



Article Investigation and Evaluation of Flue Gas Pollutants Emission in Waste-to-Energy Plant with Flue Gas Recirculation

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Abstract: The emissions of pollutants by waste-to-energy power plants, which contain more toxic substances owing to the complicated composition of municipal solid waste (MSW), such as NOx, SO₂, HCl, HF, particulate matter, and heavy metals, has attracted increasing attention worldwide. To effectively control the pollutants, a flue gas cleaning system is indispensable in the operation of MSW incineration power plants. In this study, the flue gas cleaning system in a waste-to-energy power plant with flue gas recirculation (FGR) was evaluated. The concentrations of various pollutants were measured and compared with the standards at home and abroad. The results indicated that NOx emission can be effectively reduced by FGR, and that the emission concentration of NOx may meet the national emission standards only by adopting FGR. However, the emission levels of HCl and PM exceeded the limits in legislative standards; therefore, operation optimization or retrofit of a deacidification system and bag filter were proposed to comply with the international standards and near-zero-emissions goal.

Keywords: municipal solid waste; flue gas recirculation; pollutants emission; flue gas cleaning

1. Introduction

With the rapid development of urbanization and the economy, the growth rate of municipal solid waste (MSW) has been far higher than its harmless disposal rate, the output and accumulated stock of MSW increasing year by year. In 2019, the output of MSW in China reached 0.34 billion tons, and China's MSW piles accumulated a stock of over 6 billion tons, occupying a large area of land and causing serious environmental pollution, with the "garbage siege" phenomenon existing in two-thirds of the cities in China [1]. Therefore, the harmless disposal of MSW has attracted much attention. At present, the international commonly used methods of waste disposal include landfill, composting, and waste-to-energy (WTE) [2–4]. Improper waste management can have long-term impacts on the environment, such as pollution of air, soil, surface, and ground water [4,5]. Apart from soil and ground water pollution, the major consequence of landfill is the generation of methane (CH₄) gas from the decomposition of MSW, with CH₄ contributing approximately 21% of global greenhouse gasses (GHG) [5]. Full names of all abbreviations used in this paper can be seen in Appendix A.

China, the world's largest energy consumer and importer [6], faces considerable demand for energy due to its rapid economic development. Discarded MSW can be utilized as an energy source to produce electricity and/or steam for heating [7]. Therefore, WTE is receiving increased attention. Furthermore, as the goals of "carbon peak" and "carbon neutral" were put forward by China's President Xi [8], new energy substitution,



Citation: Wang, W.; Tian, S.; Long, J.; Liu, J.; Ma, Q.; Xu, K.; Zhang, Z. Investigation and Evaluation of Flue Gas Pollutants Emission in Waste-to-Energy Plant with Flue Gas Recirculation. *Atmosphere* **2022**, *13*, 1016. https://doi.org/10.3390/ atmos13071016

Academic Editor: Célia Alves

Received: 19 May 2022 Accepted: 20 June 2022 Published: 24 June 2022

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Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). energy conservation and emission reduction, and resource recycling will become crucial choices. WTE is one of the most intuitive ways to reduce carbon emissions. For instance, MSW incineration can reduce GHG emissions such as methane, which is inevitable during landfill [9,10]; additionally, WTE can replace part of coal-fired power plants to produce the same amount of electricity and/or steam for heating, thereby reducing GHG emissions. For example, an MSW power plant with an annual capacity of 500,000 tons can generate about 200 million kWh of electricity, equivalent to saving 55,000 tons of standard coal and reducing CO_2 emissions by about 150,000 tons.

In China, the composition of MSW is complex, being primarily composed of kitchen and building waste, textiles, plastics, and metals [11,12]. The components and calorific value of MSW are related to the developing level of the economy. With the improvement in living standards, the calorific value of MSW is generally above 5000 kJ/kg [13]. With the increase in MSW incineration power plants, much public attention has been paid to pollutant emissions, which are more complex than for coal-fired plants [14-16]. The initial compositions and concentrations of pollutants in flue gas are determined by the components of MSW and the combustion mode [3]. The emitted pollutants, including NOx, acid gases (SOx, HCl, HF, etc.), heavy metals, particulate matter (PM), and toxic organic compounds [17], can seriously impact the environment and human health. To control the emission of pollutants, one method is to reduce the formation of pollutants during MSW combustion by controlling the operation parameters such as oxygen and temperature in the combustion area through an appropriate air-fuel ratio and air staging [4]. The other is post-combustion pollutants removal through the adoption of a flue gas cleaning system, which includes a series of different processes [18]: first, the removal of acid gases by means of alkaline reagents: dry neutralization with $Ca(OH)_2$ or $NaHCO_3$, and semidry neutralization with $Ca(OH)_2$ and wet scrubbing [3]; second, the removal of NOx through NH₃ or urea by selective catalytic reduction (SCR) and selective noncatalytic reduction (SNCR) [19]. Activated carbon injections are used for the removal of dioxins and heavy metals, with the removal of dust achieved through electrostatic precipitators or bag filters, the latter being more widely used in WTE plants owing to its stronger dust removal effects [20,21]. The existing research has mainly focused on the small-capacity waste incineration unit. As the technology matures, the capacity of waste incineration power plants considerably increases, and the operation and pollutant discharge characteristics of units change, so it is necessary to study the operation characteristics of large-capacity units. As the environmental requirements become stricter, it is vital to evaluate the flue gas cleaning system and control the level of pollutant emissions below national standards. The aim of this study was to evaluate flue gas pollutants emissions in a newly built largecapacity WTE plant with FGR, analyze the causes of excessive pollutants emissions during operation, and provide guidance for future operation optimization and pollutant control. The emission concentrations of pollutants, including NOx, acid gases, heavy metals, and PM, were measured and compared with the standards at home and abroad, which indicate a direction for the improvement in the flue gas cleaning system to meet international standards and the near-zero-emission goals, namely 5 mg/m^3 for soot, 35 mg/m^3 for sulfur dioxide, and 50 mg/m³ for nitrogen oxide.

2. Experiments and Methods

2.1. Materials

The MSW considered in this study was collected in Luoyang, China, and was composed of kitchen waste, curbside waste, and a smaller component of construction waste. The designed bulk density and lower heating value (LHV) of MSW were about 400 kg/m³ and 6699 kJ/kg, respectively. The designed properties of MSW are listed in Table 1. The contents of combustible matter and ash were 35.38 wt% and 20.31 wt%, respectively. The received moisture content of MSW was 44.31%, which was not conducive to stable combustion, so a sufficient leaching and fermentation process was required in the storage tank before MSW incineration, usually no less than 72 h.

MSW	Proximate Analysis (wt%) ^a				Ultimate Analysis (wt%) ^b					
	Moisture	Combustible Matter	Ash	LHV (kJ/kg)	С	Н	0	Ν	S	Cl
Received	44.31	35.38	20.31	6699	54.55	7.72	34.62	1.90	0.31	0.90
Received	44.31	35.38	20.31	6699	54.55	7.72	34.62	1.90	0.31	_

 Table 1. The designed proximate and ultimate analysis of MSW.

^a As-received basis, ar. ^b As dry ash-free basis, daf.

2.2. Plant Description

Located in Luoyang, China, the observed MSW incinerator power plant, whose waste treatment capacity was 500 tons per day, was put into operation in 2017. The process flow diagram of MSW incineration is shown in Figure 1. It primarily consisted of five subsystems: waste feeding, heat exchange, incinerator, waste heat boiler, and flue gas cleaning systems. The high-temperature flue gas produced by MSW combustion heated the feed water through heat transfer, generating superheated steam, which drove the steam turbine for further power generation. Then, after treatment by the flue gas cleaning system, the flue gas could be discharged to the atmosphere through the stack.



Figure 1. Schematic diagram of the MSW incinerator power plant.

The flue gas cleaning system (see Figure 1) was designed to gradually decrease the flue temperature. The first step was the use of a semidry reactor using lime slurry (calcium hydroxide, Ca(OH)₂ as the reactant) for the removal of acid gases such as HCl, HF, and SO₂. Then, the flue gas was further treated by the dry method (sodium bicarbonate, NaHCO₃ as the reactant) and activated carbon, which were injected into the flue gas duct before the bag filter, to remove the remaining acids, VOCs, dioxins, and heavy metals in the flue gas. The treated flue gas entered the bag filter for the removal of dust, followed by an extraction of waste heat from the flue gas by a heat exchange tube; finally, the purified flue gas was emitted into the air through the stack. Notably, FGR technology was adopted in this plant to inhibit the generation of NOx. The flue gas was extracted from the tail of the bag filter system and supplied to the corresponding part of the furnace instead of the secondary air. Due to the effectiveness of the technology, no further injection of urea solution was required under 100% maximum continuous rating (MCR) condition. However, the SNCR system was also installed for backup in case the FGR system was out of operation. The specifications of the main equipment and environmental protection facilities of this plant are listed in Table 2.

Name of Facility	Name of Item Facility		Parameter	
	Type of incinerator	-	Reciprocating mechanical grate furnace	
Incinerator and waste heat	Daily waste treatment capacity	t/d	500	
boiler	Boiler rated evaporating capacity (MCR)	t/h	51.80	
	Rated steam pressure	MPa	4.00	
	Main steam temperature	°C	400	
	Type of denitrification system	-	SNCR + flue gas recirculation	
	Flow of recirculating flue gas	m ³ /h	0~26,240	
Denitrification system	Reagents of SNCR	-	Urea	
	Number of nozzle	-	12 (three layers arrangement)	
	Flow rate of urea (per nozzle)	L/min	3.2	
Descidification system	Туре	-	Semi-dry method (rotating spray reaction towers) + dry method (NaHCO3 injection) + activated carbon injection	
Deactaincation system	Inlet temperature of semidry reactor	°C	190	
	Consumption of Ca(OH) ₂ (semidry method)	kg/t MSW	8.0	
	Consumption of NaHCO ₃ (dry method)	kg/t MSW	2.5	
	Consumption of activated carbon	kg/t MSW	0.5	
	Туре	-	Bag dust collector	
	Flue gas temperature	°C	155	
Dust collector	Removal efficiency	%	>99.80	
	Dust content at outlet (11% O ₂ , dry basis)	mg/m ³	<8.00	
	Air leakage rate	%	2%	

Table 2. Specifications of the main equipment and environmental facilities.

2.3. Sampling and Analysis

The sampling and test started after the combustion condition of the unit was stabilized for approximately 0.5 h, the test lasting for 6.0 h. To protect the monitoring instrument, pollutants emissions were consecutively sampled several times during the test. Usually, each test lasted 40 min with a 20 min cooling period for the instrument, and then we repeated the test, thereby obtaining 6 groups of sampled data during each test. The average value of the 6 groups of sampled data was used as the final result of the test, and the standard error of the measured values was recorded. To evaluate the flue gas cleaning system, the emission characteristics of acid gas, NOx, heavy metals, total organic carbon (TOC), and PM in the flue gas were considered. The unit was in normal operational condition when sampling and testing occurred on site.

We referred to the standard [22] for the sampling and testing of slag and ash. The detailed testing method of NOx was previously described [18]. In WTE power plants, NO accounts for more than 90% of the total NOx, so the NOx considered in the study was the concentration of NO. The SO₂sampling and analysis method was determined according to the standard [23]. Because of the low dew-point characteristics of SO₂, a heatable probe (model: PSP4000, M&C TechGroup, Ratingen, Germany) and heat-tracing tube were used to avoid the cryocondensation of SO₂. The system diagram of SO₂ sampling and analysis is shown in Figure 2. The sampling tube was inserted into the gas duct to extract flue gas from the rear flue. The extracted flue gas went through the heat-tracing tube and entered the flue gas preprocessor, then the concentrations of SO₂ and O₂ were measured by a multichannel flue gas analyzer (model: NGA2000-MLT3, Rosemount Inc., Shakopee,

MN, USA). To analyze the emission levels of HCl and HF, an ion chromatograph (model: PIC-10A, Qingdao Puren Instrument Co., Ltd, Qingdao, China) was used according to the standard method EPA 26 [24]. For the measurement of PM, an automatic smoke sampler (model: 3012H, Xian Yima Optoelec Co., Ltd., Xi'an, China) and the parallel sampling method were used to collect the PM from the flue gas. Then, the filter cartridge was dried, cooled, and weighed, and the difference in its weight before and after sampling was the resulting value [25]. To analyze the levels of heavy metals, 10% H₂O₂ and 5% HNO₃ solutions and an inductively coupled plasma-mass spectrometer (model: ICP-MS Agilent7500A, Agilent Technologies, Santa Clara, CA, USA) were used according to the standard method EPA 29 [26], and the TOC was measured according to the standard [27].



Figure 2. System diagram of SO₂ sampling and analysis.

3. Results

3.1. Emission Level of NOx

The emission of NOx has been a major environmental problem as it is an acid rain precursor and can form photochemical smog [28,29], which seriously threatens human health and the environment. The control of NOx emission with the aid of MSW incinerators has attracted growing concern in recent years, especially as the regulations on pollutants emissions are becoming increasingly stringent. FGR technology was used in the power plant, and the NOx emission characteristics under different flow rates of recirculating flue gas were investigated. The combustion state and the NOx emission characteristics under different conditions are shown in Table 3. Notably, the SNCR system was out of operation in all conditions considered. The recirculating flue gas had a significant influence on the emission characteristics of NOx. The NOx emission concentration decreased from 209.54 to 126.15 mg/m^3 as the FGR valve shifted from fully closed to fully open. When the FGR valve was fully closed, the NOx emission concentration was higher than the European Community (EC) emission limit [30] and the NOx emission limit of the plant, while it met the Chinese national standard (CNS) [31]. When the FGR valve was fully open, the NOx concentration met the three different emission limits, so the FGR valve was fully open in normal operation.

Item	Unit	Condition 1	Condition 2	Condition 3
Opening of recirculating flue gas damper	%	100	50	0
Flow rate of recirculating flue gas	m ³ /h	16,163.93	9345.65	0.00
Flue gas temperature at outlet of economizer	°C	193	195	198
CO ₂ emission concentration	%	14.07	14.13	13.92
CO emission concentration	mg/m^3	11.48	11.30	9.56
NOx emission concentration	mg/m ³	126.15 ± 5.60	178.90 ± 6.23	209.54 ± 6.45
limit of plant (daily average)	mg/m ³		180.00	
CNS emission limit [31] (daily average)	mg/m ³		250.00	
EC emission limit [30] (daily average)	mg/m^3		200.00	

Table 3. The combustion and operation states of the boiler.

3.2. Emission Levels of Other Acid Gases

The semidry and dry techniques were used to reduce the acid gases in the power plant. The flue gas from the waste heat boiler entered the breeching of the spray reactor, in which a suitable spiral guide plate was arranged, spiraling clockwise downward. The lime slurry entered the high-speed rotating rotary atomizer, located on top of the spray reactor, and atomized the lime slurry into droplets with a diameter of about 50 μ m. The droplets proceeded counterclockwise downward and a flue gas was formed, which was beneficial for the droplets and flue gas, and enhances the sorption of acid gases to promote the neutralization reaction. After treatment by the semidry technique, the flue gas was further treated by the dry technique, in which the NaHCO₃ was injected into the outlet of the semidry reactor before the bag filter and the residual acid gases were further processed.

The emission concentrations of acid gases in the exhaust gas are listed in Table 4. The emission concentration of SO_2 was 23.65 mg/m³, and those of HCl, HF, and TOC were 16.90, 0.58, and 0.62 mg/m³, respectively, all of which met the Chinese national standard [31]. However, the concentration of HCl was much higher than the EC emission limit [30] and the emission limit of the plant. So, other control technique for acid gases removal should be adopted or the existing device should be further improved.

Table 4. The emission concentrations of other acid gases in the flue gas.

Acid Gas	SO ₂	HCl	HF	TOC
Unit	mg/m ³	mg/m ³	mg/m ³	mg/m ³
Value	23.65 ± 2.31	16.90 ± 2.52	0.58 ± 0.06	0.62 ± 0.03
Emission limit of the plant (daily average)	30.00	8.00	1.00	10.00
CNS emission limit [31] (daily average)	80.00	50.00	-	-
EC emission limit [30] (daily average)	50.00	10.00	1.00	10.00

3.3. PM Emission Levels

PM plays an important role in the formation of fog and haze [32]. In most Chinese cities, hazy weather has become a common phenomenon, especially in winter. Due to the aerodynamic diameter, most particles originating from power plants are inhalable (PM10, with a diameter of less than 10 μ m) [33]. These particles can harm human health, and are one of the main factors influencing haze formation, which is inconvenient for human life such as leading to traffic accidents [33]. Thus, an effective and appropriate flue gas cleaning system should be selected.

In this power plant, the bag filter was selected for the removal of PM, for which the designed removal efficiency was 99.9%. In this investigation, the concentration of PM was 21.83 mg/m³, which was a little higher than the Chinese emission limit of 20 mg/m³ [31], and far beyond the EC emission limit (10 mg/m³) [30] and the plant's requirement (8 mg/m³). Therefore, to meet the international emission standard, the appropriate dust removal technique should be applied or some measures should be adopted to enhance the performance of bag filters.

3.4. Emission Levels of Heavy Metals

The composition of MSW is complex, and most MSW is not diligently sorted before entering the incinerator. Some wastes with high contents of heavy metals, such as rubber, newspapers, plastics, and batteries, are incinerated together. Heavy metals are relatively toxic pollutants in the flue gas, which cannot be generated or destroyed in the incineration process, but can be migrated or transformed [34]. Some heavy metals exist in waste water, slag, or fly ash in a solid state, whereas others change from a solid into a gas state at high temperatures, and exist in the flue gas in the form of gas or attach to particulates in the flue gas [3]. In addition to being removed by a bag dust collector, the rest of the gaseous heavy metals are discharged into the atmosphere and remain for long periods, undegraded by microorganisms in the environment. The heavy metals enter the human body through the interaction of the atmosphere, water, and organisms, and continue to accumulate. After reaching a certain amount, they cause pathological changes in human tissues and organs, and result in serious harm through carcinogenesis and teratogenesis. Compared with the heavy metals in waste water, slag, and fly ash, heavy metals discharged into the atmosphere with flue gas are easily and passively absorbed by plants and breathed into the lungs of human due to their large diffusion and wide influence ranges [35]. Therefore, effective measures should be taken to reduce heavy metal emissions with flue gas during waste incineration treatment.

In the cleaning system studied, the flue gas entered the rotating spray reaction tower, where it was well-mixed with lime slurry. The heavy metal particulates in the flue gas accumulated through mutual collision, and the diameters gradually became larger and heavier, so the particulates were removed by the baghouse. Simultaneously, the temperature of the flue gas dropped to about 140 °C after passing though the reaction tower. Due to the cooling process, the vaporized heavy metals condensed to form fine particulates, which grew through impaction and agglomeration with the lime drops. Subsequently, the flue gas passed through the NaHCO₃ and activated carbon injectors, and the heavy metals reacted with NaHCO₃ to form precipitate or were adsorbed by porous activated carbon, which was further removed by the bag dust collector. Finally, the flue gas was discharged into the atmosphere through the stack. The emission concentrations of heavy metals are shown in Table 5. The concentrations of Hg and Cd + Ti were very low after passing through the cleaning system, which were far below the requirement in the regulations. However, the concentrations of Pb + Cr were a little high, at 0.21 mg/m³, although they met the emission limits.

Hg	Cd + Ti	Pb + Cr
mg/m ³	mg/m ³	mg/m ³
1.45×10^{-4}	3.90×10^{-3}	0.21
0.05	0.05	0.50
0.05	0.10 ^a	1.00 ^b
0.05	0.05 ^c	0.50 ^d
	Hg mg/m ³ 1.45×10^{-4} 0.05 0.05 0.05	HgCd + Ti mg/m^3 mg/m^3 1.45×10^{-4} 3.90×10^{-3} 0.05 0.05 0.05 0.10 a 0.05 0.05 c

Table 5. The emission concentrations of heavy metals in the flue gas.

^a The total concentration of Cd and Ti. ^b The total concentration of Sb, As, Pb, Cr, Co, Cu, Mn, and Ni. ^c The total concentration of Cd and Ti. ^d The total concentration of Sb, As, Pb, Cr, Co, Cu, Mn, Ni and V.

3.5. Improvement in Flue Gas Cleaning System

To further improve the performance of the cleaning system and reduce pollutant emissions, system optimization or retrofit should be adopted. Considering operation optimization, the air leakage rate, which is one of the main performance indices of flue gas cleaning equipment, was calculated by testing the flue gas volume at the inlet and outlet of the equipment [36], in this case at the rotating spray reaction towers and the bag dust collector. In our study, the test results showed that the air leakage rates of the rotating spray reaction tower and bag dust collector were 4.33% and 3.56%, respectively, which are somewhat higher than the designed value (2.00%). The performance of cleaning equipment is seriously affected if the air leakage rate is too large. Excessive cold air results in the local flue gas temperature falling below the dew point temperature, causing ash deposits, dew condensation, and corrosion, so measures should be taken to reduce the air leakage rate of equipment. To further reduce the emission concentrations of acid gases and PM, wet scrubbing and wet dust removal technologies can be adopted to meet the increasingly stringent environmental emission standards.

A wet scrubbing device is usually arranged behind the bag dust collector. The flue gas from the bag dust collector enters the wet scrubber tower through the bottom, the flue gas flows up and the alkaline solution (usually Ca(OH)₂ or NaOH solution) flows down, so counter flow is formed, promoting the heat and mass transfer between the flue gas and alkaline solution, and the acid gas (SOx, HCl, HF, etc.) is removed through neutral reaction. The purified flue gas enters the downstream equipment after being defogged by the top defogger. The wet dust removal device is located behind the wet scrubber tower, in which the dusty gas can fully contact the water, and the PM in the flue gas can be separated from airflow by liquid wetting and adsorption as well as water surface tension and the static electric field. The simple flow chart of a modified flue gas purification system is shown in Figure 3.



Figure 3. Simple flow chart of a modified flue gas purification system.

4. Conclusions and Suggestions

The flue gas cleaning system in a WTE plant with FGR was investigated, and the main technologies and devices to control the emission concentrations of pollutants were introduced. The measured emission concentrations of pollutants were compared with the limits in legislative standards. The emission of NOx can be effectively reduced by adopting FGR technology, and the emission concentration of NOx can meet the international standards only by using FGR. The acid gases were significantly reduced, but the concentration of HCl was higher than the EC emission limit and the emission limit of the plant, although it may meet the CNC emission limit. The concentrations of heavy metals were low, whereas the concentration of PM was beyond the discharge limits at home and abroad.

As the environmental emission standards become more stringent, the system optimization or retrofit of the flue gas cleaning system should be adopted. The wet scrubbing and wet dust removal technologies proposed may further reduce emission concentrations to better achieve the goal of near-zero-emission.

Author Contributions: Conceptualization, W.W. and J.L. (Jun Liu); methodology, K.X.; validation, W.W. and J.L. (Jun Liu); formal analysis, Z.Z.; investigation, J.L. (Jisheng Long) and S.T.; resources, J.L. (Jisheng Long) and Q.M.; data curation, K.X.; writing—original draft preparation, W.W.; writing—review and editing, J.L. (Jun Liu); visualization, Z.Z.; supervision, W.W.; project administration, Q.M.; funding acquisition, W.W. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Project of Central Plains Science and Technology Innovation Leading Talents of Henan Province, grant number 224200510022.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

Appendix A

Table A1. Full names of all abbreviations.

Full Names	Abbreviation	Full Names	Abbreviation	Full Names	Abbreviation	Full Names	Abbreviation
nitrogen oxide	NOx	Carbon dioxide	CO ₂	Cadmium	Cd	Cuprum	Cu
sulfur dioxide	SO ₂	Nitric oxide	NO	Titanium	Ti	Manganese	Mn
hydrogen chloride	HCl	Calcium hydroxide	Ca(OH) ₂	Lead	Pb	Nickel	Ni
hydrogen fluoride	HF	Sodium bicarbonate	NaHCO ₃	Chromium	Cr	Vanadium	V
methane	CH_4	Hydrogen peroxide	H_2O_2	Antimony	Sb	Mercury	Hg
oxygen	O ₂	Nitric acid	HNO ₃	Arsenic	As		
carbon monoxide	СО	Sodium hydroxide	NaOH	Cobalt	Co		

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