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# Characteristics and mobility of heavy metals in an MSW landfill: Implications in risk assessment and reclamation

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### **Abstract**

To assess the reclamation feasibility of a landfill, the characteristics, distribution, and mobility of heavy metals in the landfill were investigated. The refuse was characterized as containing high concentrations of heavy metals, a relatively high pH, and a high ratio of NH4-N to total nitrogen (TN). The results of heavy metal distribution showed that relatively high levels of heavy metals were accumulated in the landfill. Sequential extraction revealed that the relative amounts of heavy metals were different in the samples. Zn demonstrated the greatest mobility compared to other heavy metals, whereas Cd was well retained in the landfill. Leaching experiments indicated that the mobility of heavy metals in the landfill was generally low under normal neutral conditions. However, release of heavy metals can be increased greatly when medium conditions become more acidic and aerobic.

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# **1. Introduction**

Landfill as refuse disposal is a widely accepted technology, especially in developing countries, because of its low investment and operational costs. More than 90% of the refuse is disposed by landfill in China, and China has recently closed more than 1000 landfills because of environmental concerns.

As the largest landfill in China, Shanghai Laogang Refuse Landfill was constructed in 1985 along the shore of the East China Sea, and started operation at the end of 1989. This facility treats about 5000 tonnes of refuse daily, 75% of which is generated in the city of Shanghai. Up to 2005, it had disposed of around 30 million tonnes of refuse. Our previous investigation predicted that chemical oxygen demand (COD) and biological oxygen demand (BOD) of the landfill leachate could still meet the strictest standard for pollution control of a municipal solid waste (MSW) landfill in China after a 15-year natural attenuation of the facility. The predicted time for NH4-N concentrations to reach the emission standard was 24–26 years, or even longer.

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Settlement and changes in refuse composition for total sugar, raw cellulose, biological degradable matter, volatile solids, and organic carbon were continually monitored. Mathematical simulations between these parameters and refuse age were developed based on the monitoring results. It was predicted that the refuse might be fully stabilized after approximately 22–30 years of placement, and cumulative settlement might eventually be over 30% of the initial height [\[1\].](#page-6-0)

After Shanghai Laogang Landfill was closed in 2005, the difficulty in establishing a new landfill required Shanghai municipalities to explore the possibility of reclamation of the Shanghai Landfill to renew the disposal capacity and reuse the stabilized landfill. Landfill reclamation was originally conceived as a method to address groundwater contamination problems at unsanitary landfills. Bioreactor systems, such as leachate recirculation, were applied to promote rapid stabilization of refuse in the landfill. Therefore, the MSW landfill had been promoted as a potential bioreactor rather than a waste storage facility. Reclaiming stabilized refuse in landfill offers potential economic benefits because it creates additional disposal capacity for the placement of fresh refuse, and recycles the stabilized, valuable resource. This is important for promoting the sustainable environmental development of a society. However, the presence of hazardous

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chemicals in residues of landfills is often a limit for its reclamation. Based on our previous study, most organic chemical substances will eventually either be degraded through biochemical reactions in the landfill, or be leached out from the landfill with water movement. However, the majority of heavy metals will remain in the landfill because heavy metal migration is very limited compared to the amount of metals accumulated in the landfill [\[2,3\], e](#page-6-0)specially in anaerobic processes. The slow movement of heavy metals is the result of heavy metals being subjected to strong sorption on soil particles, precipitation under anaerobic conditions, and chelation with inorganic and organic ligands in landfills [\[4,5\].](#page-6-0)

Before the reclamation of a landfill can be implemented and the residues can be reused outside the landfill environment, it is necessary to comprehensively characterize heavy metals in the landfill. There are limited published studies in this area. Flyshmmar described heavy metals in an MSW deposition cell and their transformation and distribution [\[6,7\].](#page-6-0) By analyzing heavy metal content in soil reclaimed from an MSW landfill, Pradeep et al. assessed the possible limitation of reuse of the reclaimed soil outside of the landfill [\[8\].](#page-6-0) Remon et al. reported soil characteristics in a former metallurgical landfill, assessed the risk of heavy metal mobility, and evaluated reclamation feasibility of the landfill [\[9\].](#page-6-0)

The objective of this study was to investigate the characteristics and migration of heavy metals in Shanghai Laogang Landfill. Additionally, the effect of disposed time on the stabilization of heavy metals and leaching was also investigated to identify any indications of possible environmental hazards of the reclaimed residue, and the possibility of heavy metal migration under different conditions.

# **2. Materials and methods**

#### *2.1. Description of the landfill site*

Shanghai Refuse Landfill, the largest landfill in China, was constructed in 1985 along the shore of the East China Sea, and it is located on land that was formed by silt deposition carried by the Yangtze River. This facility initiated its operation at the end of 1989. Occupying a total area of about  $6 \text{ km}^2$ , the landfill consisted of many clay landfill cells dug in situ with 10 ha for each cell. Refuse of different years was disposed in different landfill cells with a filling height of 4 m. The landfill leachate was collected by a leachate collecting system under the bottom of the cell and discharged through a separate pipeline. Two landfill leachate treatment systems were constructed in the landfill, and a gas collection system was installed in each closed cell to collect gaseous products.

#### *2.2. Sampling and basic analysis*

# *2.2.1. Refuse sampling*

Because refuse samples are highly heterogeneous, a systematic sampling plan was designed. Refuse samples were collected from four chosen sites of different landfill cells that had been closed over a period from 1991 to 1997. First, surface vegetables and cover soil were removed manually, and refuse samples were collected at 1-m intervals from the surface to the maximum depth of 4 m. Refuse samples of at least 50 kg were collected at each depth, and total of 16 samples were collected from 4 sampling sites in each landfill cell. The sampling area was approximately  $2-3$  m<sup>2</sup>, depending on the section area of the dig.

All the refuse samples from each landfill cell were mixed and homogenized. After the samples were oven-dried at  $50^{\circ}$ C, the basic physical composition of the samples was measured by weighing the components. Nondegradable materials, such as stones, glass bottles, plastic films, rubber, etc., were removed. For further basic chemical analysis, the remaining sample was sieved through a 10-mm sieve. After nondegradable materials remaining on the 10-mm sieve were removed, the remaining sample was crunched and ground to less than 10 mm by hammer, ball miller, and grinder, if necessary. The entire sample that passed through the 10-mm sieve was homogenized and stored in a container for analysis. These fractions were used to represent the entire sample pool collected from the landfill because these fractions presumably contain most of available and reactive metals in the refuse [\[8\].](#page-6-0)

The characteristics of the refuse samples were determined according to procedures described by the SW-846 Manual (USAEPA). Sample pH values were measured in a distilledwater extract of 1:10 (w/v) by a PHB-4 pH meter. TOC content was determined by SSM–TOC (TOC-Vcpn SHIMADZI, Japan). The cation exchange capacity (CEC), organic matter, and total nitrogen (TN) of the refuse samples were determined according to most-accepted procedures [\[10\].](#page-6-0)

# *2.2.2. Landfill leachate*

At least 2 L of landfill leachate was collected from the same landfill cells that the refuse samples were collected, and the leachate samples were stored in 5-L polyethylene bottles. All leachate samples were kept in the dark and transported to the lab for analysis within 24 h of sampling.

The pH of the leachate samples was measured directly using a PHB-4 pH meter. The COD, TN, and NH<sub>4</sub><sup>+</sup>-N were determined according to standard procedures [\[11\].](#page-6-0)

#### *2.3. Heavy metal distribution*

Approximately, 0.250 g of refuse sample was transferred to a digestion vessel. Six milliliters of 65% HNO<sub>3</sub>, 1 mL of 30% H2O2, and 1 mL of 40% HF were added to the vessel. The sample was then digested in Microwave Solvent Extraction Labstation (ETHOSE, Italy) for 20 min, and vented for 3 min. The digestion solution was filtered through a 0.45-µm membrane and the filtrate was diluted to 100 mL in a flask. The concentrations of heavy metals were determined by ICP–OES (Optima 2100DV, Perkin-Elmer, USA). Quality control procedures were carried out including duplicates and blanks during the analytical procedure.

To determine the concentrations of heavy metals in the landfill leachate, 50 mL of landfill leachate was placed in a digestion vessel. Seven milliliters of  $65\%$  HNO<sub>3</sub> and 1 mL of  $30\%$  H<sub>2</sub>O<sub>2</sub>

Table 1 Extraction sequence procedure

Step	Extractant	Fraction	Target forms
	$1 M MgCl2$ (pH 7)	Exchangeable	Exchangeable cations
$\overline{2}$	1 M NaAC (pH 5)	Acid soluble	Carbonates
$\mathcal{R}$	$0.04 \,\mathrm{mol} \,\mathrm{L}^{-1}$ $NH2$ HCl (pH 3)	Reducible	Mn and Fe oxides
$\overline{4}$	$30\%$ H <sub>2</sub> O <sub>2</sub> (pH 2)	Oxidizable	Organic matter or sulfides

were added to the vessel. The mixture was then digested and analyzed using the same procedures described above.

## *2.4. Sequential extraction*

A series of progressively stronger extraction procedures were employed to fractionate metals into exchangeable, acid-soluble, reducible, oxidizable, and residual fractions. Details of the sequential extraction procedure are given in Table 1 [\[12,13\].](#page-6-0) After filtration through a  $0.45$ - $\mu$ m membrane, the concentrations of heavy metals in the extracted fractions were analyzed by ICP–OES. Blank extractions (without samples) were performed throughout the entire procedure for each set of analyses.

#### *2.5. Leaching experiment*

To investigate the mobility of heavy metals in refuse with water, metal leaching by deionized water was examined. The details of the experimental protocol are as follows [\[14\].](#page-6-0)

A refuse sample of 50 g was placed in a polypropylene container and deionized water was added until a liquid/solid ratio of 10 was achieved. The container was agitated on a controlledtemperature horizontal vibrator at 110 rpm for 8 h and then stabilized for 16 h. The mixture was filtered through a mediumspeed quantitative filter paper (5B), and the filtrate was collected in a 200-mL volumetric flask. After the filtrate was digested, the concentrations of heavy metals were analyzed by ICP–OES. To give a better representation of sample variability, the leaching experiment was replicated three times, and the measure-



ments of the concentrations of heavy metals were carried out in triplicate.

# *2.6. pH-dependent leaching*

To investigate the leaching behavior of heavy metals under comparatively low pH, a pH-dependent leaching experiment was carried out in this study. The procedural details are as follows [\[15\].](#page-6-0)

A refuse sample of 20 g was placed in a reactor and deionized water was added until liquid/solid ratio of 10 was achieved. The pH controller was set at desired values of 4 and 6 via connections to acid  $(0.1N HNO<sub>3</sub>)$  delivery pumps and reservoirs. The solution pH value was controlled by a pH sensing and controlling unit. The reactors were agitated by a magnetic stirrer for 8 h. The mixtures were filtered by medium-speed quantitative filter paper (5B), and the filtrates were collect in 200-mL volumetric flasks. After the filtrates were digested, the concentrations of heavy metals were analyzed by ICP–OES.

# **3. Results and discussion**

# *3.1. Basic analysis*

Heterogeneity of the refuse samples was expected. Although extensive efforts were made to improve sample representation and data reliability, the heterogeneity made it difficult to accurately determine the chemical characteristics of the refuse samples. Many factors contribute spatial variation, including waste input variability, disposal manner, and the randomly chosen sites. A summary of the data from the chemical analyses of the residues and leachates is given in Tables 2 and 3.

# *3.2. Heavy metal distribution in different landfill cells*

The total metal content in refuse of the landfill is listed in [Table 4.](#page-3-0) The heavy metal contents did not change greatly with the landfill age. According to the national soil environmental standard (GB15168-1995), the concentrations of Ni, Cr, and Pb

	1991	1993	1995	1997	
pH	$7.65 \pm 0.14$	$7.58 \pm 0.12$	$7.39 \pm 0.19$	$7.45 \pm 0.27$	
Organic matter $(\%)$	$12.20 \pm 0.37$	$15.40 \pm 0.42$	$13.70 \pm 0.34$	$11.50 \pm 0.29$	
TOC $(\%)$	$10.50 \pm 0.32$	$11.30 \pm 0.28$	$9.30 \pm 0.26$	$9.33 \pm 0.32$	
$CEC$ (mmol $kg^{-1}$ )	$159.0 \pm 5.2$	$128.0 \pm 3.7$	$146.0 \pm 3.1$	$170.0 \pm 4.7$	
$TN$ (mg $L^{-1}$ )	$0.42 \pm 0.051$	$0.39 \pm 0.041$	$0.36 \pm 0.038$	$0.46 \pm 0.036$	

Table 3

Chemical characteristics of landfill leachate



<span id="page-3-0"></span>



Sampling date is March 15, 2005.

were under the third standard of the soil environment for all samples. The content of Cu was also generally under the third standard, with the exception of the sample from 1993. The content of Zn was two to three times the limit value.

The concentrations of the heavy metals were similar to those reported by other authors [\[6,16\]. F](#page-6-0)lyshmmar et al. (1998) investigated the concentration of some metals in the fine fraction of MSW samples (less than 2 mm) from a 20-year-old landfill cell. Our mean concentrations of Pb and Cr in the refuse samples were comparable to the results reported by Flyshmmar for a Swedish test landfill cell, whereas our mean Zn, Ni, Cd, and Cu concentrations were significantly higher than those observed by Flyshmmar. It should be noted that the refuse sample that Flyshmmar analyzed was only the fine portion (less than 2 mm) of the sample, different from those used in this study.

As shown in Table 5, heavy metal concentrations in leachate samples were relatively low and below the limits of the integrated wastewater discharge standard (GB8978-1996), although the concentration of Cr is higher in comparison to the concentrations reported by Christensen. The metal with the highest concentration leached from the landfills was Cr, followed by Pb and Ni, and only minor amounts appeared to be leached. The heavy metal concentrations in the refuse were significantly higher than those in the landfill leachate, which was in agreement with the results from other studies [\[3,17\]. T](#page-6-0)he high retention of most heavy metals in the landfill is attributed to the establishment of a top layer after the landfill was closed that maintained a reducing environment. The anaerobic conditions with limited water access in the landfill promoted the growth of sulfur-reducing organisms, and the reduced sulfur may have promoted metal precipitation in the landfill as sulfides. In addition, humic sub-





Sampling date is March 20, 2005.

<sup>a</sup> Not detected, detection limit is  $0.002$  mg L<sup>-1</sup>.

<sup>b</sup> Not detected, detection limit is 0.005 mg L<sup>-1</sup>.





stances produced from degradation of organic substances could also have retained heavy metals in the landfill by adsorption or surface complexation [\[18\].](#page-6-0)

#### *3.3. Sequential extraction*

Metal fractionation using sequential extraction procedures can provide useful information for risk assessment because the fraction pattern is a good indicator of metal distribution and chemical characteristics. Therefore, the reactivity, mobility, and bioavailability of metals at different environmental conditions can be estimated [\[19\].](#page-6-0) The distribution of heavy metals and the percentage of fractionation of heavy metals are shown in [Table 6](#page-4-0) and Figs. 1–6. The different patterns in the distribution of metals suggest fundamental and qualitative differences in the chemical characteristics of these metals in terms of binding and reactivity in these samples.

The exchangeable fraction of metals is generally considered the mobile and available form, and therefore more susceptible to be released. Compared with other heavy metals, Zn and Cr were the highest in the exchangeable fraction, indicating that Zn has a high potential of creating hazardous effects on the environment. On the other hand, only a small fraction of Pb and Ni was present



Fig. 2. Fractionation ratio of Zn.

<span id="page-4-0"></span>Table 6 Extraction procedure of heavy metals at different fractions

Element	Time $(a)$	Extraction concentration (mg $g^{-1}$ )				Mobility	
		Exchangeable	Acid soluble	Reducible	Oxidizable	Residual	
Zn	1991	0.088	0.244	0.834	0.221	ND	
	1993	0.116	0.333	0.881	0.227	$\rm ND$	
	1995	0.094	0.319	0.758	0.224	ND	
	1997	0.073	0.403	1.131	0.255	ND	
Pb	1991	0.008	ND <sup>a</sup>	ND	0.062	0.205	0.253
	1993	ND	0.023	0.048	0.198	0.139	0.658
	1995	ND	0.022	ND	0.149	0.264	0.393
	1997	ND	0.009	ND	0.109	0.216	0.353
Ni	1991	0.003	0.004	0.02	0.005	0.026	0.55
	1993	$ND^b$	0.005	0.019	0.012	0.025	0.331
	1995	ND	0.005	0.01	0.005	0.04	0.585
	1997	ND	0.003	0.019	0.005	0.017	0.6194
Cr	1991	0.01	0.013	0.069	0.02	0.011	0.912
	1993	0.011	0.009	0.076	0.031	0.027	0.821
	1995	0.005	0.022	0.069	0.032	0.023	0.845
	1997	0.005	0.017	0.079	0.027	$\rm ND$	
Cd	1991	ND <sup>c</sup>	$\rm ND$	ND	$\rm ND$	0.0018	$\boldsymbol{0}$
	1993	ND	ND	<b>ND</b>	ND	0.0016	$\boldsymbol{0}$
	1995	ND	ND	<b>ND</b>	ND	0.003	$\boldsymbol{0}$
	1997	$\rm ND$	ND	ND	$\rm ND$	0.001	$\boldsymbol{0}$
Cu	1991	ND <sup>g</sup>	0.03	<b>ND</b>	0.233	0.097	0.73
	1993	0.002	0.041	<b>ND</b>	0.294	0.199	0.628
	1995	ND	0.025	<b>ND</b>	0.204	0.1	0.695
	1997	ND	0.036	<b>ND</b>	0.191	0.075	0.751

<sup>a</sup> Not detected, detection limit is 0.005 mg L<sup>-1</sup>.<br><sup>b</sup> Not detected, detection limit is 0.002 mg L<sup>-1</sup>.

Not detected, detection limit is  $0.002$  mg L<sup>-1</sup>.

<sup>c</sup> Not detected, detection limit is  $0.001 \text{ mg L}^{-1}$ .

in the exchangeable fraction for the sample from 1991, and Pb and Ni showed almost no content in the exchangeable fraction in all of the samples. The low content in the exchangeable fraction of Cu and Cd might be explained by the low solubility product constant of Cu (8.5 × 10<sup>-45</sup>) and Cd (3.6 × 10<sup>-29</sup>) as sulfides.

The main proportion of Zn and Cr were recovered in the reducible fraction, while Cr and Zn were also found in the aciddissoluble and the oxidizable fractions. This suggests that Zn and

Cr might strongly bind to manganese oxides and ferric hydroxides, precipitate as sulfides, or be adsorbed on organic substances [\[20–21\].](#page-6-0) As shown in [Figs. 1–6,](#page-3-0) Cu in the oxidizable fraction showed the highest concentration compared to the other metals. In general, more than 50% of the Ni, Cd, and Pb were in the residual fraction. Acid-soluble and residual fractions of Cu and Ni were also found in all of the samples. The content of Cu and Pb in the oxidizable fraction was greater than the other



Fig. 3. Fractionation ratio of Cd.



Fig. 4. Fractionation ratio of Pb.



Fig. 5. Fractionation ratio of Ni.

metals in that fraction, which indicated that much more Cu and Pb were bound to organic matter or precipitated as metal sulfides compared with other metals. Cd was exclusively found in the residual fraction. In addition, Ni was mainly found in the residual fraction, although a high concentration of Ni was also found in the reducible fraction. Based on these results, the binding of heavy metals to manganese oxides or ferric oxides and their precipitation as sulfides seem to play important roles in the stability of heavy metals in the landfill.

To study metal retention in each sample, fraction extraction factors were calculated ([Table 6\).](#page-4-0) The mobility factor is defined as the sum of content extracted except in the last step (residual fraction) divided by the total content of the sample. The higher the value, the greater is the relative metal mobility [\[22\].](#page-6-0) The results indicated that, in general, the fractionation patterns of Zn and Cu did not differ greatly with the landfill age. Cd demonstrated the greatest stability and was more strongly bound in the landfill compared with the other heavy metals, whereas the mobility of Zn was considered the greatest in all refuse samples. The differences in the mobility of various heavy metals were



Fig. 6. Fractionation ratio of Cu.





caused by the solubility of respective sulfides, hydroxides, or other precipitates, as well as the degrees and modes of complexation with organic substances [\[18\].](#page-6-0)

#### *3.4. Leaching test*

A leaching test by deionized water was used to evaluate the potential environmental risk associated with refuse under water. As shown in Table 7, the content of heavy metals released from the refuse was less than 1% of the total heavy metal content. This indicated that the high contents of heavy metals in the refuse were mostly insoluble, and their tendency to leach under natural conditions was probably very low. This was because the mobilization of heavy metals was restricted by the presence of buffering substances in the landfill during the anaerobic process; the buffering substances are able to buffer solutions in neutral to weak alkaline solutions.

In contrast, more Zn was leached compared with other metals. This was consistent with the extraction result in which Zn exhibited the highest mobile fractions of all the heavy metals investigated in this study. On the other hand, Cd showed the lowest leaching because it existed only in the residual fraction. In addition, the leachability of heavy metals by deionized water was generally lower than the sequence extraction. This may be the result of the high pH values of the residues, which limited the amount of metals to be released into solution.

# *3.5. pH-dependent leaching*

The effects of pH on metal leaching are presented in Table 8. As shown in the table, a general trend can discerned that more heavy metal was leached at pH ∼ 4. At a low-pH value, Zn, Cu, and Ni demonstrated high leachability. The highest leaching concentration of Zn was more than 29 mg L−1. Compared with the results of sequent extraction, it can be found that the increased Zn leaching was probably due to its high acid-soluble proportions.





<span id="page-6-0"></span>At a high pH value, the heavy metals can be immobilized by sulfides or oxides, or by binding to organic substances. With the decrease of pH, Zn bound to carbonates can be dissolved and released. The same portion of Zn could not be released during the leaching by deionized water because the residues had a strong buffering capacity to keep the medium pH greater than 7. The result of pH-dependent leaching can help explain the exact mechanism involved in heavy metals releasing from residues. When residues are mixed with water of low pH, the protons in the low-pH medium will deplete the buffering capacity, resulting in dissolution or destruction of carbonates to release heavy metals bound to the carbonates. If pH is further decreased, the heavy metals adsorbed to manganese and ferric oxides may also be released from the residues. If pH values were kept below 2, the heavy metals bound as sulfides may be released when the medium condition is suitable for the oxidation of sulfides. Therefore, it can be concluded that heavy metals in the residues can be an important environmental issue at low-pH and highredox-potential conditions. It can be concluded that the potential leachability of heavy metals was determined by the pH value of the refuse. However, the leaching data with the deionized water might not be conclusive in determining whether or not the heavy metals in refuse would cause secondary pollution because the metal leachability by water often strongly depends on pH.

## **4. Conclusion**

In this study, we investigated refuse samples in a closed landfill. The results of basic analyses indicated that the landfill was at its methane-producing phase, and the chemical characteristics of the refuse become stabilized after being disposed for more than 10 years. In general, the residue contained relatively high levels of heavy metals. Although the content of each heavy metal in the refuse varied, it did not change greatly with the landfill age. A low concentration of heavy metals in the leachate suggests that the heavy metal became stabilized in the landfill and only a small quantity of heavy metals was discharged in the closed landfill.

A sequential extraction procedure was used to evaluate the most labile forms of heavy metals. The results showed that the mobile fractions of the heavy metal were quite different and largely element-dependent. High concentrations of Zn were associated with the soluble, exchangeable, and carbonate fractions, whereas other heavy metals were mostly associated with the residual mineral components.

The leaching tests showed that heavy metals released from the refuse were low. It can be concluded that the heavy metals could be considered as quite insoluble, and their leaching was probably low under normal neutral conditions, although the refuse contained high concentrations of potentially toxic heavy metals. However, the fact that heavy metals are insoluble under normal neutral conditions does not necessarily imply that heavy metals are immobilized and pose no hazard to the environment and human health. The pH-dependent leaching results demonstrated that heavy metals in the residues can be an important environmental issue at low-pH values. Excavation of the refuse from the landfill would lead to an increase in mobilization of the

deposited metals when redox conditions are suitable. Therefore, an accurate risk assessment of the refuse after reclamation must include pH and redox studies to identify potential hazards. In addition, the impact of the leaching of organic pollutants and the interactions among elements in the residue need to also be addressed prior to a reclamation project.

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