The Effect of γ-Irradiation on the Biodegradability of Landfill Leachate

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Synopsis. A preliminary examination of the enhancement of biodegradability for landfill leachate, humic acid, and fulvic acid solutions, and aqueous solutions of polyethylene glycol-6000 by irradiation by ^{60}Co γ-rays under aeration was performed. In all cases, the BOD_5/TOC and BOD_5/COD ratios increased with the dose. This suggests that the γ-irradiation modified these refractory materials to more biodegradable compounds.

The object of the treatment of landfill leachate is to remove organic substances and nitrogen contained in large amounts in the leachate. It is not easy to remove the organic substances by microbial- and physical-chemical treatment processes, because most of these substances consist of high-molecular-weight fulvic-like and humic-like materials.[1] In the determinations of the biodegradability for many kinds of waste water, Murakami *et al.* recently showed that landfill leachate was an especially refractory waste and was hardly treated by a present biological process.[2]

There has been much interest in the possible application of ionizing radiation to the treatment of waste water. The principal effects that can be turned to advantage are related to disinfection, the degradation of organic compounds, modifications in the biodegradability, and changes in the colloidal properties.[3]

The authors have examined the radiation-induced degradation of organic substances in the leachate generated from the landfill in Tokyo Bay by the irradiation of ^{60}Co γ-rays under aeration at room temperature. It was found that these substances were degradated by oxidation reaction, leading to a decrease in the chemical oxygen demand(COD) and the total organic carbon(TOC), and the conversion to lower-molecular-weight compounds, while oxygen-containing compounds, such as organic acids and carbon dioxide, were also produced.[4]

These results suggest that the irradiation is capable of enhancing the biodegradability for the leachate, since the biodegradability is dependent upon the chemical composition[5] and the molecular-weight distribution.[6] In some experiments carried out by Alexander, the irradiation produced a slight decrease in the biodegradability of the primary effluent.[7] Thus, preliminary experiments for the modification of the biodegradability of the landfill leachate by irradiation by ionizing radiation were undertaken.

Experimental

The leachate (COD, about 2000 ppm), solutions of humic acid and fulvic acid separated from the leachate, and dilute aqueous solutions of polyethylene glycol-6000** were used as the samples. These solutions were put into a glass vessel about 30 mm in diameter and irradiated under aeration with γ-rays from a 185 TBq (5 kCi) ^{60}Co source at room temperature. Almost all the irradiations were performed at the dose rate of 95 Gy·s^{-1} (0.57 Mrad·h^{-1}), as determined by means of a ferrous sulfate solution. After irradiation, the COD (permanganate method at 100 °C), TOC (Toshiba-Beckman Type Model 102 Total Organic Carbon Analyzer), and biochemical oxygen demand(BOD_5) were measured. For the determination of BOD_5, the return sludge from a sewage-treatment process was cultured with skim milk, and the filtrate was used for seeding.

**The mean molecular weight is about 6000.

Results and Discussion

There are several methods for determining the biodegradability, but a general method has not yet been established. It is known that, simply, the biodegradability can be estimated from the ratio of the BOD to the theoretical oxygen demand(ThOD) and that it becomes higher with an increase in the ratio. However, the ThOD can not be estimated for such complicated components as the leachate and the samples denatured by irradiation. In such cases, the TOC or COD can be used instead of the ThOD.[8]

The BOD_5, COD, TOC, and pH values in the leachate and in solutions of humic acid, fulvic acid, and polyethylene glycol-6000 are summarized in Table 1 as a function of the dose. As can be seen in Table 1, the COD and TOC in the leachate, humic acid, and fulvic acid decreased monotonously; on the contrary, the BOD_5 increased with an increase in the dose except for the BOD_5 in humic acid at 86 kGy (8.6 Mrad). Then, the BOD_5/COD and BOD_5/TOC ratios increased with the dose from almost zero in unirradiated solutions. The BOD_5/TOC ratios in these solutions are shown in Fig. 1 as a function of

Table 1. Changes in COD, TOC, and BOD_5 by γ-irradiation

Sample	Dose 10^4Gy(Mrad)	pH	COD	TOC	BOD_5
			ppm		
Leachate	0	8.6	375	262	~6
	1.71	8.5	292	222	81
	3.36		223	200	100
	5.34[a]	8.5	169	166	102
Humic acid	0	10.6	607	540	30
	0.86	7.2	598	516	61
	1.71	6.7	529	480	76
	3.42	4.5	390	352	104
	8.55	3.6	182	260	57
Fulvic acid	0	7.2	370	360	20
	1.80	6.0	309	318	101
	3.42	4.9	239	265	122
PEG-6000	0	5.8	403	532	~3
	0.57	3.7	400	530	97
	1.71	3.3	409	520	181
	3.42	3.2	395	528	332
	9.69		339	300	273

Dose rate: 95 Gy·s^{-1} (0.57 M rad·h^{-1})

a) 148 Gy·s^{-1} (0.89 M rad·h^{-1})

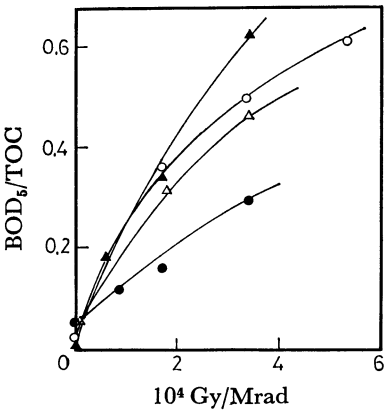

Fig. 1. Enhancement of BOD_5/TOC as a function of dose.
○: Leachate, ●: humic acid, △; fulvic acid, ▲: PEG-6000.

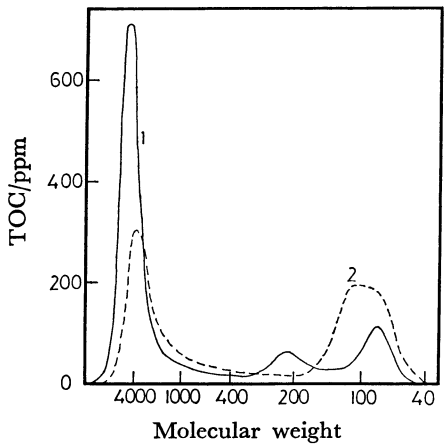

Fig. 2. Effect of irradiation on molecular weight distribution of humic acid solution.
1: Unirradiation, 2: 20 kGy (2.0 M rad).

the dose. The pH lowered with dose except in the leachate. It is because of the pH value's dependence on the buffer-capacity that there was little change in the pH of the leachate. These facts indicate that the irradiation modified the properties of those organic substances, so that they became more biodegradable compounds.

The same experiments were undertaken for the aqueous solutions of polyethylene glycol-6000, which is a refractory material for a biological-treatment process.[6] In this case, no marked change was seen in the COD and TOC up to at least 34 kGy (3.4 Mrad), while the BOD_5 increased markedly with the dose. Therefore, the ratios of BOD_5 to COD and of BOD_5 to TOC increased with the dose; moreover, these values were higher than those of the leachate (Fig. 1). The polyethylene glycol molecule seems to be easily modified to more biodegradable compounds.

The one reason for the enhancement of the biodegradability of these refractory materials by γ-irradiation may be the formation of lower-molecular-weight substances and of organic acids, such as acetic acid.[4] A typical finding regarding the molecular-weight distribution by gel-permeation chromatography of the humic acid solutions is shown in Fig. 2. From Fig. 2, one can find that the higher-molecular-weight components were degradated to lower ones by irradiation. The drops in the BOD_5/TOC and BOD_5/COD ratios of the humic acid solution at the dose of 86 kGy (8.6 Mrad) may be dependent upon the formation of less biodegradable materials by the high-dose irradiation.

These results show that the irradiation of ionizing radiation might increase the efficiency of the biological treatment process; however, more detailed experiments will be required.

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