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Evaluation of binary surfactants on the flotation performance of incinerator fly ash

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ABSTRACT

Carbon constituents and polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) in incinerator fly ash can be removed by floatation, but the floatation performance is poor due to weathering. The effects of four binary surfactants (i.e., SDS-Tween 80, CTAB-Tween 80, Span 20-Tween 80 and SDS-CTAB) on the flotation process were discussed. Results show that SDS-Tween 80 with the mixture ratio of 3:7 is superior to the other surfactant mixtures in decarburization and PCDD/Fs removal, and can remove 91.4% of carbon constituents and 89.3% of PCDD/Fs. The content of PCDD/Fs in the tailings is 2.6 ng I-TEQ/g which meets the landfill standard of PCDD/Fs.

Keywords: Incinerator fly ash; Flotation; Binary surfactant; Decarburization; Polychlorinated dibenzo-p-dioxins and dibenzofurans

1. Introduction

Coronavirus Disease 2019 (COVID-19) is a major global public health concern due to high morbidity and can be transmitted through exposure to infected hospital solid waste (HSW). The increasing use of medical services results in the generation of excess infectious HSW including discarded disposable masks, gloves and other protective equipment. If HSW is not disposed properly, it will inevitably result in secondary infection and the spread of the epidemic. Incineration is the major disposal pathway for infectious HSW in China and can destroy pathogens and reduce the weight of waste by more than 70%. However, hospital solid waste incinerator (HSWI) fly ash was also classified as hazardous waste due to its high concentrations of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and toxic heavy metals [1–3].

HSWI fly ash also contains considerable amounts of carbon constituents including unburned carbon (UC) and powder activated carbon (PAC) which are injected into the flue gas purification system to absorb PCDD/Fs. It is well known that PCDD/Fs and other organic pollutants in fly ash are enriched in UC during incineration [4]. Every gram of PAC can absorb 105~115 ng PCDD/Fs, so the adsorption capacity of PAC is remarkable [5]. Therefore, carbon constituents are the enrichment source of PCDD/Fs in HSWI fly ash [6]. Flotation is a physical–chemical separation technology, which is generally used to treat mineral, fly ash, wastewater and polluted soil. It is found that PCDD/Fs and carbon constituents in HSWI fly ash are both

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lipophilicity and hydrophobic, which facilitate simultaneous removal of these two substances by flotation [7].

As the daily output of HSWI fly ash is relatively small (about 300–500 kg/d), most HSWI fly ash is usually put into designated stockpiles within 3–6 months before treatment [8]. During storage, the surface of UC may be oxidized naturally to form more oxygen-containing functional groups. Therefore, the weathering process can reduce the hydrophobicity of the UC surface and have a great negative impact on its floatability, which makes it difficult to float out the carbon constituents with common oily collectors alone [2,9], but the addition of surfactants can improve the flotation performance [10].

In the decarburization flotation process, the addition of surfactant has the following advantages: (a) the adverse effect of oxygen-containing groups on the surface of UC is eliminated by the adsorption between the surfactants and surfaces of carbon particle [11,12]; (b) improving the wettability of carbon particles make kerosene easily adhere to the surface of carbon particles; (c) the emulsification and dispersion of kerosene in the slurry is improved, which increase the collision probability of kerosene and carbon particle constituents. Therefore, the flotation efficiency can be improved [13]. The types of surfactants mainly include cationic, anionic, and non-ionic surfactants which have a great influence on flotation performance [14,15]. In our previous study, it was found that the single non-ionic Tween 80 had higher carbon and PCDD/Fs removal efficiency than cationic cetyltrimethylammonium bromide (CTAB), anionic sodium dodecyl sulfate (SDS) and no surfactant. Furthermore, the optimum flotation condition was determined as follows: Tween 80 concentration of 0.015 g/kg ash and slurry pH = 7 [16]. Compared with a single surfactant, the mixture of dual surfactants has a synergistic or antagonistic effect and is widely used in mineral flotation, soil washing and washing fields [17,18]. Surfactant mixtures can not only obtain relatively high activity, but also reduce the total amount and the cost of surfactants [19]. The excellent performance of mixed surfactants has stimulated many researchers to explore the potential combination patterns [20]. Ahn et al. [21] compared the effect of single and binary surfactant mixtures on the adsorption of Cd^{2+} by granular activated carbon. Yuan et al. [22] found the desorption ability of hexachlorobenzene from spiked kaolin with surfactant mixtures of SDBS-Tween 80 was better than that with single sodium dodecyl benzene sulfonate (SDBS). Huang et al. [23] disclosed that the synergistic effect of binary surfactant mixtures (Span 20-Tween 80) was higher than that of the single surfactant at the optimum hydrophilic–lipophilic balance (HLB) value. However, there is little research on the effect of mixed surfactants on the flotation performance of incinerator fly ash.

The objective of this research is to select appropriate binary surfactants to promote the removal of carbon constituents and PCDD/Fs in HSWI fly ash. Four kinds of dual mixed surfactant (i.e., anionic–non-ionic, cationic–non-ionic, non-ionic–non-ionic and anionic–cationic) were prepared to investigate the flotation performance of HSWI fly ash. The effect mechanism was expressed by measuring the zeta potential and surface tension.

2. Materials and methods

2.1. Materials

The fly ash sample was produced in the rotary kiln of HSW incineration plants in southern China, collected from the bag filter and stored at the garbage dump for 6 months. The moisture content of the ash sample is 9.5%. The ash sample was homogeneously mixed and screened to remove particles larger than 840 μm, and then was dried at 378 K for 24 h.

2.2. Methods

All flotation tests were carried out in a 1 L Denver flotation cell with an impeller speed of 2,000 rpm, an airflow rate of 1.2 L/min, and a slurry concentration of 100 g/L. The kerosene of 3.0 g/kg ash and methyl isobutyl carbinol (MTBC) of 0.1 g/kg ash were added into the slurry respectively and stirred before flotation [16]. After flotation, froths and the tailings are filtered, dried and weighed respectively, and then analyzed.

In this study, four kinds of dual mixed surfactants (SDS-Tween 80, CTAB-Tween 80, Span 20-Tween 80 and SDS-CTAB) at certain ratios were added to improve the floatability (Table 1). The mixed rations were ranged from 1:9 to 9:1. To facilitate comparison, other operating conditions were kept constant and the pH value of the original slurry was adjusted to 7 at a fixed surfactant dose of 0.015 g/kg ash. In order to evaluate the effect of dual mixed surfactants on decarburization performance, carbon removal efficiency (CRE) was evaluated.

$$\text{Carbon recovery efficiency}(\%) = \frac{\text{Carbon}_{\text{Froths}} \times \text{Mass}_{\text{Froths}}}{\left(\text{Carbon}_{\text{Froths}} \times \text{Mass}_{\text{Froths}} + \text{Carbon}_{\text{Tailings}} \times \text{Mass}_{\text{Tailings}}\right)} \quad (1)$$

where $\text{Carbon}_{\text{Froths}}$ is the mass fraction of carbon in the froths, in %; $\text{Mass}_{\text{Froths}}$ is the mass of froths, in g; $\text{Carbon}_{\text{Tailings}}$ is the mass fraction of carbon in the tailings, in %; $\text{Mass}_{\text{Tailings}}$ is the mass of tailings, in g.

Table 1
Characteristics of used surfactants

Surfactant	Structural formula	Critical micelle concentration (mol/L)	Hydrophilic–lipophilic balance	Type
Tween 80	$C_{64}H_{124}O_{27}$	1.2×10^{-5}	15.0	Non-ionic
Span 20	$C_{18}H_{34}O_6$	6.1×10^{-5}	8.6	Non-ionic
SDS	$C_{12}H_{25}SO_4Na$	8.3×10^{-3}	13	Anionic
CTAB	$C_{19}H_{42}BrN$	9.2×10^{-4}	15.8	Cationic

The contents of 17 toxic PCDD/Fs congeners were analyzed by high resolution gas chromatography-high resolution mass spectrometry (HRGC-HRMS). The sample pretreatment was conducted according to the modified version of the US EPA Method 23 (20), and the detailed analytic method was described in our previous paper [16]. Three replicates of PCDD/Fs analyses were made for obtaining reliable data and the results were determined as the average of three measurements.

The surface tension is determined by DataPhysics DCAT 21 combined with the William plate method. Zeta potential was measured by a zeta potential 230 analyzer at room temperature [16]. The loss on ignition (LOI) for the sample was determined as the weight loss at 1,123 ± 25 K for 3 h, and results showed that the LOI of fly ash was 15.8% because it contained a large amount of PAC. Major compounds in HSWI fly ash were shown in Table 2.

Table 2
Major compounds in HSWI fly ash

SiO_2	14.3
CaO	23.6
Al_2O_3	3.65
Fe_2O_3	3.31
MgO	1.09
K_2O	4.58
Na_2O	17
SO_3	4.83
Cl	22.4
TiO_2	0.99
F	1.19
LOI	15.8

3. Results and discussion

3.1 *Effect of mixed surfactants on decarburization performance*

Different types of single surfactants promote different mechanisms of flotation decarbonization. Anionic SDS can be adsorbed on the oxidized surface of the carbon constituents, which increases the hydrophobicity of the carbon surface. Cationic CTAB might interact with the UC surface by electrostatic attraction [15]. The polar oxygenated functional groups of non-ionic Tween 80 and Span 20 might interact with the oxygenated sites on the UC particle surface [24]. Furthermore, their nonpolar aliphatic chain might interact with PAC by hydrophobic bonding [15,25].

The effect of mixed ratios of surfactant mixtures on CRE and LOI of the tailings are shown in Fig. 1. When SDS-Tween 80 mixture was used as the surfactant, CRE firstly increased and then steadily decreased. While mixed ratio = 3:7, CRE reached its maximum (91.4%) which is higher than 90.6% and 82.1% of singe Tween 80 and SDS [16]. Under this condition, the LOI of the tailings was the minimum (5.4%) which was lower than 6.0% stipulated by the American Society for Testing Material. When cationic CTAB-Tween 80 mixture was used as the surfactant, the CRE decreased with the increase of mixing ratio. The highest CRE (85.9%) was obtained at the mixed ratio = 1:9, while the LOI of the tailings acquired a minimum (9.1%). For two kinds of non-ionic surfactant mixtures of Span 20-Tween 80, the best result was attained at mixed ratio = 3:7. Besides, the highest CRE was only 79.8% for the SDS-CTAB mixture at the mixed ratio = 5:5, and the LOI of the tailings was 12.1%. Under the optimum mixed ratio, the promoting effect of binary surfactants was better than that of single surfactant Tween 80, SDS or CTAB [16].

On the whole, the synergistic effects of surfactant mixtures were ranked as: SDS-Tween 80 > Span 20-Tween 80 > CTAB-Tween 80 > SDS-CTAB. This result may be attributed to the HLB of surfactant mixtures and the interaction between surfactants and UC/PAC surface. Generally speaking, surfactants with high HLB values have more oxygen-containing functional groups, which is more beneficial to the removal of UC. Furthermore, the synergistic effect is the best when the surfactant mixtures have the optimum HLB. The results about the non-ionic Span 20-Tween 80 mixtures were in accordance with that reported for Huang et al. [23] who concluded that the optimum HLB value is 13.5. The synergistic effect of SDS-Tween 80 is like this situation. Furthermore, the hydrophobic part of Tween 80 and SDS was mainly adsorbed on the hydrophobic surface of carbon constituents [22]. No synergy of CTAB-Tween 80 was observed, probably because their HLB were very close. Tween 80 may interact with PAC and UC by the hydrophobic bond of the nonpolar hydrocarbon chain [13,15]. In addition, non-ionic Tween 80 can emulsify the kerosene and increase the number of oil droplets. Because there is no Tween 80, the synergistic effect of SDS-CTAB mixtures was relatively poor. The combination of anionic–cationic surfactants might inactivate and bring the negative effect due to interaction between opposite charges [19].

The flotation performance of fly ash was superior to the research of Huang et al. [23] when the Tween 80 and SDS mixtures were used [2]. This may be due to the different characteristics of two types of fly ash, such as particle size, the content of PAC and chlorides. In addition, the surfactant concentration and pulp pH also affected the flotation results [20].

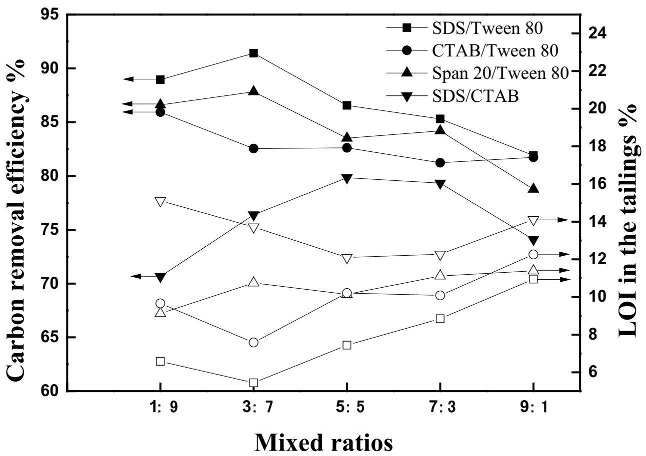


Fig. 1. Effects of mixed surfactants on decarburization performance.

3.2. Effect of mixed surfactants on surface tension and zeta potential of the slurry

An appropriate surfactant can improve the flotation performance by reducing the surface tension of the solution. The synergistic effect in the mixed surfactant system may be related to reducing the surface tension [20]. The effect of surfactant mixtures on the surface tension of the slurry is shown in Fig. 2. According to our previous results, when a single surfactant of Tween 80, CTAB and SDS were used, the surface tensions of slurry were 73.2, 74.9 and 75.3 mN/m, respectively. The surface tension of surfactant mixtures was lower than that of the calculated average value of the single surfactant, which might be due to the synergy effect of surfactant mixtures. Low surface tension is beneficial to improve the adhesion of bubbles, the dispersion of oil collectors and flotation recovery [15]. When SDS: Tween 80 ratio = 3:7, a sharp decrease of surface tension was observed, these binary surfactants may be more readily adsorbed on the UC surface, which improves the performance of decarburization flotation. The same situation was observed in Span 20-Tween 80 combinations. In addition, the surface tension of the slurry might relate to the high salt concentration due to the chloride dissolution in HSWI fly ash [26].

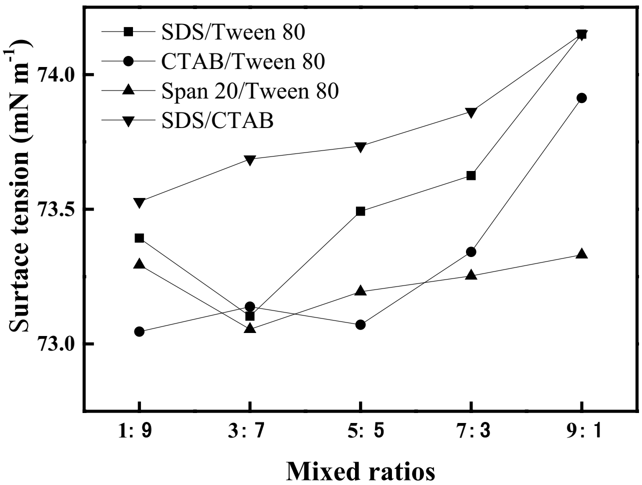


Fig. 2. Effect of surfactant mixtures on the surface tension of slurry.

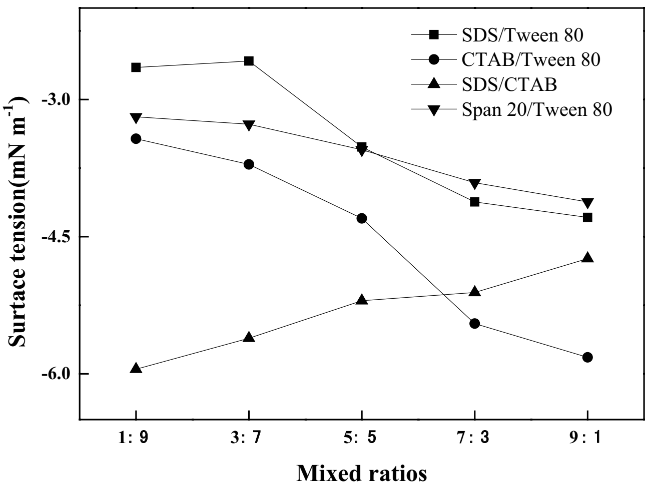


Fig. 3. Effect of surfactant mixtures on zeta potential of slurry.

In order to better understand the flotation behavior, the zeta potential of slurry in different surfactant mixtures was measured (Fig. 3). The zeta potential curve showed the opposite trend in the presence of CTAB-Tween 80 and SDS-CTAB, because the zeta potential increases with the increase of CTAB concentration, while the zeta potential of other surfactants decreases. When the mixed ratio changed, Span 20-Tween 80 mixture did not distinctly make the zeta potential decrease. These observations are consistent with the results of carbon removal. Generally, the removal efficiency reaches the maximum value below or above the isoelectric point, or the minimum absolute value of zeta potential [27,28]. The minimum value of zeta potential at mixed ratio = 3:7 could explain the high decarburization performance of the SDS-Tween 80 mixture.

3.3. Effect of mixed surfactants on PCDD/F removal

The removal mechanism of PCDD/Fs in HSWI fly ash may be divided into the three steps: (1) Dissolution: part of PCDD/Fs might deviate from the solid matrix or dissolve after adding surfactant mixtures; (2) Adsorption: PCDD/Fs in solution may be adsorbed in carbon constituents due to effective performance; (3) Flotation: PCDD/Fs were floated from solution into the froths together with carbon constituents. In this process, the carbon constituents were acted as carriers of PCDD/Fs [29]. In addition, there is close adsorption between gaseous-phase PCDD/Fs and porous PAC, and these low chlorinated PCDD/Fs combine strongly with PAC during the flotation process.

We have previously confirmed that PCDD/Fs removal is consistent with the carbon removal under the action of surfactant [16]. According to the above decarburization flotation, the best-mixed ratios for SDS-Tween 80, CTAB-Tween 80, Span 20-Tween 80 and SDS-CTAB mixture are 3:7, 3:7, 1:9 and 5:5, respectively. Under these conditions, PCDD/Fs removal effects and total toxic equivalent (TEQ) values in the tailings are shown in Fig. 4. When SDS-Tween 80, CTAB-Tween 80, Span 20-Tween 80 and SDS-CTAB mixture were

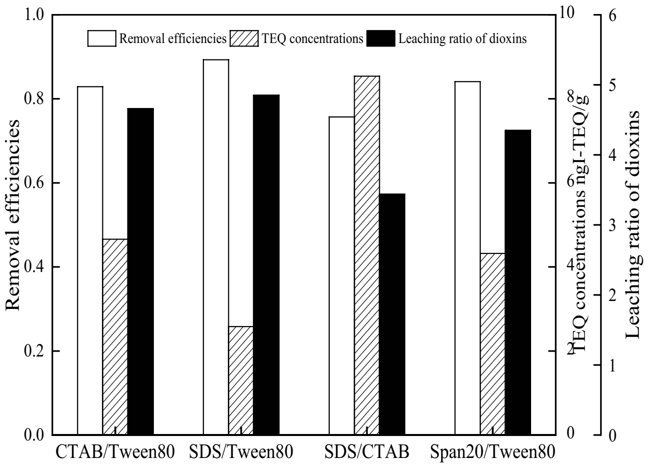


Fig. 4. Effects of surfactant mixtures on dioxin removal.

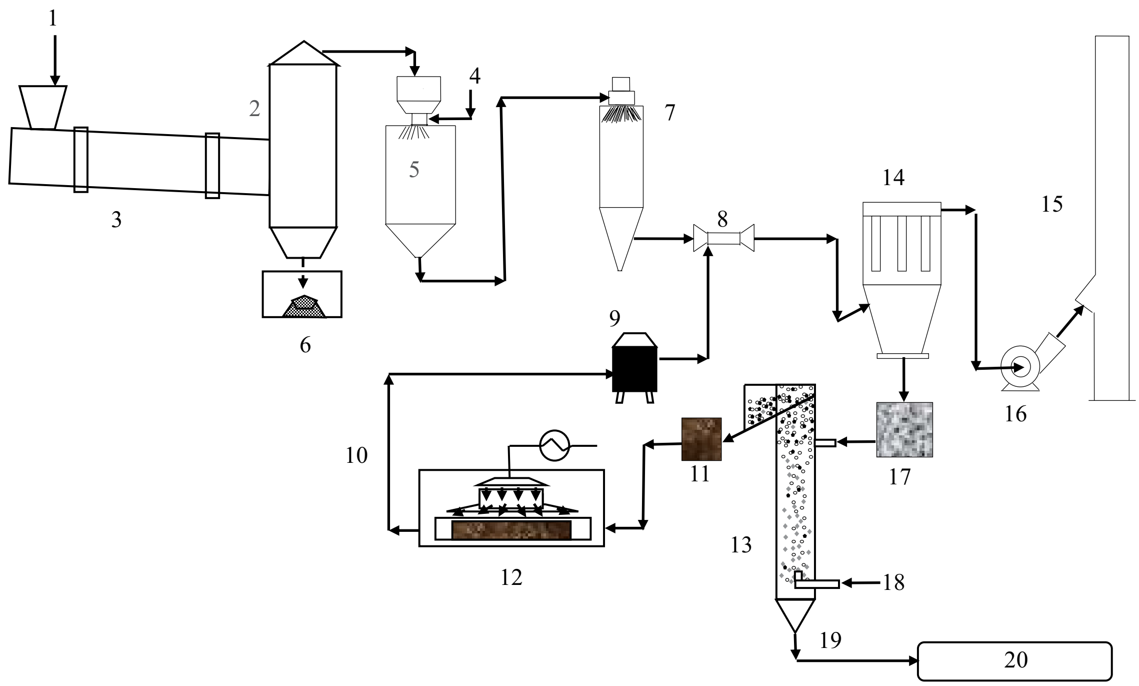
used, the total PCDD/Fs removal efficiencies were 89.3%, 82.9%, 84.1% and 75.7%, and TEQ values in the tailings were 2.58, 4.66, 4.32 and 8.54 ng I-TEQ/g, respectively. The results indicated that the removal efficiency of SDS-Tween 80 was higher than that of the other three binary surfactants and higher than that of single Tween 80 and SDS, respectively [16].

As mentioned above, for SDS-Tween 80 mixture, the surface hydrophobicity of the UC and PAC particles can be greatly improved due to the hydrogen bond of the anionic SDS and the hydrophobic bond of Tween 80 [25]. In addition, Tween 80 can shield the electrostatic head–head repulsion forces and increase the adsorption of anionic SDS [20]. Therefore, SDS-Tween 80 mixture could promote the adhesion of PCDD/Fs and improve the removal efficiency of PCDD/Fs. At the same time, after flotation with SDS-Tween 80, the TEQ value of the tailings is less than 3 ng I-TEQ/g which meets the standards of urban domestic waste landfills, and the tailings can be landfilled.

It is worth noting that PCDD/Fs in fly ash are easily leached out from in surfactant solution, especially in chloride solution [18,30]. After flotation, a small amount of PCDD/Fs would remain in the solution. Under the mixture of SDS-Tween 80, CTAB-Tween 80, Span 20-Tween 80 and SDS-CTAB mixtures, the residue rates of PCDD/Fs were 4.85%, 4.66%, 4.35% and 3.44%, respectively (Fig. 4). The results show that the residue rate of PCDD/Fs was high when SDS-Tween 80 was used as a surfactant. In addition, the removal of PCDD/Fs in the flotation is complicated, so it is necessary to further study the relationship between PCDD/Fs removal rate and waste residue removal rate. After flotation, the remaining solutions containing PCDD/Fs should be treated by photogradation or catalytic degradation to degrade PCDD/Fs before final disposal [31]. In addition, it is necessary to study the effect of surfactant concentration on the residual rate of PCDD/Fs.

3.4. Application prospect of flotation technology of HSWI fly ash

A large amount of HSWI fly ash is produced after burning HSW, and its safe disposal has attracted wide attention, especially during the epidemic in COVID-19. Flotation has been proved to be an effective treatment method for HSWI fly ash. Compared with single surfactants, SDS-Tween 80 mixtures can improve the floatability of fly ash under a certain compound ratio. Because PCDD/Fs is lipophilic and hydrophobic, most PCDD/Fs and carbon constituents will be separated and then enriched in the froths by flotation. The froth products after flotation account for about 19% of the original quality [16]. Thus, it is necessary to further treat the froths for reducing the PCDD/Fs emissions. According to our previous research on the froths, microwave treatment can not only destruct PCDD/Fs (above 98%) but also realize the regeneration of carbon constituents in the treated (Fig. 5) [32]. The treated froths can be injected again into air pollution control devices in the incinerator as an effective PCDD/Fs adsorbent. The volume and toxicity of the tailings would be decreased greatly, therefore, the tailings could be directly landfill disposed.



1—Medical waste; 2—Recombustion chamber; 3—Rotary kin; 4—Water; 5—Quench tower; 6—Bottom ash; 7—Semi-dry scrubber; 8—PAC injection; 9—PAC storage tank; 10—Pulverization; 11—The froths; 12—Microwave sintering furnace; 13—Flotation column; 14—Bag filter; 15—Stack; 16—Induced fan; 17—Raw fly ash; 18—Air; 19—Tailings; 20—Landfill disposal;

Fig. 5. Flow scheme of the combination of flotation and microwave treatment.

4. Conclusions

The effects of SDS-Tween 80, CTAB-Tween 80, Span 20-Tween 80 and SDS-CTAB on carbon and PCDD/Fs removal from HSWI fly ash during the flotation process were studied. The results show that the removal efficiency of binary surfactant is higher than that of single surfactant, especially when the ratio of SDS-Tween 80 mixture is 3:7. Under these conditions, the removal efficiencies of carbon constituents and PCDD/Fs are 91.4% and 89.3%, respectively. In addition, the synergistic effect of surfactants was characterized by measuring the surface tension and zeta potential. Floatation followed by microwave treatment may be an alternative method for the detoxification of HSWI fly ash.

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