

THE EFFECT OF INSIDE CIRCUMSTANCE OF THE HAZARDOUS WASTE LANDFILL ON THE LEACHING BEHAVIOR OF HARMFUL HEAVY METALS

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ABSTRACT: In Japan, among three landfill classes, the hazardous waste landfill is used for disposing of hazardous waste of which leaching of heavy metal exceeds its criteria. To prevent the emission of a hazardous substance to the surrounding environment, the hazardous waste landfill is required to be constructed with a robust concrete wall and base, and the top must be capped. Because of the containment structure, rainwater infiltration and air intrusion hardly occur. This means the leaching potential of hazardous heavy metals in waste remains forever. But considering long term safety, their leaching potentials must be better reduced. In this study, the influence of interior condition in the hazardous waste landfill on the leaching of heavy metals was examined from the accelerated weathering experiments for dust which is disposed of at the hazardous waste landfill. As the results, a decrease in the leaching amount of Cd, Pb, Mn, and Zn was confirmed in a wet environment where adsorbed water was present on the particle surface. On the other hand, in the dry and CO₂ saturated environment, an increase in the leaching amount was observed. The former phenomenon was thought to be the formation of hydroxide at the particle surface, and the latter was considered to be the influence of adsorption of CO₂ to Fe₃O₄ and ZnO. Though further scientific verification is required for each phenomenon, the findings obtained in this study indicate that the leaching of heavy metals from wastes in the hazardous waste landfill may change over time depending on the interior condition.

Keywords: Hazardous waste landfill, Dust, Weathering, Heavy metals, Leaching

1. INTRODUCTION

In Japan, solid waste landfill is classified into three types. They are inert waste landfill, non-hazardous waste landfill, and hazardous waste landfill [1]. Among them, in hazardous waste landfill (strictly controlled type landfill site), hazardous waste which exceeds criteria for hazardous heavy metals leaching is disposed of. The hazardous waste landfill is required to be designed and constructed with robust concrete wall and base, and the top must be capped (Required structure is indicated in Fig. 1). The concept of this landfill is containment of hazardous substance and eliminates the chance of reaction with water and air. In general, in non-hazardous waste landfill, various substances are subjected to many reactions and gradually stabilized. Even for hazardous heavy metals, it is known that their leaching potentials reduce due to the long-term weathering effect in non-hazardous waste landfill. But according to the concept of the hazardous waste landfill, these reactions cannot be expected because air and water intrusion cannot be expected. This means that the leaching potential of hazardous heavy metals will be kept forever. However, there is no guarantee that the container structure will be permanently

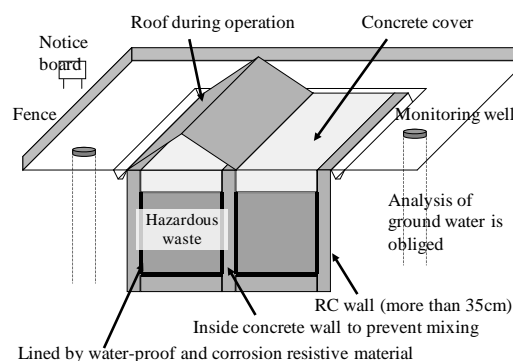


Fig.1 Required structure of hazardous waste landfill (strictly controlled type landfill site) in Japan (drawn based on [2])

maintained because of the impact of natural deterioration of structure or disaster.

In order to prevent sudden emission of hazardous substance due to these occasions in future, some measure must be taken. There are two candidates considered to be able to achieve it. The one is multi-barrier system. To surround the site by various barrier systems such as clay barriers, impact of heavy metal to the surrounding environment can be mitigated. The other is to reduce the leaching potential of hazardous

substance itself from solid waste. For incineration residue, the aging phenomenon is known to be effective for reducing the mobility of heavy metals [3]-[5]. The aging or weathering of the incineration residue is occurred by the contact with rainwater and carbon dioxide in air. As mentioned above, the hazardous waste landfill is surrounded by concrete walls and roof. The waste inside the landfill is isolated from the outside environment. This means that active control of inside circumstance is necessary to create the condition preferable for aging or weathering.

However, it is not clear what kind of interior condition management (humidity, gas atmosphere, temperature, etc.) is desirable for reducing of heavy metal leaching potential by aging/weathering. Storage of hazardous waste in the containment-type hazardous waste landfill such as the hazardous waste landfill in Japan) is semi-permanent. That is, an extremely long time can be used. There is room for study whether there can be favorable inside circumstance that can gradually change the heavy metals contained in the hazardous waste into a hardly soluble state within that long time. Controllable parameters may be gas atmosphere, humidity, temperature, etc. These need to be made clear.

In this study, the effect of inside circumstance of the hazardous waste landfill on the leaching behavior of harmful heavy metals was investigated. The electric furnace dust that is designated as hazardous waste was used and it was subjected to the accelerated weathering experiment. The objective of the study is to elucidate suitable condition for reducing heavy metal leaching potential from the waste disposed of in the hazardous waste landfill and to propose the active control method to achieve it.

2. MATERIAL AND METHODS

2.1 Material

According to the statistics of Ministry of Environment, Japan, there are 24 hazardous waste landfills registered [6]. However, among them, the landfill in operation is only one or two. At one hazardous waste landfill, interview was performed regarding kinds of waste accepted at the site. The major waste accepted and disposed of were sludge, cinder, and dust as indicated in Fig 2. Each waste was disposed of at this hazardous waste landfill site because leaching of certain heavy metal indicated in Fig 2 exceeds acceptance criteria designated by government.

However, it was difficult to obtain these waste samples at the landfill site due to their contracts. Thus, similar hazardous waste samples were

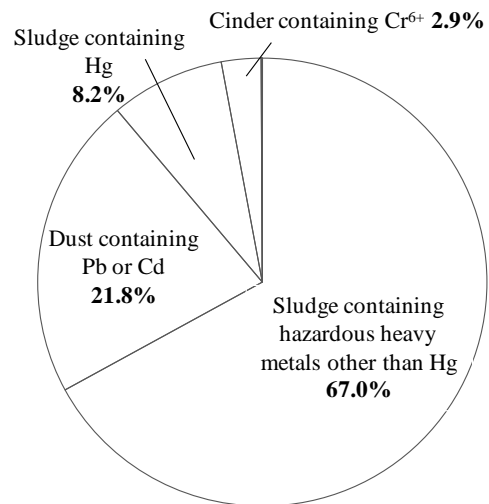


Fig. 2 Percentage of type of waste disposed of at one hazardous waste landfill (2014-2017)

obtained from waste treatment company which generates these wastes that have similar hazardous characteristics. The waste samples were sludge and two dusts. At first, leaching test (Japanese leaching test, JLT-13 [7]) was conducted for each of them. Since one dust sample from electric furnace had high heavy metal leaching potential, it was selected for the experiment in this study.

For the electric furnace dust, elemental composition was analyzed by XRF (Horiba MESA-800) at first, then, mineral composition was analyzed by XRD (Rigaku Rint-2000). Table 1 shows the elemental composition of the dust and Fig.1 shows the diffractogram by XRD analysis.

Table 1 Elemental composition of the electric furnace dust analyzed by XRF

Element	Fe	Zn	Cr	Ca	Ni
%	32.6	21.0	12.0	8.6	5.8

Element	Si	Cl	K	Mn	Pb
%	5.3	3.8	3.7	3.0	2.7

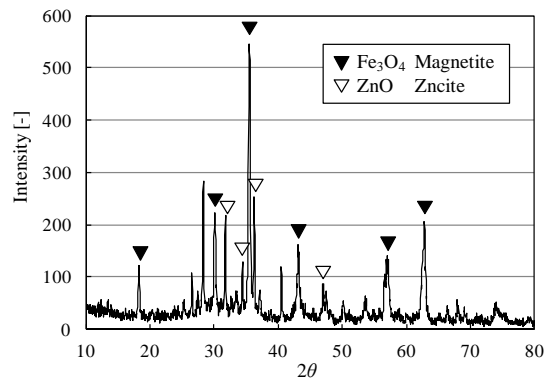


Fig. 3 Diffractogram of the dust sample by XRD

Table 2 Content and Leaching amount of metals

Element	Ca	Cd	Cr	Fe	Mn	Ni	Pb	Si	Zn
Content [mg/kg]	64139	117	23045	176074	12866	29301	10210	29903	84528
Leaching [mg/kg]	63.5	50.5	0.1	0.1	110.2	47.8	6.8	49.7	1109.5
Ratio [%]	0.1	43.3	0.0	0.0	0.9	0.2	0.1	0.2	1.3

Ratio = Leaching amount / Content

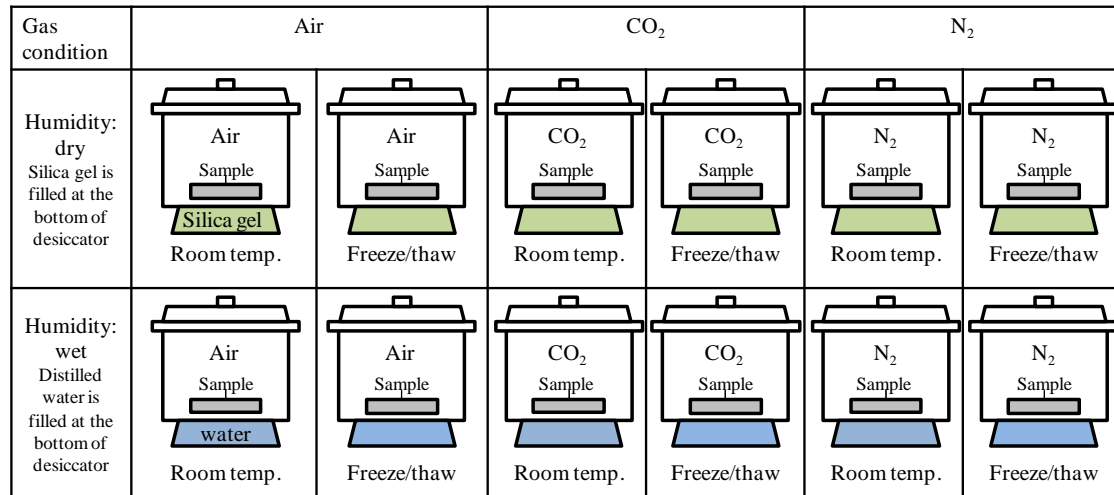


Fig. 4 Series of accelerating weathering experiment

As can be seen Table 1, major component of the dust is Fe and Zn. Besides, from Fig. 3, Fe is contained as Magnetite (Fe₂O₃) and Zn is contained as Zincite (ZnO). Since both compounds have adsorption capability, they may affect heavy metal leaching characteristics from the dust.

Next, for determining of the more accurate content, the contents of Ca, Cd, Cr, Fe, Mn, Ni, Si, Pb, and Zn, which were the main components of the dust by XRF analysis, were measured by ICP analysis (SHIMADZU ICPE-9000) after hydrofluoric acid digestion of the dust. Also, the leaching amount was measured again by the JLT-13 leaching test of liquid-solid ratio 10. ICP again used for the analysis of eluate of the leaching test.

Table 2 shows the measurement results of the metal content and the leaching amount. Although the leaching ratio (= leaching amount / content) of main elements such as Fe and Ca is low and it seems to exist in a hardly soluble form, about 43% of the content of cadmium was leached out.

2.2 Accelerated Weathering Experiment

By considering possible interior circumstance as the inside condition of hazardous waste landfill, a total of 12 series of combinations of gas, humidity, and temperature were set up (Namely, 3 gas conditions, 2 humidity conditions, and 2 temperature conditions (3*2*2=12)). Samples

were put in the dish and it was placed in the chamber of which condition was set at each weathering condition. Each condition was set as follows;

Gas: a) Air, b) N₂ gas, c) CO₂ gas, the chamber was purged by each gas.

Humidity: a) Dry (hygroscopic silica gel was filled in the bottom of the chamber), b) wet (A metal tray was placed at the bottom of the chamber and filled with distilled water)

Temperature: a) Leave at room temperature (Just leave as it is), b) Freeze and thaw repetition (the sealed chamber was put in the freezer once a day. After the sample was frozen, leave it in the room and thaw it).

The samples were cured in chambers under each condition (Fig. 4). Ten samples were prepared for each weathering series and were put in each chamber. From each series, a sample was collected one by one weekly, and it was subjected to the leaching test.

Leaching test was conducted as follows; approximately 2g of a sample taken from each weathering series and then it was weighed. the sample was put into a screw bottle, and 20mL of distilled water was added to make L/S = 10, then it was shaken at 200 rpm for 6 hours with a bench shaker. Thereafter, the supernatant was filtered through a 0.45 μm membrane filter to obtain a test solution, and the concentration of the target metal

was analyzed by ICP. The pH was also measured with a HORIBA pH meter.

3. RESULTS AND DISCUSSIONS

The influence of the repetition of freezing and thawing on the leaching behavior of heavy metals was not observed. Thus, only the results of standing at room temperature are explained below. Moreover, in the following, drying conditions with N₂ gas atmosphere is considered as blanks. This is because, under this condition, no change in the leaching was observed for any of the metal elements (this is considered to be the condition that weathering hardly occurs). The results of Cd and Pb are shown in Fig. 5 and Fig. 6. In addition,

Fig. 7 shows the Mn results that showed similar behavior to Cd. And Fig. 8 shows the results of Zn that showed a marked change in the leaching behavior especially among the elements with high contents.

Under wet conditions (right in each figure (depicted as “wet”), the Cd, Pb, Mn, and Zn were confirmed to decrease in the amount of leaching compared to the result obtained in dry condition (each figure in left), regardless the gas atmosphere. The results in wet condition were below the blank line (i.e., the average of the leaching amount under dry/N₂ condition) shown in the figure. Especially, Leaching amount of Cd and Mn showed clear trend of decrease with elapsed week.

Under wet conditions, adsorbed water is

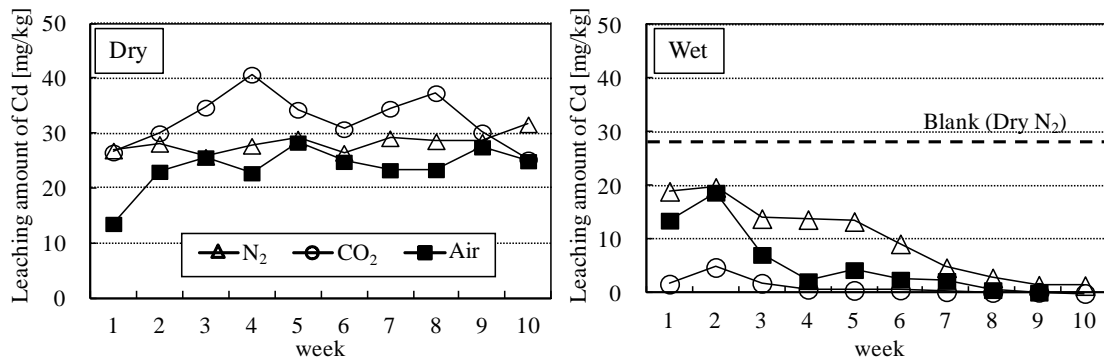


Fig. 5 Change of Cd leaching from dust under each weathering condition

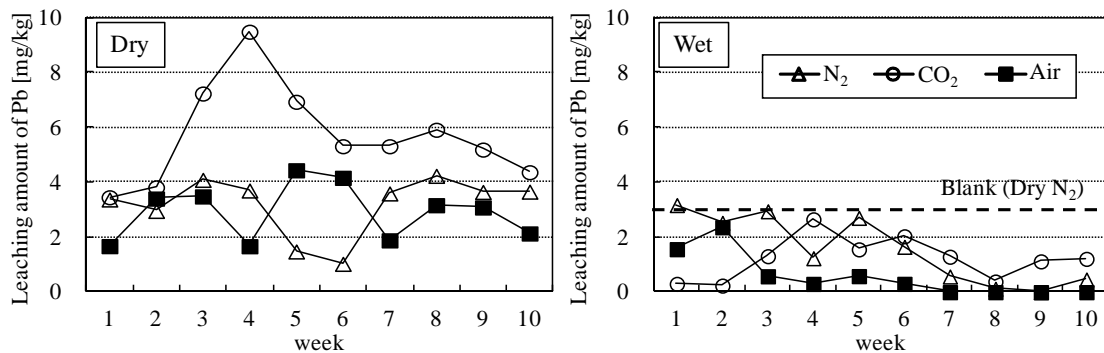


Fig. 6 Change of Pb leaching from dust under each weathering condition

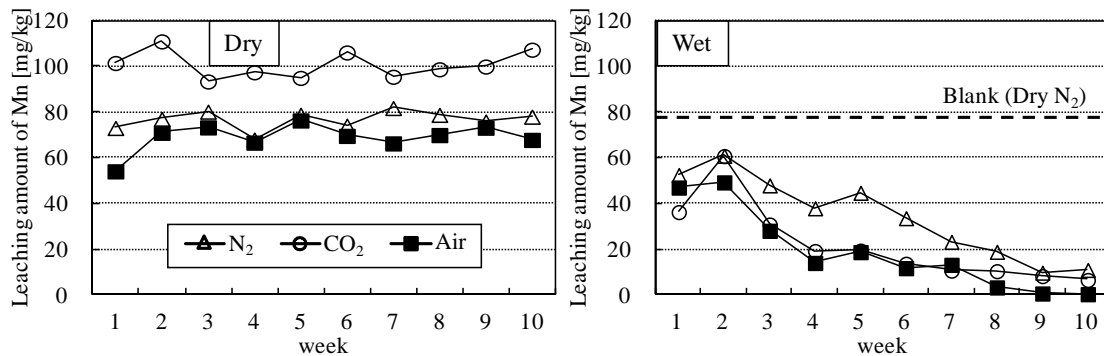


Fig. 7 Change of Mn leaching from dust under each weathering condition

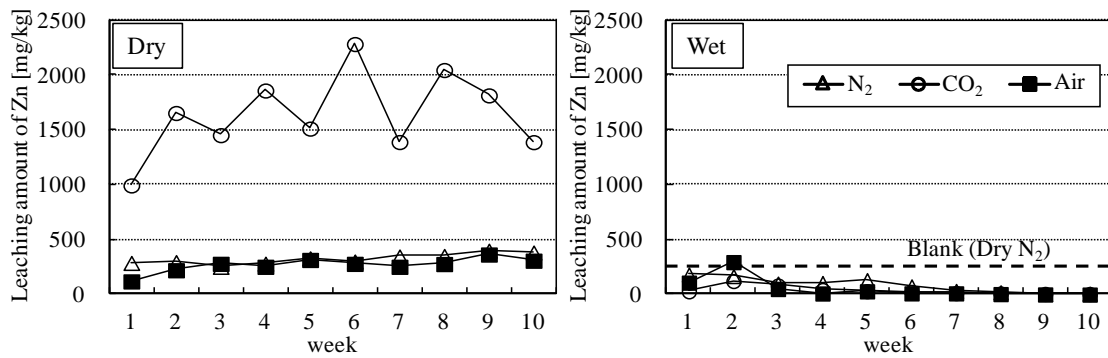


Fig. 8 Change of Zn leaching from dust under each weathering condition

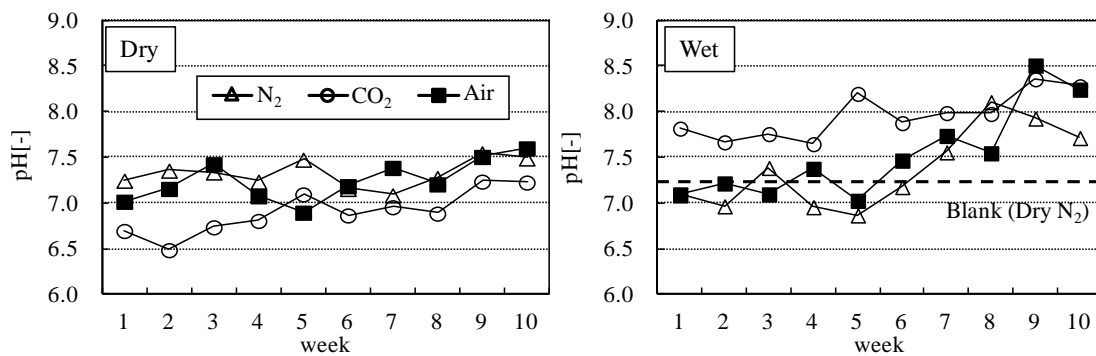


Fig. 9 Change of pH in leaching test under each weathering condition

present on the particle surface because of high humidity, and a gradual increase in pH is confirmed (Fig. 9), so it is considered that the formation of hydroxides of these metals is a factor in the decrease in leaching amount. Cd (Fig. 5) shows a remarkable decrease in leaching amount under wet/CO₂ conditions compared to other gas condition. It is considered that CO₂ in the gas phase is dissolved in surface adsorbed water of the particles to form carbonate.

On the other hand, under the drying conditions shown in the left figure, an increase in the leaching amount of Cd, Pb, Mn, and Zn was confirmed under CO₂ exposure conditions (marked as circle), and a significant increase was particularly observed in Zn. Since there is no water in the dry atmosphere, the increase in the leaching amount is likely to be the influence of the direct reaction between the gas phase CO₂ and the solid particle of dust sample. It is possible that Fe₃O₄ and ZnO, which are the predominant components of the sample, both have adsorption capability, and have suppressed the leaching of heavy metals by working as an adsorbent during the leaching test in case when they do not yet contact with CO₂. There is some reports that these oxides adsorb gas phase CO₂ [8]-[10]. As a result of the dust sample being exposed to CO₂ for a long time, the adsorption capacity is lowered due to the occupation of the adsorption site by CO₂, resulting in the increase of

metal leaching amount may have happened.

4. CONCLUSIONS

The influence of the inside circumstance in the hazardous waste landfill on the leaching behavior of heavy metals were examined from the accelerated weathering experiment for electric furnace dust which is a kind of hazardous wastes to be disposed at the hazardous waste landfill. Among the main components, a decrease in the leaching amount of Cd, Pb, Mn, and Zn was confirmed in a wet environment where adsorbed water was present on the particle surface. On the other hand, in the dry and CO₂ saturated environment, an increase in leaching amount was observed. The former phenomenon was thought to be the formation of hydroxide through particle surface adsorbed water and the formation of carbonate for Cd by CO₂ gas dissolution, and the latter was considered to be the influence of adsorption of gas phase CO₂ to Fe₃O₄ and ZnO, but further scientific verification is required for each of them. The findings obtained in this study indicate that the leaching of heavy metals from hazardous wastes disposed of in a hazardous waste landfill may change over time depending on the interior circumstance. And it means that active control of the inside circumstance of containment structure may be effective to reduce the risk of future emission of these hazardous heavy metals.

More specifically, if electric furnace dust is disposed of in a hazardous waste landfill, it is effective to control the humidity to a high level for turning hazardous heavy metals into insoluble state. In particular, if it is desired to suppress Cd leaching, to make the gas atmosphere inside landfill to high CO₂ gas concentration (by using CO₂ gas injection, etc.) is effective for carbonate formation. In this study, the conditions of saturated vapor pressure and CO₂ of 100% were adopted to accelerate weathering. However, since the storage period of waste in the hazardous waste landfill is extremely long, it is deemed to be possible to progress insolubilization of hazardous metals without making these conditions extremely high. Until now, no attention has been paid to the internal condition of hazardous waste landfills, and there has been no particular control. Of course, the conditions under which leaching suppression proceeds depend on the type of waste (dust, sludge, ash, etc.). One pit in hazardous waste landfill sites in Japan is regulated to be 250 m³ or less, so the size is small. Therefore, it is feasible to limit the type of waste accepting to each pit to one kind and promote weathering by controlling the interior circumstance to the favorable condition for each waste. What this research suggests is that leaching suppression of hazardous heavy metals is possible by the control of inside condition and acceleration of weathering and it is achieved by using extremely long storage period.

5. ACKNOWLEDGMENTS

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