Treatment of municipal sewage sludge by electron beam irradiation

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Abstract Irradiation treatment of sewage sludge was carried out using an electron beam accelerator. The effects of irradiation dose, sludge water content and sludge thickness on the breakdown of sludge flocs, thus the formulation of soluble chemical oxygen demand (SCOD) and soluble total nitrogen (STN), and the reduction of MLSS and endogenous OUR, and so on, were investigated. It was observed that, with increased doses, the SCOD, STN and UV absorption intensity of the sludge supernatant increased rapidly with similar trends, and MLSS considerably decreased, all indicating that a large amount of the cell contents of sewage sludge were released by electron beam irradiation. The results of endogenous OUR also showed that a large number of microorganisms in sewage sludge were killed or deactivated. Zeta potential sludge became more positive with increased doses, implying that the dewatering performance of sewage sludge was not adversely affected. It was also found that the thickness of sludge was an operation parameter of great importance in the electron beam treatment of sewage sludge due to the relatively short penetration depth in water/sludge of the high-energy electrons.

Key words Electron beam, Sewage sludge, SCOD, Endogenous OUR

1 Introduction

The treatment of sewage sludge has become one of the most critical environmental issues worldwide. In China, it is estimated that more than 4.2 million tons (dry weight) of waste sewage sludge is produced each year, at an annual growth rate of 10%. In addition to the heterotrophic and autotrophic bacteria which are responsible for wastewater purification, sewage sludge contains a large amount of pathogenic microorganisms and organic contaminants, and high concentration of heavy metals. Sewage sludge treatment contributes to 30%–50% of the total operation costs of a wastewater treatment plant, and a cost-effective method is a must.

Ionizing radiation, such as gamma ray and electron beam, is a powerful tool to treat wastewater^[1], and it is also a promising method for sludge treatment due to its high efficiency in reducing pathogens and decomposing non-biodegradable hazardous pollutants such as pesticides, PAHs and PCBs^[1,2]. Since 1960s, gamma ray sewage sludge treatment has been

extensively studied, and demonstration plants have been built and run for several years^[2-7]. In the past decade, electron beam treatment of sewage sludge has been explored^[8-11]. In this paper, E-beam treatment of sewage sludge of different water contents and thicknesses is studied.

2 Materials and methods

2.1 Materials

Sludge samples (~99% water and ~1% dry materials) were taken from the secondary settling tank in a municipal wastewater treatment plant in Jiading district, Shanghai, China. The water content was reduced to about 97% and 90% by gravity thickening and centrifugation, respectively.

2.2 Irradiation

Sewage sludge samples of ~99%,~97% and ~90% water content and in thickness of ~0.3, ~0.5 and ~1.0 cm were irradiated to 2, 5, 10, 15 and 20 kGy by 1.8 MeV 1mA E-beam from an electron accelerator at

Supported by Shanghai Natural Science Foundation (Grant No. 09JC1416900)

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ambient temperature.

2.3 Analytical methods

The sludge samples before and after irradiation were centrifuged (40 min, 4500 rpm). Their supernatant for soluble chemical oxygen demand (SCOD) was measured with a Hach DR/890 Colorimeter, and UV absorption spectra were obtained with a Hitachi UV-3010 spectrophotometer. The soluble total nitrogen (STN) and mixed liquor suspended solids (MLSS) were assayed according to standard methods^[12]. The zeta potential of particles in centrifuged (2 min, 1800 rpm) sludge supernatant was measured by a Concentrated Zeta Potential Analyzer (DelsaNano C, Beckman Coutler).

The specific oxygen uptake rate (SOUR) during the endogenous respiration phase, i.e., in the absence of substrate from external sources, of the sludge samples was measured using a closed respirometric unit of a BOD₅ bottle, in which the dissolved oxygen (DO) concentration was measured using a YSI Model 5100 oxygen analyzer. The non-irradiated sludge samples of different water contents (~99%, ~97% and ~90%), were diluted to the same MLSS (=2.0 g/L) by distilled water before the DO measurement. The same dilution factor was

applied to an irradiated sample of the same water content. The sludge sample in the BOD₅ bottle was continuously stirred, and the DO value was obtained every 30 second. The OUR was determined by linear regression from the slope in the DO concentrationtime plotting. For convenient comparison, the SOUR of samples before and after irradiation was calculated by the following equation:

SOUR (in $mgO_2 \cdot h^{-1} \cdot g^{-1}MLSS$) = OUR/(original MLSS before irradiation/dilution factor).

3 Results and discussion

3.1 Mixed liquor suspended solids

The irradiation dose effect on mixed liquor suspended solids in 0.5-cm thick sludge samples of different water contents, and in 97% water-content samples of different thicknesses, was shown in Table 1. The MLSS decreased with increasing doses and more MLSS loss was seen for samples of lower water content (higher biosolid concentration). The 0.5-cm sample thickness was the best in terms of the MLSS removal, as it is the closest to the optimum range of 1.8 MeV E-beam in water.

Dose / kGy	Water contents of 0.5-cm thick samples			Thickness of 97% water-content samples		
	99%	97%	90%	0.3 cm	0.5 cm	1.0 cm
0	12.9	32.2	98.8	32.2	32.2	32.2
2	11.4	28.3	93.8	30.4	28.3	31.3
5	10.4	26.7	92.7	27.2	26.7	28.9
10	9.3	25.8	91.1	26.1	25.8	27.6
15	8.2	26.4	89.4	27.2	26.4	28.2
20	8.0	24.7	88.7	25.2	24.7	25.7

Table 1 MLSS (in g/L) of sludge samples of different water contents and thicknesses irradiated to 0-20 kGy

The organic contents of the sludge flocs, such as non-biodegradable substances and especially various microorganisms, were broken up by free radicals such as $\cdot OH$, $\cdot e_{aq}$, $\cdot H$, $\cdot O_2^-$ and $\cdot HO_2$, which were generated from irradiation of water and further reactions, for example:

 $H_2O \rightarrow 2.7 \cdot OH + 0.6 \cdot H + 2.6 \cdot e_{ag}^{-} + 3.2 H^{+} + 0.7 H_2O_2 +$

$$0.45 H_2 + \cdots$$
 (1)

$$\mathbf{e}_{\mathrm{aq}} + \mathbf{O}_2 \longrightarrow \mathbf{O}_2 \tag{2}$$

$$\cdot H + O_2^- \rightarrow \cdot HO_2 \tag{3}$$

3.2 Soluble chemical oxygen demand

Figure 1a shows the SCOD in sludge samples of different water contents irradiated to different doses. The SCOD increased with the dose, being 685, 1605 and 2770 mg/L for sludge samples of 99%, 97% and

90% water contents, respectively. And it increased with decreasing initial water contents. This is because that more biosolid was involved in the reaction with free radicals generated in the irradiation. The significant increase of SCOD was also attributed mainly to the breakup of microbial cells which lead to the release of soluble intracellular substances, and the decomposing of insoluble intracellular substances might also increase SCOD, especially at higher irradiation doses. Fig.1b shows the SCOD in 97% water-content sludge samples of different thicknesses irradiated to different doses, and the sludge thickness of 0.5 cm was the best in this regard. Figs.1a and 1b also show that the efficiency of irradiation process tends to be lower when the doses were higher than approximately 5 kGy.



Fig.1 SCOD-dose curves of (a) 0.5-cm thick sludge samples of different water contents and (b) 97% water-content sludge samples of different thicknesses.

3.3 Soluble total nitrogen

Figure 2a shows the STN of 0.5-cm thick sludge samples of different water contents irradiated to 0–20

kGy. The STN, which was very low initially, increased rapidly with the dose, but decreased with increasing water contents. STN, including ammonia nitrogen, nitrite nitrogen, nitrate nitrogen, and soluble organic nitrogen, was mainly released by irradiation of proteins and nucleic acid of sludge microorganisms. As a result, more STN was released when the sludge concentration was higher. The STNs of 97% water-content sludge samples in different thicknesses are shown in Fig.2b. At 20 kGy, the STN concentration increased from 2.7 mg/L to 71.9, 85.4 and 65.7 mg/L for 0.3-, 0.5- and 1.0-cm sample thickness, respectively.



Fig.2 STN-dose curves of (a) 0.5-cm thick sludge samples of different water contents and (b) 97% water-content sludge samples of different thicknesses.

3.4 UV absorption spectra

Figure 3 is the UV absorption spectra of the supernatant of a sludge sample (97% water content, 0.5 cm thick) before and after irradiation. The peak near 260 nm is the absorbance peak of nucleic acids. The absorption peak of proteins is often near 280 nm due to the amino acids such as tryptophan, tyrosine

and cysteine. So the broad absorption band from 240 nm to 300 nm in Fig.3 should be overlapped spectra of nucleic acids and proteins generated from microorganisms in the Sludge. Effects of irradiation dose and sludge water content on the UV absorption intensity at 260 nm are shown in Fig.3b, with a 0.5-cm thick sludge sample. It looks very much like Fig.1a.



Fig.3 UV absorption spectra (a) of sludge samples (water content=97%, thickness=0.5cm) irradiated to different doses, and (b) the absorption at 260 nm *vs.* dose by 0.5-cm thick sludge samples of different water contents.

3.5 Endogenous SOUR

The endogenous SOUR of activated sludge is the oxygen consumption rate in absence of external substrate, so it is suitable for examining the concentration of active biomass. Fig.4 shows effects of the irradiation dose, sludge water content and thickness on the endogenous SOUR of sludge samples. It is noted in Fig.4a that at ≤ 10 kGy the endogenous SOUR decreased rapidly, indicating that a large amount of microorganisms were deactivated or killed by irradiation, whereas above 10 kGy the decrease of endogenous SOUR became much slower. And the sludge sample of 0.5 cm thickness had the lowest

SOUR of all sludge samples, leaving other parameters unchanged (Fig.4b), indicating that the highest energy efficiency can be achieved at this thickness.



Fig.4 Endogenous SOUR-dose curves of (a) 0.5-cm thick sludge samples of different water contents and (b) 97% water-content samples of different thicknesses.

3.6 Zeta potential

Effects of the irradiation dose and sludge thickness on Zeta potential of particles in centrifuged sludge supernatant are shown in Table 2. Sludge flocs carry negative charge due to the anionic functional groups, such as carboxylic, so the measured zeta potential was all negative. From an electrokinetic point of view, zeta potential becoming more positive towards zero is good for sludge settling and dewatering. In Table 2, it is noted that the zeta potential increased with the dose. Therefore, the dewatering property of irradiated sludge cannot be adversely affected.

Table 2Zeta potential (in mV) of centrifuged particle in 97%water-content sludge samples of different thicknesses irradiatedto various doses

Dose	Sample thickness					
(kGy)	0.3 cm	0.5 cm	1.0 cm			
0	-15.18	-15.18	-15.18			
2	-13.59	-14.28	-14.76			

5	-13.15	-14.72	-14.35	
10	-13.09	-14.37	-14.55	
15	-11.79	-14.39	-13.78	
20	-11.02	-13.14	-10.59	

4 Conclusion

Sewage sludge was treated by electron beam under various conditions. It was found that electron beam could effectively deactivate or kill the sludge microorganisms, and make them release proteins, nucleic acids and other organic substances, which results in the increasing of SCOD and STN concentrations in the supernatant and the decreasing of MLSS and endogenous OUR of the irradiated sludge. The SCOD, TN, and intensity of UV absorption spectra showed the similar trends with the variation of irradiation dose, sludge water content and thickness, respectively. The zeta potential measurement indicated that the dewatering performance was not adversely affected. The thickness of irradiated sludge was an important parameter since the penetration of electron beam is much shorter compared to gamma rays.

References

- 1 IAEA. IAEA-TECDOC-1598, Vienna, 2008.
- 2 Wang J L, Wang J Z. J Hazard Mater, 2007, 143: 2–7.
- 3 Borrely S I, Cruz A C, Del Mastro N L, *et al.* Prog Nucl Energ, 1998, **33:** 3–21.
- 4 Gautam S, Shah M R, Sabharwal S, *et al.* Water Environ Res, 2005, **77:** 472–479.
- 5 Etzel J E, Born G S, Stein J, *et al*. Am J Public Health N, 1969, **59:** 2067–2076.
- 6 Lessel T, Suess A. Radiat Phys Chem, 1984, 24: 3–16.
- 7 Chu L, Wang J, Wang B. Biochem Eng J, 2011, **54:** 34–39.
- 8 Zheng Z, Kazumi J, Waite T D. Radiat Phys Chem, 2001,
 61: 709–710.
- 9 Chmielewski A G, Zimek Z, Brylsandelewska T, *et al.* Radiat Phys Chem, 1995, **46:** 1071–1074.
- 10 Park W, Hwang M H, Kim T H, et al. Radiat Phys Chem, 2009, 78: 124–129.
- 11 Shin K S, Kang H. Appl Biochem Biotech, 2003, **109**: 227–239.
- 12 APHA. Standard Methods for the Examination of Water and Wastewater, 20th ed. Washington, DC, 1998.