

## SS and CODremoval in Artificial Stone Wastewater Using Coagulation Process

Ping ZHANG\*

Collaborative Innovation Center for Water Pollution  
Control and Water Safety in Karst Area  
Guilin University of Technology  
Guilin 541004, China  
e-mail: 497474987@qq.com, www.glut.edu.cn

Lin-feng CHEN

Collaborative Innovation Center for Water Pollution  
Control and Water Safety in Karst Area  
Guilin University of Technology  
Guilin 541004, China

Qi-meng ZHANG

Collaborative Innovation Center for Water Pollution  
Control and Water Safety in Karst Area  
Guilin University of Technology  
Guilin 541004, China

Sheng-min WANG

Collaborative Innovation Center for Water Pollution  
Control and Water Safety in Karst Area  
Guilin University of Technology  
Guilin 541004, China

Qing-lin XIE

Collaborative Innovation Center for Water Pollution  
Control and Water Safety in Karst Area  
Guilin University of Technology  
Guilin 541004, China

Nan-chun CHEN

College of Materials Science and Engineering and  
Ministry-province Jointly-constructed Base for State  
Key Laboratory of Processing for Nonferrous Metals  
and Featured Materials  
Guilin University of Technology  
Guilin, 541004, China  
e-mail: cnc@glut.edu.cn

**Abstract**—The obvious features of artificial stone wastewater are high SS and COD concentration, and the wastewater cannot meet the discharge standard. Three-stage coagulation procedure was adopted to dispose wastewater by controlling the different coagulants, coagulant dosage and pH value in this study. The results showed that aluminum sulfate was acted as coagulant and the dosages were 56.70mg/L, 16.00mg/L and 7.50mg/L respectively in the three-stage coagulation procedure, the supernatant SS of wastewater was 8.8mg/L, the turbidity was 6.9 NTU and COD was 73.65 mg/L. Based on the three-stage coagulation procedure, the supernatant was further to be adsorbed by diatomite. When the pH was 6.40, the dosage was 20 g/L, and the supernatant COD was 16.80 mg/L, the supernatant, treated by three-stage coagulation procedure, can be directly used for production.

**Keywords**—Artificial stone processing wastewater; SS and COD; Coagulation; Adsorb Introduction.

### I. INTRODUCTION

The main pollutants of artificial stone processing wastewater were SS and COD. It was result from stone powder came from stone cutting and edging, and the residues of auxiliary raw material, used for processing, such as resin adhesive, curing agent and so on Powder in the water showed the state of suspended and colloidal, the concentration of SS was 1000mg/L or more[1-3]. The main components of residues were resin acids and their saponification products, unsaturated fatty acids and their

saponification products, lignin and their degradation products, etc. At present, only part of SS can be removed by coagulation and sedimentation [4-9].

### II. MATERIAL AND METHODS

Wastewater from Guangxi Li Sheng Stone Industry Co., Ltd., the water property was present in table 1.

TABLE I. THE WATER QUALITY OF ARTIFICIAL STONE PROCESSING WASTEWATER

index	SS(mg/L)	Turbidity(NTU)	COD(mg/L)
value	1688	1145	8640

Three Coagulants, aluminum sulfate, poly aluminum, ferric chloride, were used in the experiment. The coagulant was prepared into 10 g/L solution.

The first coagulation: 600mL wastewater was took into a 1000mL stirring cup, the wastewater was adjusted to a pH value, a certain amount of coagulation was added into the stirring cup and stirred with six-unit coagulant mixer. The process of six-unit coagulant mixer: the first stage, stirred for 30s, with a speed of 300r/min; the second stage, stirred for 3min, and the stirring speed was 150r/min; the third stage, stirred for 5min, the speed was 150r/min. After then rest for 20min, the supernatant was taken and the COD, SS and turbidity were measured.

The second coagulation: 500mL supernatant of the first coagulation was added into a 1000mL stirring cup, and then dispose the supernatant with the same as the first coagulation

stirring process, Resting for 20min after completing stir, the supernatant was taken, and then COD, SS and turbidity were measured.

The third coagulation: 400mL supernatant of the second coagulation was added into a 1000mL stirring cup, adjusted the pH value, added in a certain amount of coagulation and stirred with six-unit coagulant mixer with the same as the first coagulation stirring process, Resting for 20min after completing stir. Finally, the COD, SS and turbidity were measured in the supernatant. Absorption: 50mL supernatant of the third coagulation was injected into 100mL centrifuge tube, and different dosage diatomite was added into the centrifuge tubes respectively. The supernatant was adjusted to a pH value. After then oscillating the tubes with Constant water bath panoscillating for 6h, Centrifuged the sample with a speed of 4000 r/min for 5min, then COD in the supernatant was measured.

COD was measured by dichromate digestion in a Microwave Oven with ECH-II microwave heater; SS was tested by weight method with WSZ-100 and WSZ-800 turbidity meter; the pH value was measured with a UB-7 pH meter; the coagulation experiment was carried out with a ZR4-6 type six-blending coagulator.

### III. RESULTS AND DISCUSSION

#### A. The Effect of Different Coagulants on COD and SS

Aluminum sulfate, poly aluminum chloride, ferric chloride were selected as coagulant, the wastewater was adjusted to the different pH values, the dosage of three coagulants was 83.33mg/L (table 2 to 4)

TABLE II. COAGULATION OF ALUMINUM SULFATE WITH DIFFERENT PH VALUES

pH	SS (mg/L)	Turbidity (mg/L)	COD (mg/L)	Removal SS (%)	Removal turbidity (%)	Removal COD (%)
5.6	10.2	35	6235.44	99.39	96.86	27.83
6.4	10.2	32	5995.34	99.40	97.13	30.61
6.9	8.2	25	5448.61	99.51	97.76	36.94
7.6	8.1	15	5048.58	99.52	98.65	41.57
8.4	9.2	21	5236.32	99.45	98.12	39.39
10.0	8.5	25	5246.86	99.50	97.76	39.27

With the increasing of pH value (Table2), the removal rate of COD increased gradually. In addition, when the pH value was 7.6, the removal rate of COD was the best with the value of 41.57%. However the pH value was range from 8 to 10, removal rate of COD was decreased slightly. Besides, after coagulation the removal rate of SS and turbidity in wastewater both reached over 99% and 95% respectively.

Table 3 was shown that poly aluminum chloride was selected as the coagulant, with the increasing of pH value, the removal rate of COD increased gradually. When the pH value was 10.0, the removal rate of COD was the best with 37.77%. The removal rate of SS and turbidity both reached over 99% and 95% respectively after coagulation.

That ferric chloride was selected as the coagulant, with the increasing of pH value, the removal rate of COD

increased gradually. When the pH value was 10.0, the removal rate of COD was the best, and the value was 33.65%. The removal rate of SS and turbidity both can reach over 99% and 95% respectively after coagulation.

It was observed that the three coagulants can remove SS and turbidity in the wastewater efficiently. Although the removal rate value of COD was not high, 1853.44~3591.42mg/L of COD can be removed, and the coagulation effect of aluminum sulfate was better than aluminum chloride and ferric chloride. Therefore, aluminum sulfate was the best coagulant, and its optimum pH was 7.6. The coagulation experiments with different dosages were made with aluminum sulfate as coagulant (Table 5).

With the increasing of dosage, the removal rate of COD was increased gradually and the best was 43.86% when the dosage was 56.7mg/L, but the removal rate declined when added the dosage continually. Thus the best dosage of coagulant was 56.7mg/L. After coagulation, the removal rate of SS can reached over 99%, and the removal rate of turbidity was above 95%.

Under the dosage was 56.7 mg/L, pH=7.6, removal rate of SS and turbidity were high. After the coagulation, SS was 8.2 mg/L and the turbidity was 16 NTU, meeting the discharge standard, but COD removal rate just was 43.86%. Although the value of COD was declined from 8640.00 mg/L to 4850.21mg/L, it was still beyond the reach of discharge standard. Therefore, to proceeding the second and third coagulation on the basis of the first coagulation experiment was necessary for treatment process.

TABLE III. COAGULATION OF POLY ALUMINUM CHLORIDE WITH DIFFERENT PH VALUES

pH	SS (mg/L)	Turbidity (mg/L)	COD (mg/L)	Removal SS (%)	Removal turbidity (%)	Removal COD (%)
5.6	10.4	55	6304.66	99.38	95.07	27.03
6.4	10.2	43	6179.35	99.40	96.14	28.48
6.9	8.3	41	5758.88	99.51	96.32	33.35
7.6	8.3	35	5696.68	99.51	96.86	34.07
8.4	9.2	36	5536.32	99.45	96.77	35.92
10.0	8.7	36	5376.62	99.48	96.77	37.77

TABLE IV. COAGULATION OF FERRIC CHLORIDE WITH DIFFERENT PH VALUES

pH	SS (mg/L)	Turbidity (mg/L)	COD (mg/L)	Removal SS (%)	Removal turbidity (%)	Removal COD (%)
5.6	10.7	55	6786.56	99.37	95.07	21.45
6.4	10.5	43	6554.87	99.38	96.14	24.13
6.9	10.3	41	6223.45	99.39	96.32	27.97
7.6	9.7	35	6088.66	99.43	96.86	29.53
8.4	9.2	36	5936.77	99.45	96.77	31.29
10.0	8.5	36	5732.46	99.50	96.77	33.65

**TABLE V. THE RESULTS OF DIFFERENT DOSAGES THAT ALUMINUM SULFATE AS COAGULANT AND THE OPTIMUM PH=7.6**

Dosages (mg/L)	SS (mg/L)	Turbidity (mg/L)	COD (mg/L)	Removal SS (%)	Removal turbidity (%)	Removal COD (%)
16.7	10.1	32	5732.96	99.40	97.13	33.65
30.0	9.9	31	5386.44	99.41	97.22	37.66
43.3	8.5	23	5055.34	99.50	97.94	41.49
56.7	8.2	16	4850.21	99.51	98.57	43.86
70.0	9.0	21	5046.64	99.47	98.12	41.59
83.3	8.7	23	5089.45	99.49	97.94	41.09

### B. The Effects of the Second and Third Coagulation Selecting Aluminum Sulfate as Coagulant

The dosage of aluminum sulfate was 56.7mg/L, the first coagulation supernatant (COD was 4850.21mg/L, SS was 8.2 mg/L) under the dosage of coagulant was 56.7mg/L and pH=7.6 was adjusted to the various values of pH, and carried out the second coagulation, the results were present in table 6. Table 7 was shown the experimental results on the conditions that optimum pH and different dosages. There were no analysis about the removal of SS and turbidity, on account of the supernatant containing the low concentration of SS and turbidity. Therefore, only the SS and turbidity values of the supernatant after second coagulation were listed.

**TABLE VI. THE SECOND COAGULATION RESULTS OF DIFFERENT PH AND ALUMINUM SULFATE AS COAGULANT**

pH	SS (mg/L)	Turbidity (mg/L)	COD (mg/L)	Removal COD (%)
5.6	12.4	6.5	488.56	89.93
6.4	10.2	8.0	496.43	89.76
6.9	10.2	7.6	464.68	90.42
7.6	8.9	7.5	448.35	90.76
8.2	10.3	5.7	482.36	90.05
10.2	9.6	10.5	472.86	90.25

The removal efficiency of COD was the best when pH value was 7.6, with the value of 90.76%. The SS of the wastewater after coagulation was 8.9 mg/L, the turbidity was 7.5 NTU and the COD was 448.35 mg/L.

That optimal pH value of 7.6, the removal rate of COD increased with the increasing of the dosages, but it increased slowly when the dosage was more than 16 mg/L. Thus, from the point of view of economic, the optimum dosage was 16mg/L. After the second coagulation, the SS, turbidity and COD of supernatant were 10.4 mg/L, 10.1NTU and 462.42 mg/L respectively. As a result of the secondary coagulation, the COD removal efficiency and the removal rate were more than 90%, but the COD of effluent was 462.42 mg/L, which could not meet the discharge standard. Therefore, it is essential to carry out the third coagulation based on the coagulation of previous experiment. The optimum pH values of the first and second coagulation were 7.6, and the third

coagulation pH was also 7.6. During carrying out the third coagulation of different dosages (table 8).

**TABLE VII. THE SECOND RESULTS OF DIFFERENT DOSAGES AND OPTIMUM PH, ALUMINUM SULFATE AS COAGULANT**

The dosages (mg/L)	SS (mg/L)	Turbidity (mg/L)	COD (mg/L)	Removal COD (%)
8	13.6	10.9	531.98	89.03
12	13.2	10.3	495.78	89.78
14	12.2	10.3	488.67	89.92
16	10.4	10.1	462.42	90.47
30	9.8	10.0	460.15	90.51
44	8.8	8.4	456.46	90.59
56	8.0	8.2	450.96	90.70
70	8.6	7.8	438.35	90.96
84	13.0	10.2	446.36	90.80

That optimal pH value of 7.6, the COD removal rate increased with the increasing of the dosages. The dosage of 7.5mg/L was the best, however the dosage increased continually, the removal rate of COD was decreased, so the optimum dosage was 7.5mg/L. After the third coagulation, SS was 8.8mg/L, turbidity was 6.9NTU, and COD was 73.65mg/L. It could meet the Integrated Wastewater Discharge Standard. The removal rate of SS, turbidity and COD was 99.48%, 99.38% and 99.15% respectively.

**TABLE VIII. THE THIRD RESULTS OF DIFFERENT DOSAGES AND OPTIMUM PH, ALUMINUM SULFATE AS COAGULANT**

The dosages (mg/L)	SS (mg/L)	Turbidity (mg/L)	COD (mg/L)	Removal COD (%)
2.5	8.6	7.1	80.00	82.70
5	8.0	7.0	80.00	82.70
7.5	8.8	6.9	73.65	84.07
10	8.9	6.4	78.40	83.05
15	9.8	7.4	120.45	73.95
30	9.6	9.3	136.46	70.49
45	8.9	4.7	144.48	68.76
60	9.4	6.8	200.64	56.61
75	9.3	8.3	144.00	68.86
90	10.2	13.2	128.00	72.32

### C. The Effect of Diatomite Adsorption on the Removal of COD and SS

After the three times coagulation, the removal rate of SS, turbidity and COD were more than 90%, but the rest of COD was 73.65mg/L, still beyond the discharge standard. Therefore, experiment of adsorption treatment was further to be done continually. The diatomite was chosen as adsorbent and took the supernatant (COD was 86.4mg/L) after coagulation 50mL, the pH value was adjusted to 5.6, 6.4, 6.9, 7.6, 8.4 and 10.0 respectively [10]. 5g adsorbent was added to each of the supernatant. Adsorption results. Thus the optimum pH was 6.4.

50mL supernatant (COD was 86.4mg/L) after coagulation was taken, the pH value was adjusted to 6.4, and then the different dosages of adsorbent was added to the supernatant. Under the condition that optimal pH value of 6.4, the COD removal rate increased with the increasing of

the dosages of diatomite. When the dosage of diatomite was 1g, the removal rate was the highest. If the dosage increased continually, the removal rate of COD decreased (fig.3). Thus the optimum dosage of diatomite was 20g/L. The COD of the supernatant after absorbing was 16.18 mg/L, and the removal rate of COD was 80.56%, reaching the Integrated Wastewater Discharge Standard.

#### IV. CONCLUSIONS AND SUGGESTION

The comparison showed that aluminum sulfate was the suitable coagulant, and the dosages of three times coagulation were 56.7mg/L, 16mg/L and 7.5mg/L respectively. The supernatant obtaining after treating the artificial stone wastewater with coagulation, contained the SS with the concentration of 8.8mg/L, the turbidity with the concentration of 6.9NTU, the COD with the concentration of 73.65mg/L, the supernatant after coagulation met the secondary standard number in GB8978-1996. When the pH was 6.4 and the dosage was 20g/L, diatomite as the adsorbent treated the supernatant obtained after the coagulation with adsorbing. The COD could decrease to 16.8mg/L reaching the primary standard number A level (COD was 50mg/L). At last, it could be used for production directly.

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#### REFERENCES

- [1] Yong Li, Guofeng Li, Hui Yan. The treatment of wastewater in stone industry. *Stone*, 2016, 05:32-34 (in Chinese).
- [2] Anonymous. Environmental Research; Reports from Afyon Kocatepe University advance knowledge in environmental research. *Ecology, Environment and amp; Conservation*, 2009.
- [3] Minghui Xie. Discussion about the countermeasures of pollution control in Stone Industry. *Environmental Management*, 2007, 06:63-64 (in Chinese).
- [4] Shaohua Feng. Practical technology for wastewater treatment in stone slab processing. *China Water and Wastewater*, 2010, 14:109-112 (in Chinese).
- [5] Xiongping Lin. Analysis on the recycling method about the treatment of wastewater in stone industry. *Chemical Engineering and Equipment*, 2009, 05:191-192 (in Chinese).
- [6] Weixun Chen, Yanhong Chen, Jiyu Zhang. The study of granite processing wastewater flocculation treatment technology. *Environmental Engineering*, 2011, 01:46-50 (in Chinese).
- [7] Yunfeng Zhang, Meilin Huang, Xiuzhen Xu. Discussion on treatment method of wastewater from stone processing. *Energy and Environment*, 2011, 05:84-85 (in Chinese).
- [8] Bo Xu. Example of treatment engineering on Stone wastewater. *Techniques and Equipment for Environmental Pollution Control*, 2003, 08:81-83 (in Chinese).
- [9] Liming Cai, Kaihan Chen, Xiaoshuang Hong, Tingfei Wang. Study on lime demulsification/brine flocculation process for wastewater treatment in stone industry. *Industrial Water Treatment*, 2012, 02:51-55 (in Chinese).
- [10] Shaoying Guo. Advanced treatment on tail water from municipal wastewater treatment plant by modified diatomite. Master Dissertation, Fujian Normal University, 2010 (in Chinese).