Asian Journal of Chemistry; Vol. 26, No. 10 (2014), 3105-3108

ASIAN JOURNAL OF CHEMISTRY

http://dx.doi.org/10.14233/ajchem.2014.16940

Combination of Ozone and Ceramic Raschig Ring to Improve Efficiency of Treatment of Landfill Leachate

VAN HUU TAP[1,*], TRINH VAN TUYEN[2], NGUYEN HOAI CHAU[2] and DANG XUAN HIEN[3]

[1]Faculty of Environmental and Earth Science, Thai Nguyen University of Sciences, Thai Nguyen City, Vietnam
[2]Institute of Environmental Technology, Vietnam Academy of Science and Technology, 18 Hoang Quoc Viet Road, Hanoi, Vietnam
[3]School of Environmental Science and Technology, Ha Noi University of Science and Technology, 1 Dai Co Viet Street, Hanoi, Vietnam

*Corresponding author: Tel: +84 975 326936; E-mail: vanhuutap@gmail.com

Received: 27 December 2013; *Accepted*: 23 February 2014; *Published online*: 10 May 2014; AJC-15190

The combination with ozone/ceramic raschig ring (O_3/CRR), with three strips of surface area of the ceramic raschig ring is 356, 539 and 728 m^2/m^3) at a concentration of O_3 is 2.882 g Hr^{-1} studied to improve performance of landfill leachate treatment in Nam Son site, Soc Son, Hanoi city, Vietnam. Volume of landfill leachate used for each pilot lot is 1 L. The combination of ozone and the ceramic raschig ring for landfill leachate treatment has improved significantly treating performance. In experiment with the ceramic raschig ring, surface contacting area is 728 m^2/m^3, performance of treatment of colour, COD and TOC has increased comparison with experiment without the ceramic raschig ring is 8, 16 and 7 % respectively. The amount of residual O_3 after treating in the experiment available the ceramic raschig ring (728 m^2/m^3 of surface area) is also much lower than the experiment without the ceramic raschig ring. Average amount residual O_3 corresponding the experiment with and without the ceramic raschig ring is 0.632 and 1.639 g respectively after 100 min of treatment. At experimental conditions available the ceramic raschig ring (728 m^2/m^3 of surface area), the content of O_3 identified uses 4.812 Kg O_3/Kg COD.

Keywords: Ceramic Raschig Ring, COD, Landfill leachate, Ozonation, Total organic carbon.

INTRODUCTION

Disposal of solid waste by burial is quite common method to treat solid waste in most countries around the world. This method is simple and easy to do but produce large amounts of landfill leachate, complex components, high pollution concentrations that are difficult to treat. Especially, persistent organic substances, such as humic compounds, fulvic and humic acid and polycyclic compounds, *etc*., inorganic compounds such as ammonium and anions Cl^-, HCO_3^-, CO_3^{2-} and SO_4^{2-}. Therefore, the study of treatment methods of landfill leachate bringing high performance is an essential issue. Composition and properties of landfill leachate vary due to component of solid waste and landfill age. Landfill leachate released from the new landfill sites available a high concentration of organic substances mainly volatile acids. If the ratio of BOD/COD is high, it should be treated with biological methods that get a good effect.

In contrast, landfill leachate released from the old landfill sites contains mostly non-biodegradable organic compounds. The ratio of BOD/COD is low, thus treatment by biological methods get low efficiency. Chemical and physiochemical methods used to reduce the concentration of non-biodegradable organic matters and increase the ratio of BOD/COD.

Recently, researchers used ozone to treat non-biodegradable organic compounds of landfill leachate due to its high oxidation potential[1-4]. Ozone was used to treat COD and colour of landfill leachate. With ozone concentrations of 400 mg/L/h, 50 % COD and 90 % colour at pH 11 in 180 min of reaction are removed[5]. use of 1.3 g O_3/g COD, 30 % COD and 21 % TOC of landfill leachate are treated. Ozone is often used to improve the efficiency of landfill leachate treatment. The ozonazion process is used in the pre-treated phase of landfill leachate[6-8]. However, there are many studies on treatment of landfill leachate by ozone agent to enhance processing efficiency after processes of chemical, physics and biological treatment[1,9,10] combined $O_3/H_2O_2/Fe^{2+}$ to improve the efficiency of landfill leachate treatment in Malaysia. The efficiency of treatment gains 87 % COD and 100 % colour.

In this study, a new agent is put in to enhance the solubility of O_3 from the gas blend in to water (landfill leachate) to enhance treated efficiency of organic compounds in landfill leachate in Vietnam. This is the combination of ozone and ceramic raschig ring (CRR): (O_3/CRR). The main objective of this study is to improve efficiency of landfill leachate treatment with ozone by combining ozone and ceramic raschig ring (O_3/CRR). Ceramic rasching rings are used to improve the mass transfer of O_3 from gas phase to liquid phase.

EXPERIMENTAL

Sampling landfill leachate: Landfill leachate untreated is taken from the reservoir in Nam Son landfill, Soc Son district, Hanoi city, Vietnam. The area of landfill is 54.07 hectares, divided into 9 buried boxes and started burying operation from 1999. The landfill receives daily 4,000 tons of domestic solid waste from Ha Noi city. Landfill leachate is released 1,600 m^3 per day[11]. Landfill leachate taken was collected in 20 L plastic cans in the period from July to October in 2013 and transported to the laboratory. Characteristics of landfill leachate are shown in Table-1. Indicators were analyzed according to the Standard Methods for the Examination of Water and Wastewater[12].

TABLE-1
CHARACTERISTICS OF QUALITY OF LANDFILL LEACHATE FROM NAM SON IN THIS RESEARCH

Parameters	Unit	Value	Average
pH	-	7.87-8.52	8.20
Colour	Pt-Co	2022-3050	2536
COD	mg L^{-1}	2178-3987	3083
BOD	mg L^{-1}	572-1452	1012
TOC	mg L^{-1}	955-1010	982
Total N	mg L^{-1}	724-1806	1265
NH_4^+	mg L^{-1}	670-1668	1169
Total P	mg L^{-1}	4.28-6.30	5.29
Cl^-	mg L^{-1}	1724-2469	2097
CO_3^{2-}	mg L^{-1}	181-234	208
HCO_3^-	mg L^{-1}	7557-10389	9873
SO_4^{2-}	mg L^{-1}	99-282	191

Analytical methods: COD, colour, pH and O_3 were measured before and after treatment according to the Standard Methods for the Examination of Water and Wastewater[12]. TOC was measured on TOC analyzer: TOC-Vcph (TCVN 6634:2000).

Experiment systems: The system consists of two reactors: one reactor without ceramic raschig ring (CRR) and other reactor containing ceramic raschig ring (Fig. 1a and 1b). Corresponding landfill leachate is treated by all system available reactor with or without ceramic raschig.

The experimental apparatus consists of: (1) tank containing landfill leachate (2) landfill leachate pump (3) the reactor, (4) ozone generator, (5) the ozone absorption device (6) ceramic raschig ring.

Ozone generator: Model: Lino (lin 4.10 L) capacity of 10 g per hour, manufactured by Lino J.S.C in Vietnam. Equipment fixed automated clocks and meter to measure input gas flow (1-20 l per minutes). Gas from the ozone generator for reactor from the bottom of the reactor center.

The reactor: the reactor is made of cylindrical heat-resistant mica, 1,000 mm height, 54 mm internal diameter and 3 mm thickness. On reactor's wall, there are valves fixed to take water samples. At reactor's bottom, the gas supply valve mounted on the valve and water discharging water samples after treating. On reactor's top, there is the gassing valve and simultaneously it can connect to ozone absorbing device.

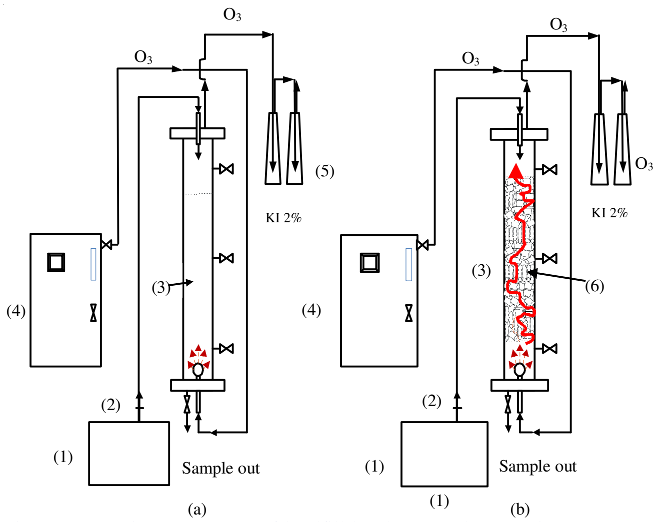

Fig 1. Experiment systems of landfill leachate (a) the experiment system with reactor uncontaining ceramic raschig ring, (b) the experiment system with reactor containing ceramic raschig ring

Ceramic raschig ring: Ceramic raschig rings have mechanical strength and surface area that is many times higher than other conventional materials are. It was made by Tropical Environment Company in Viet Nam. Chemical composition mainly are SiO_2, Al_2O_3, ceramic raschig rings have the ability to withstand high temperatures up to 800 °C and to be sustainable in high acidic or base medium. Chemical composition: > 60 % SiO_2, 17-23 % Al_2O_3, <1 % Fe_2O_3, 1-2 % CaO_2, 2-4 % K_2O + Na_2O. Features of ceramic raschig are described in Table-2 and Fig. 2.

Use of O_3 process/ceramic raschig for treating landfill leachate: The experiments in combining O_3/CRR were made at pH 8 and reaction time of 100 min. Experiments' aim is to assess the influence of the surface area of ceramic raschig and O_3 to capacity of treatment of landfill leachate. The experiment is performed with values of contact area of ceramic raschig's surfaces of 0; 356; 539 and 728 m^2/m^3. Volume of landfill leachate and airflow of ozone generator are unchanged when comparing with the experiment without ceramic raschig ring.

When taking O_3 to reactor available ceramic raschig ring that will increase the contact time of O_3 and wastewater (at red line in Fig. 1b). This means that the increase of O_3 solubility in water will increase the reaction of O_3 with organic substances in landfill leachate. Ceramic raschig ring is used in this experiment has a smooth surface, non-corrosion, non-reaction with O_3. Use of ceramic raschig ring with three sizes (Table-2) for three different kinds to evaluate treating performance. The experiments were performed at 25 ± 5 °C.

TABLE-2
FEATURES OF CERAMIC RASCHIG RING

Internal diameter (mm)	Dimensions (D*H*mm)	Surface area (m^2/m^3)	Porosity (%)	Quantity (rings/m^3)	Weight (Kg/m^3)
2	2*10*3	728	44	2,880,000	1,215
4	4*12*2.5	539	54	910,000	1,156
6	6*10*3	356	59	1,042	931

Note: D: Internal diameter (mm), H: Hight (mm), mm: thickness of wall of ring (mm), features of ceramic raschig ring was measured by esearcher

(a): Ceramic raschig rings (728 m^2/m^3) (b): Ceramic raschig rings (539 m^2/m^3) (c): Ceramic raschig rings (356 m^2/m^3)

Fig. 2. Three kinds of ceramic raschid rings with surface area

RESULTS AND DISCUSSION

In this study, before treatment by O_3/CRR, landfill leachate is pre-treated by coagulation with PAC 1,500 mg L^{-1}. Experiments of combination O_3 and ceramic raschig ring also conducted as the pH 8 and reaction time of 100 min. In these experiment of effect of surface area of ceramic raschig ring, the conditions are the same with above experiments: airflow is generated from Ozone Generator (7 L/min corresponding ozone concentrations generated about 2,882 mg Hr^{-1}), volume of test sample (1 L), landfill leachate also pretreated by flocculation-coagulation with PAC 1500 mg L^{-1}. The treatment efficiency of landfill leachate is compared with concentrations after flocculation. The experiments were carried out simultaneously on the same concentrations on colour, COD and TOC of landfill leachate. Concentrations are as follows: COD: 2,142-2,333 mg L^{-1}, colour: 1,650-1,808 Pt-Co, TOC: 825-890 mg L^{-1}. The experiments were performed three times, denoted by E1, E2 and E3 with following conditions: reactor without ceramic raschig and 3 reactors with ceramic raschig ring available 356, 539 and 728 m^2/m^3 of the surface area, respectively. Experimental results are shown in the Fig. 3.

The experimental results of effects of the surface area of ceramic raschig ring with the conditions as above original pollutants have showed that the treatment efficiency has not changed much between experiments with the reactor systems without ceramic raschig ring and the reactor systems with ceramic raschig available up to 356 m^2/m^3 of surface area. There was COD removal (the reactor without ceramic raschig ring: 30-33 %, the reactor with ceramic raschig ring 356 m^2/m^3 : 32 %), TOC removal (the reactor without ceramic raschig: 25-33 %, the reactor with ceramic raschig ring 356 m^2/m^3 : 27-35 %) and colour removal (the reactor without ceramic raschig ring: 66-74 %, the reactor with ceramic raschig ring 356 m^2/m^3 : 67-79 %). However, when the surface area increased over 539 m^2/m^3, the treatment efficiency of organic compounds in landfill leachate in this study has increased significantly. Especially, treatment efficiency of COD and TOC achieved the highest performance of experiments on the reactor with

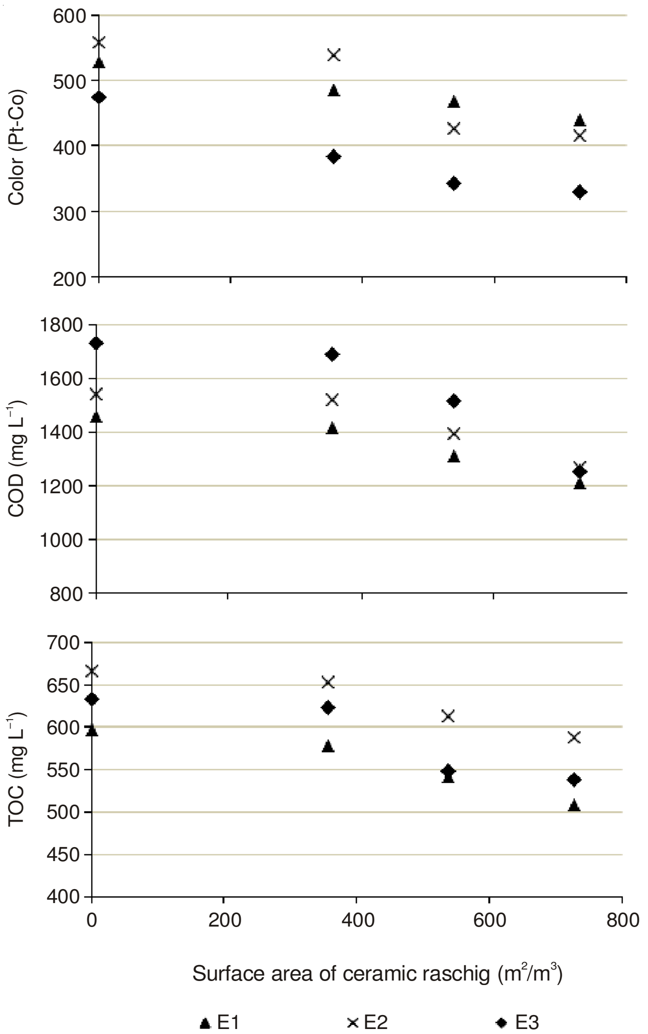

Fig. 3. Effect of surface area of ceramic raschig on Colour, COD and TOC removal in treatment by O_3/ceramic system at pH 8 after 100 min of treatment

ceramic raschig ring available up to 728 m^2/m^3 of surface area. The treating performances for colour, COD and TOC corresponding the highest achievement are 74- 82, 42-49 and 34-

TABLE-3
RESIDUAL O_3 CONCENTRATION AFTER EXPERIMENTS IN COMBINATION OF O_3 AND CERAMIC RASCHIG

Surface aea of ceramic raschig ring (m^2/m^3)	Experiment					
	E 1		E 2		E 3	
	Amount of O_3 in (g)	Amount of O_3 out (g)	Amount of O_3 in (g)	Amount of O_3 out (g)	Amount of O_3 in (g)	Amount of O_3 out (g)
0	4.628	2.503	4.413	2.479	5.368	2.789
356	4.628	2.408	4.413	2.360	5.368	2.670
539	4.628	2.193	4.413	2.097	5.368	2.455
728	4.628	1.549	4.413	1.501	5.368	1.763

42 % in the experimental systems without ceramic raschig ring in the same conditions of the initial pollutants, performance of the above indicators is 66-74 %, 30-33 and 25-33 %, respectively.

The experimental results indicate that, O_3 is an agent poorly soluble in water from aeration. It has expressed through treatment efficiency not really high in reactor with ceramic raschig ring up to 356 m^2/m^3. It is not so different from the experiments of the reactor without ceramic raschig ring. It is only to use the reactor containing ceramic raschigring with up to 539 m^2/m^3 of surface area, performances increase significantly. In the experiment with the reactor available ceramic raschig ring with 728 m^2/m^3 surface area, the treating performances for colour, COD and TOC are higher than those experiments with reactor unavailable ceramic raschig ring corresponding are 8 %, 12-16 and 7 % and achieved the highest performance in the experiment of this study. As a result, the larger surface area has increased mass transfer of the gas phase to the liquid phase. The cause can explain: when the large surface area of ceramic raschig, contact time of the gas phase and liquid phase will increase that increase O_3 transition from the gas phase to the liquid phase. Therefore, the amount of O_3 in water increases, the reaction of O_3 with organic compounds in landfill leachate also increases. This result is substantiated *via* the results table of analysis of the amount of O_3 before and after the reaction of experiments of combination of O_3 and ceramic raschig ring (Table-3).

Analysis results of amount of O_3 before and after reaction have showed that the amount of residual O_3 in experiments with the reactor available ceramic raschig ring is lower than that of the reactor without ceramic raschig. At experiments with ceramic raschig available 728 m^2/m^3 of surface area, the amount of residual O_3 measured is the lowest concentration in all three experiments. This is to prove that the amount of O_3 dissolved in water is the greatest. At this condition, after 100 min of reaction, average amount of O_3 generated will provided 4.803 g. The average reduction of COD is 997.8 mg in 1 L of water, amount of O_3 consumed in average to treatment for 1 kg COD was 4.8 Kg O_3/Kg COD. Some previous studies have shown the level of consumption of O_3 to treat COD landfill leachate such as[2,13] consumes 1.5 and 16 KgO_3/Kg COD when using in combination to O_3/H_2O_2 to treat landfill leachate, while it is shown that less consumption is 1 KgO_3/Kg COD showed 1.6 KgO_3/Kg COD being results of consumption when using in combination to O_3/Fenton to treat landfill leachate.

Conclusion

In this study, the experimental results have identified that experiments of combination O_3/ceramic raschig ring have significantly increases in the treating performance of organic compounds in landfill leachate of experiment available 728 m^2/m^3 surface area. The treatment efficiency of colour, COD and TOC is over 10 % higher than that the experimental system without ceramic raschig ring. O_3 solubility from aeration to the liquid phase of experiments with 728 m^2/m^3 of surface is also much higher than that of the experiments without ceramic raschig ring. O_3 solubility from air phase to the liquid phase of experiments with 728 m^2/m^3 of surface area is also much higher than that of the experiments without ceramic raschig ring. The combination O_3/ceramic raschig is a treating solution with a good effect in landfill leachate treatment. Experiments also showed that it has to use 4.8 Kg O_3 to treat 1 Kg COD.

REFERENCES

1. S.S. Abu Amr, H.A. Aziz, M.N. Adlan and M.J. Bashir, *J. Environ. Sci. Health A*, **48**, 720 (2013).
2. C. Tizaoui, L. Bouselmi, L. Mansouri and A. Ghrabi, *J. Hazard. Mater.*, **140**, 316 (2007).
3. D.M. Bila, A. Filipe Montalvão, A.C. Silva and M. Dezotti, *J. Hazard. Mater.*, **117**, 235 (2005).
4. C. Ratanatamskul and P. Auesuntrachun, *Int. J. Environ. Waste Manage.*, **4**, 470 (2009) .
5. P. Van Aken, N. Lambert, J. Degrève, S. Liers and J. Luyten, *Ozone Sci. Eng.*, **33**, 294 (2011).
6. S. Cortez, P. Teixeira, R. Oliveira and M. Mota, *J. Hazard. Mater.*, **182**, 730 (2010).
7. P. Haapea, S. Korhonen and T. Tuhkanen, *Ozone Sci. Eng.*, **24**, 369 (2002).
8. I. Kamenev, L. Pikkov and J. Kallas, *Proc. Estonian Acad. Scientific Chem.*, **51**, 148 (2002).
9. A. Vilar, S. Gil, M.A. Aparicio, C. Kennes and M.C. Veiga, *Water Practice Technol*, **1**, 3 (2006).
10. C. Di Iaconi, R. Ramadori and A. Lopez, *Biochem. Eng. J.*, **31**, 118 (2006).
11. N.H. Khanh, Landfill Environment and Technology of Landfill Leachate Treatment, Publish House of Technological Science, Vietnam (2009).
12. APHA, Standard Methods for the Examination of Water and Wastewater, 19th American Public Health Association, Washington DC (2012).
13. F. Wang, D.W. Smith and M.G. El-Din, Oxidation of Aged Raw Landfill Leachate with O_3 Only and O_3/H_2O_2 and Molecular Size Distribution Analysis, Proceedings of the 16th World Congress of the International Ozone Association, IOA, Las Vegas, USA, pp. 1-21 (2003).