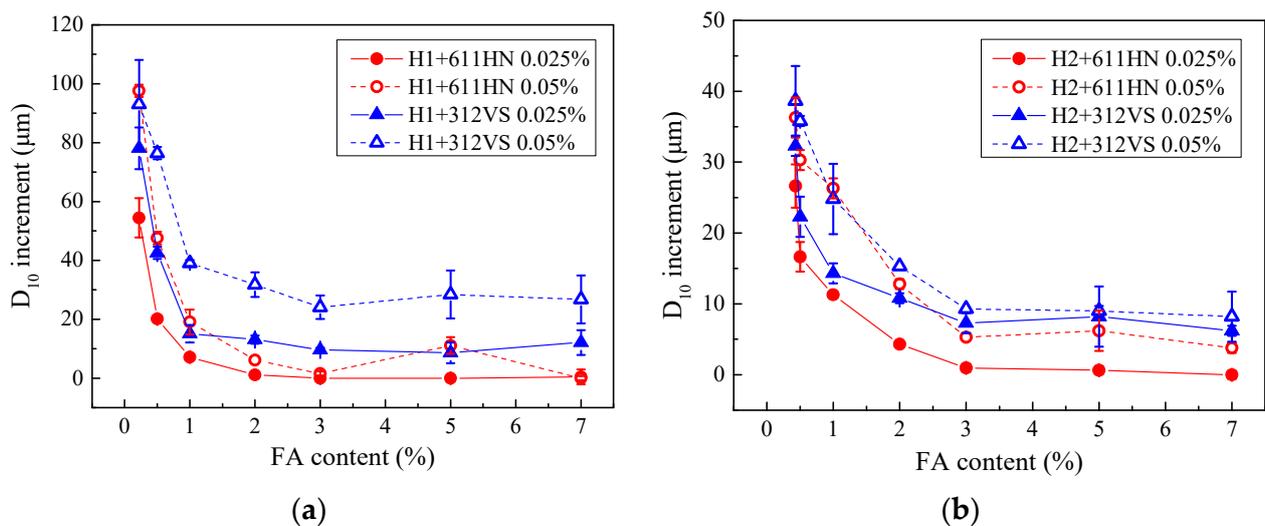


Figure 2. (a) D_{10} increment of the H1 slurry with different FA contents after flocculation. (b) D_{10} increment of the H2 slurry with different FA contents after flocculation.

Figure 3a,b shows the SRF of the H1 and H2 slurry with different FA contents after flocculation. Figure 3a indicates that the SRF increased rapidly and then basically stabilized with the increase of the FA content. The influence of FA on the filtration of the H1 slurry is significant when the FA content was in the range of 0.22~2.00%, and the influence was not obvious when the FA content exceeded 2.00%. The filtration results of the H2 slurry had a similar trend to the H1 slurry. The FA was very sensitive to the effect of filtration when the FA content was between 0.40% and 3.00%, as is shown in Figure 3b. The FA content basically did not affect the SRF when the FA content was greater than 3.00%.

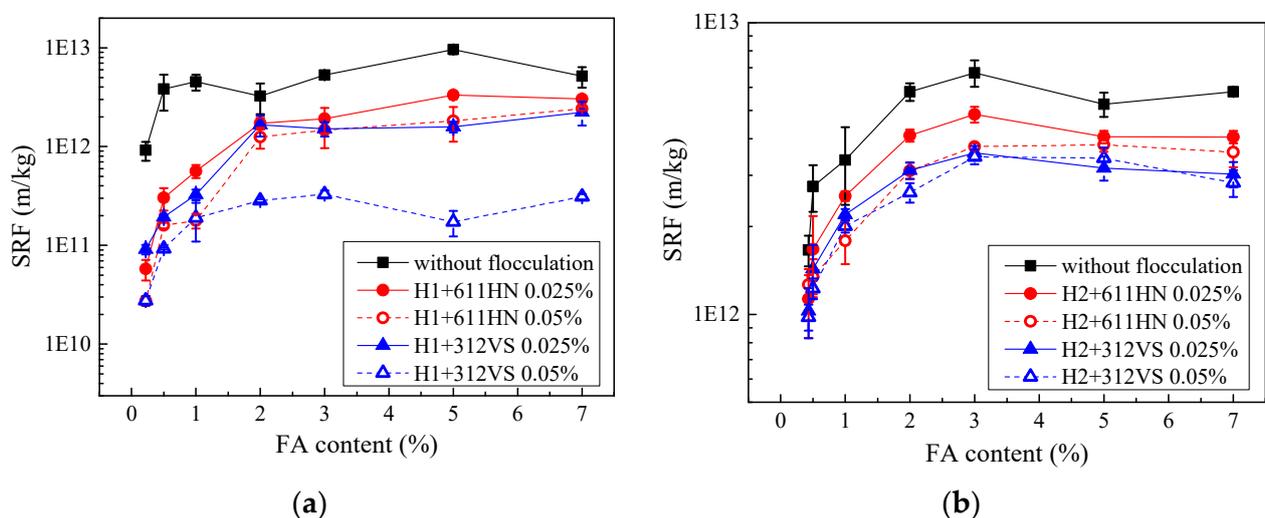


Figure 3. (a) SRF of the H1 slurry with different FA contents after flocculation. (b) SRF of the H2 slurry with different FA contents after flocculation.

3.2. Flocculation and Separation Results of the Eight Dredged Slurry Samples

Figure 4a,b shows the flocculation and separation results of the eight waste slurry samples. Figure 4a illustrates that the eight types of waste slurry had obvious differences in their D_{10} increments after flocculation. The D_{10} increment varied from 8.86 to 78.11 μm with 312VS and varied from 7.33 to 30.19 μm with 611HN. The SRF changed from 5.77×10^{10} to 6.25×10^{12} m/kg (see Figure 4b), showing a significant difference.

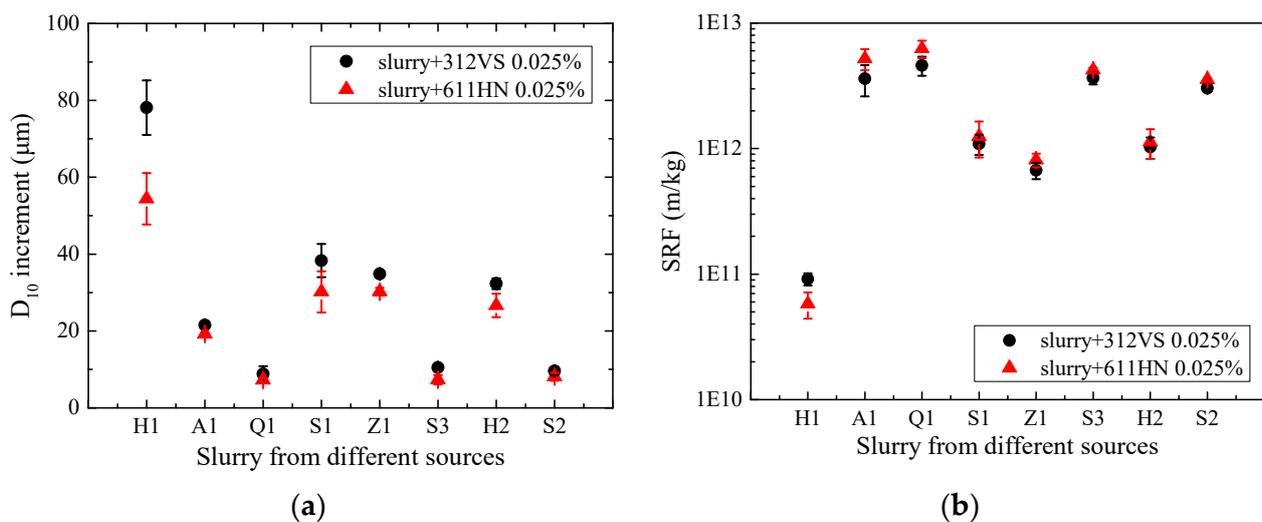


Figure 4. (a) D_{10} increments of the eight dredged slurry samples after flocculation. (b) SRFs of the eight dredged slurry samples after flocculation.

3.3. Zeta Potential Results

Figure 5a shows the zeta potential of H1 and H2 with different FA contents. For the H1 slurry, the zeta potential dropped rapidly from -9.07 to -27.33 mV when the FA content was 0.22~2%. The zeta potential remained basically stable when the FA content increased from 2% to roughly 7%. For the H2 slurry, the zeta potential decreased rapidly from -16.53 to -26.93 mV. The zeta potential was basically unchanged when the FA content exceeded 3%.

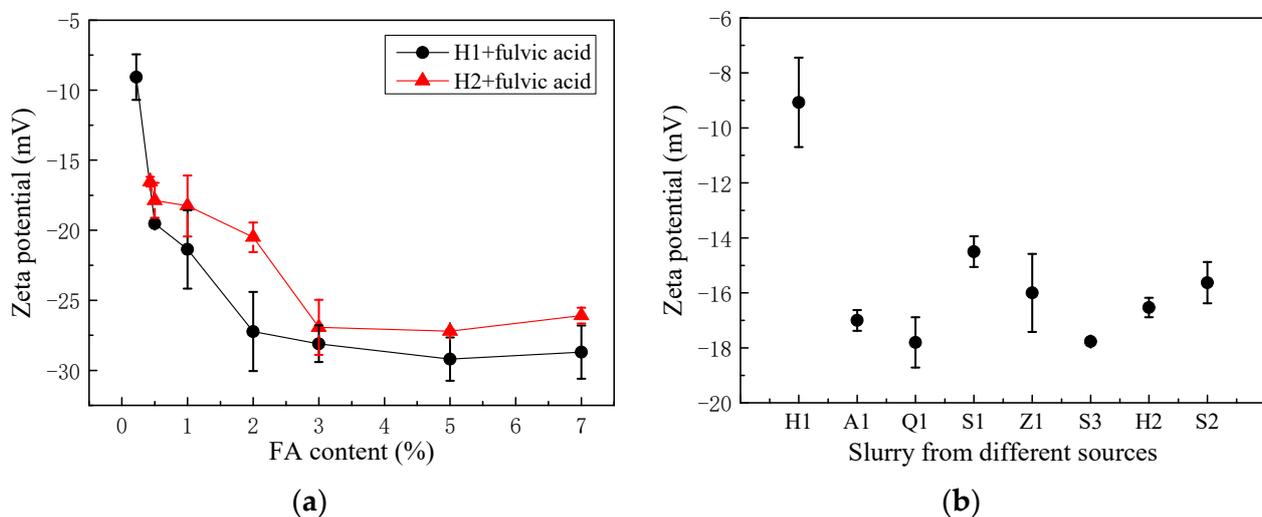


Figure 5. (a) Zeta potentials of H1 and H2 with different FA contents. (b) Zeta potentials of the eight dredged slurry samples.

Figure 5b indicates the zeta potentials of the eight different sources of waste slurry. The zeta potentials of the different slurry samples were obviously different. The absolute zeta potential of the H1 slurry was the lowest (-9.07 mV), and the absolute zeta potential of the Q1 slurry was the highest (-17.8 mV).

4. Discussion

Based on the above results, the influence of FA in waste-dredged slurry from different sources on flocculation and separation was discussed. It can be seen from Figures 2 and 3 that when the FA content was the single variable, FA significantly affected the flocculation

and separation results in the range of 0.22~3%. According to Table 2, the FA content in the waste slurry from different sources was between 0.22% and 0.62%, which was within the range that sensitively affected the results of flocculation and separation. In Figure 4, there are also obvious differences in the flocculation and separation results of the eight waste slurry samples. Therefore, it was necessary to clarify whether FA in the waste slurry was a sensitive factor that affected the results of flocculation and separation.

Pearson correlation analysis between the FA contents in waste slurry from different sources and indices of flocculation and separation (D_{10} increment and SRF) was carried out, as is shown in Figure 6. Figure 6a,b shows the correlation between the FA content of waste slurry and the D_{10} increment, and the Pearson correlation coefficients were -0.36 and -0.29 , respectively. Figure 6c,d shows the correlation between the FA content and the SRF of the waste slurry, and the Pearson correlation coefficients were 0.49 and 0.49 , respectively. The low correlation coefficients revealed that the FA content in the waste slurry was not a sensitive factor that affected the results of flocculation and separation.

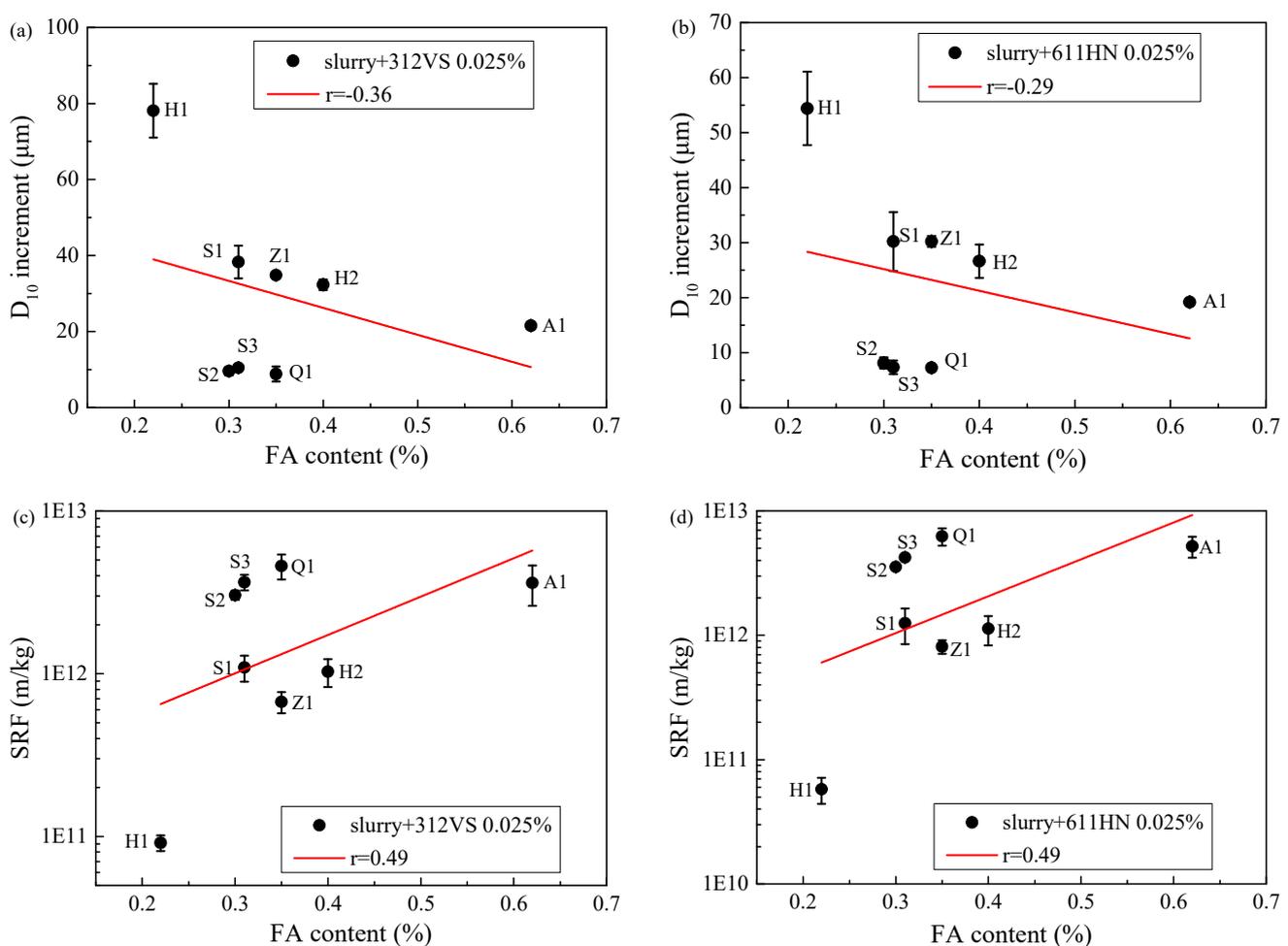


Figure 6. Linear correlation between the FA content and the test results (eight waste slurry samples from different sources). (a) D_{10} increment (0.025% 312 VS). (b) D_{10} increment (0.025% 611HN). (c) SRF (0.025% 312 VS). (d) SRF (0.025% 611HN).

According to the above discussion, the FA content significantly affected the results of flocculation and separation when the FA content was used as the single variable. For the eight waste slurry samples with different FA contents, there were differences in the results of flocculation and separation, but the correlation between the FA content and the flocculation and separation results was low (Figure 6). These two parts of the results seem to be contradictory, so it is necessary to explore further for internal reasons.

The zeta potential of the H1 and H2 slurry decreased rapidly and then stabilized with the increase of the FA content in Figure 5a, which was similar to the trend of results for flocculation and separation in Figures 2 and 3. Therefore, it could be assumed that FA affected the results of flocculation and separation by affecting the zeta potential of the slurry. To prove this point, the H1 slurry was taken as an example to analyze the correlation between the FA content and the results of flocculation and separation, as shown in Figure 7, which demonstrated high relevance ($r = 0.89, 0.93, 0.90, 0.98$). This basically proved that adding FA to the slurry essentially changed the zeta potential of the slurry and then influenced the results of flocculation and separation. The zeta potential changed significantly ($-9.07 \sim -27.23$ mV) when the FA content was in the range of 0~3%, so the effect of flocculation and separation also changed significantly. The zeta potential was basically unchanged ($-27.33 \sim -28.70$ mV) when the FA content increased from 3% to roughly 7%, so the effect of flocculation and separation trended toward being stable.

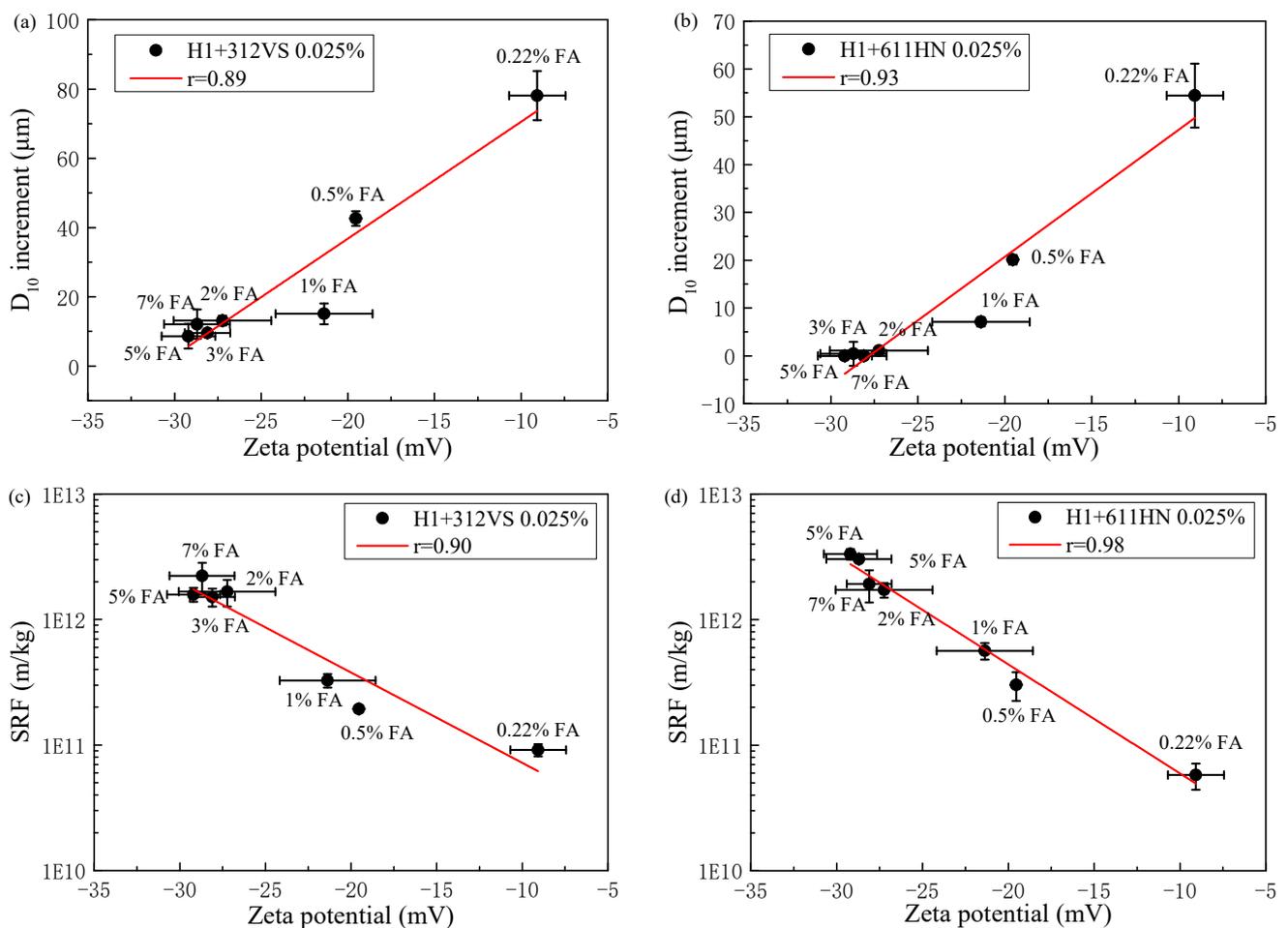


Figure 7. Linear correlation between the zeta potential and test results (H1 slurry with different FA contents). (a) D_{10} increment (0.025% 312 VS). (b) D_{10} increment (0.025% 611HN). (c) SRF (0.025% 312 VS). (d) SRF (0.025% 611HN).

The zeta potential of waste slurry from different sources and the flocculation and separation results were analyzed for correlation. As is shown in Figure 8, the zeta potential of the waste slurry had good correlation with the D_{10} increment ($r = 0.92, 0.87$) and the SRF ($r = 0.91, 0.92$). Although the properties of the waste slurry from different sources were quite different, the zeta potential of the waste slurry was the main factor affecting the flocculation and separation results.

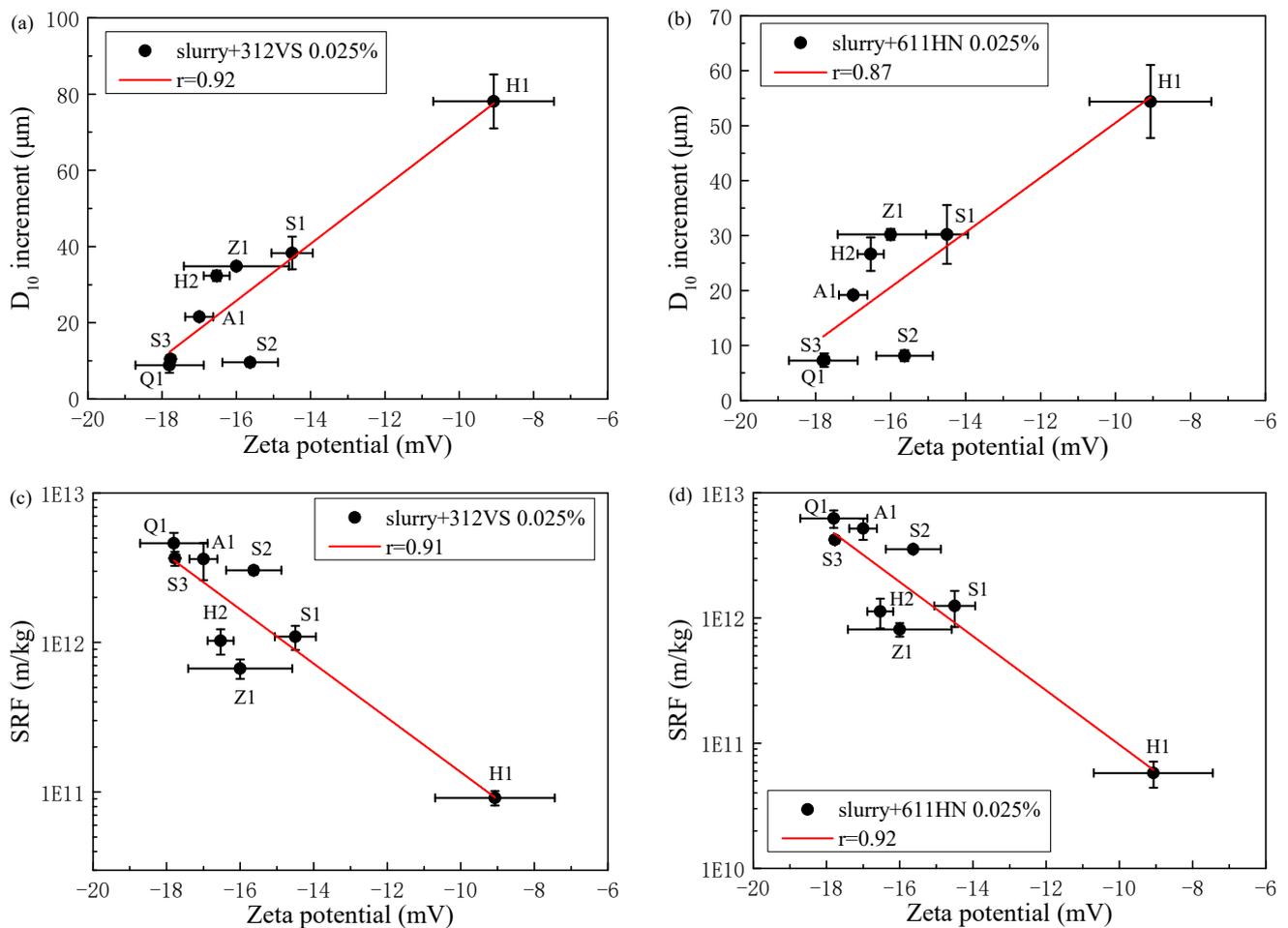


Figure 8. Linear correlation between the zeta potential and test results (eight waste slurry samples from different sources). (a) D_{10} increment (0.025% 312 VS). (b) D_{10} increment (0.025% 611HN). (c) SRF (0.025% 312 VS). (d) SRF (0.025% 611HN).

FA in waste slurry was not a sensitive factor that affected the flocculation and separation results. The reason for this could be that not only could FA affect the zeta potential, but other factors in the slurry could also affect the zeta potential. In the research of soil and water conservation, it has been found that clay minerals significantly impact the flocculation effect [23–25], and the flocculation results change when clay minerals and humus are present simultaneously [26,27]. Therefore, it could be inferred that although FA affected the flocculation and separation results, other components in the slurry also affected the results. All these components in the slurry probably impacted the flocculation results by changing the zeta potential, which needs to be studied further.

5. Conclusions

(1) This paper carried out an experiment on the influence of the FA content on the flocculation and separation results. The effect was significant when the FA content was in the range of 0~3% and not obvious when the FA content exceeded 3%.

(2) There were significant differences in the flocculation and separation of dredged waste slurry from different sources. Under the same flocculation conditions, the D_{10} increment could differ by nearly 10 times, and the SRF differed by 2 orders of magnitude. The FA content in different waste slurry samples ranged from 0.22% to 0.62%, but FA was not a sensitive factor affecting the flocculation results.

(3) Fulvic acid mainly affected the results of flocculation and separation by affecting the zeta potential of the waste slurry. The zeta potential in the dredged slurry determined the flocculation and separation results, and fulvic acid was only one of the factors that

affected the zeta potential. Therefore, in the design of the flocculation and dewatering of waste slurry, only the zeta potential of the slurry needs to be considered, and the FA content is not necessary to consider.

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Conflicts of Interest: The authors declare no conflict of interest.

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