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COMBUSTING FUEL FORMED FROM WASTE.
REDUCTION IN EMISSION OF CHROMIUM, NICKEL AND LEAD

Thermal waste treatment processes dramatically reduce their volume and mass. Unfortunately, they also cause environmental nuisance associated with emissions of harmful substances. The emissions of toxic heavy metals into the air are of particular interest. Low effectiveness of dust extraction equipment for submicron size particles does not provide ecological safety. The paper presents the results of heavy metal (chromium, nickel and lead) emissions from fuel formed from waste combustion. It presents the possibility of reducing that emission by binding heavy metals in the residue scraper. As immobilising metal additives, substances such as: V_2O_5 and $Na_2B_4O_7$ were used. Obtained reduction of heavy metal emission was of up to tens percent. In a series of studies, where in addition to $Na_2B_4O_7$ and V_2O_5, $CaCO_3$ was added, an additional effect of metal retention in the residue scraper was obtained. Chalk binds chlorine released from the fuel combustible material, which in turn prevents the transformation of heavy metals to form chlorides. Binding of hydrogen chloride from the gas atmosphere of the boiler to calcium chloride, eliminates the ability to create heavy metal chlorides. Addition of chalk reduces also its HCl emission and thus minimizes corrosion of the boiler components.

1. INTRODUCTION

The recovery of heat generated in the process and its effective use are slowly becoming mandatory. Functioning of waste incineration installations as power plants, heating plants or heating power plants is subject to local opportunities and needs for the transfer and use of forms of produced energy. Implementation of thermal waste treatment must be safe for the environment, cannot constitute a toxicological hazard. This in turn is a condition for the atmosphere protection and the establishment of appropriate legal regulations.

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Disposal is required by a constantly increasing amount of generated waste, and the EU enforced Poland to increase the share of sorted waste fractions that will be subject to recycling processes, and in case of combustible waste – energetical recycling. Thus, an increase in the production of fuels formed from waste will take place. This means that the waste will be treated as raw materials, and their combustible components will to some extent substitute conventional fuels.

Heavy metals, as they are present in all types of waste in different concentrations and various forms, pose a serious problem of environmental contamination. In the case of their emission to the atmosphere during thermal utilization, they can adsorb on the surface of dust, fly ash, or exist as individual particles. The intensity of evaporation of metals and their compounds in the furnace, apart from the tension of the vapour, depends on many complex and linked factors such as: temperature, oxidizing or reducing conditions and the presence of halogens.

Heavy metals are typically found in the waste as inorganic compounds. Many of these compounds are non-active substances during the combustion process and leave the furnace unchanged. Then the metals are in the residual ash. Heavy metals leave waste incineration systems also in other ways [1–4]. Fly ash trapped in equipment for heat recovery and flue gas cleaning contains heavy metals. Fractions of metals originally present in the waste can also be found in exhaust gases emitted by the systems.

Some metals and like-metal compounds present in waste materials are volatile and are a subject to evaporation in conditions similar to those that occur during the incineration of waste [5]. The high temperature of combustion causes some trace amounts of various metals present in the waste to evaporate. Their steam penetrates to the exhaust gases, which takes them through the furnace system. During cooling of the exhaust gas, vapor condensates in two ways – a uniform (homogeneous) to create new particles and mixed (heterogeneous) adsorbing on the surface of ash particles. Homogeneous condensation leads to the formation of particles with a diameter of less than 1 mm. Heterogeneous condensation depends on the relation of size of the outer ash surface to the weight of the ash particles. Small particles have a greater concentration of volatile metal than the original waste. The concentration of metals such as silver, cadmium, lead and antimony in the particles emitted from combustion appliances is from 30 to 100 times greater than would be expected if this were not the case of the evaporation process. The emission of metals increases with respect to the relation of homogeneous to heterogeneous condensation [5].

Metals in combustible materials can react in installations for the incineration of waste. These reactions result in the reduction of metal oxides. Created mixtures often evaporate more easily than the original substances. There are two main ways for metal to leave the waste incineration installation. One is the evaporation of metal at some points in combustion system, the other is taking the particles containing non volatile metals. Volatility of metals is the main characteristic determining the behavior of metal in waste incineration. Volatility of metals is characterized by the temperature volatility.

Metal volatility temperature is the temperature at which the actual metal vapor pressure is 0.1 Pa. When metal has such a vapor pressure, the evaporation can occur. It was examined that more than 30% of volatile metals, including 10% of metals at volatility temperature above 1143 K are not intercepted. The temperature increase and the consequent increase in gas pressure cause the increase in metal emissions. Vapor pressure of all metals increases rapidly with increasing temperature. Tenfold pressure increase is a result of temperature increase of only 20 K. The vapor pressures for each of the metal adopt values from a very wide range. However, typically the values of these pressures are very small (less than 0.1 Pa) and only small amounts of various metals may evaporate. Significant influence on the vapor pressure of many metals (caused by formation of metal chlorides) has also the amount of chlorine contained in the waste. Chlorides are more volatile than the corresponding metal oxides or free metals. The effective vapor pressure of many metals increases with increasing concentration of chlorine and a large quantity of metals is converted into their chlorides [6]. Sørum et al. [7, 8] studied the volatility of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn in the incineration of municipal waste. They found that the mobility of metals is affected not only by the composition of the waste undergoing thermal transformation but also by the process parameters. The volatile behavior of Cd, Cr, Hg and Pb shows no significant influence, while As, Cu, Ni and Zn are strongly influenced by one or more of the parameters, temperature, fuel/air and chlorine/metal ratios. The problem of heavy metal emissions from fuel combustion, biomass and sewage sludge occupies many researchers [9–12]. Amand and Leckner [13] analysis shows the metal emissions from co-combustion of sewage sludge and coal/wood in fluidized bed. Distribution of ashes between bottom and fly ash is typical of combustion fluidized bed CFB, about 30/70% but in some cases Hg and Cd were only in the fly ash. Chinese scientists [14] studied effectiveness of sorbents on emission reduction of cadmium into the atmosphere during combustion of waste. As sorbent they used calcium oxide (CaO), alumina (Al_2O_3) and silicon dioxide (SiO_2). CaO sorbent proved to be weak in the binding of cadmium. Waste was incinerated at temperatures from 1003 K to 1303 K. When blowing into the furnace, sorbent performance of Al_2O_3 and SiO_2 reached 80%. Studies on the effects of sulfur compounds on the binding of Cd by the sorbents were also conducted. The presence of sulfur as elemental sulfur and Na_2S improved the performance of sorbents. Sulfur in the form of Na_2SO_3 and Na_2SO_4 resulted in increased Cd concentration in the axhausted gases and lower efficiency of binding of Cd by the sorbents. Folgueras et al. [15, 16] investigated the effect of temperature (up to 1373 K, with low heating rates, ca. 10 K/min) on the release of trace elements from blends of coals and sewage sludge in the laboratory furnace; as expected, vaporization increased with the temperature for a number of elements. From the correlation of the volatilization extents with the amount of Cl, Ca/S and other inorganic compounds in the blends, they concluded that chlorine enhances vaporization, whereas the interaction of trace elements with the major ash compounds favours retention.

High temperature processes have a major impact on the formation and reduction of heavy metal compounds. It is connected with changes in free energy and vitality of thermal dissociation under the influence of temperature.

Metal oxide reduction reactions occur at elevated temperatures. Reducing agents present in the processes of reduction of metal oxides are: carbon, carbon monoxide, silicon, magnesium, aluminum. At a temperature of 1173 K, metal oxides form a series, according to their decreasing reductivity: Cu_2O, PbO, NiO, CdO, SnO_2, Fe_3O_4, FeO, ZnO, Cr_2O_3, MnO. As a general rule determining the effect of temperature on the reaction rate, it can be seen that the higher the temperature, the faster the reduction is.

The melting point of various metals can be an approximate indication of their relative volatility. Volatility of individual metals depends on the type of compounds in which the metal occurs. The melting temperature of several metals and their compounds is given in Table 1.

Table 1

Melting points of some metals and their compounds [K]

Metal	Element	Chloride	Sulfate	Sulfide	Oxide
Cd	594	841	1273	2023	1773
Cr	2148	1097 ($CrCl_3$) 177 (CrO_2Cl_2)	373	1823	2539
Cu	1356	893 ($CuCl_2$) 703 ($CuCl$)	decomposes at $T > 473$	decomposes at $T > 493$	1599
Ni	1728	1274	1121	1070	2257
Pb	601	774 ($PbCl_2$) 258 ($PbCl_4$)	1443	1387	1159
Zn	693	556	873	1293	2248

Metals contained in the materials to be combusted can undergo reactions with these substances in the plants. There are two types of reactions. For the former type, processes occur when in the materials, next to metals, organic substances are located. A very common trigger of these reactions is chlorine. The other group of reactions occurs as a result of high temperature, leading to the generation of pollution at the place of waste incineration. The reactions generally come down to release oxygen from metal oxides. The resulting new compounds are more volatile than the original ones. Simultaneously, fumes coming from waste, in the zone of low temperatures and high concentrations of oxygen, undergo following reactions leading back to its original, more complex and condensed form. Here occur two forms of condensation – homogeneous and heterogeneous ones.

The aim of this study was to determine the mobility of metals Cr, Ni, Pb in the process of waste incineration and fuels formed from waste. To reduce the elevation of metals from the combustion chamber in a stream of exhaust gases, fuel additives were

used in the form of divanadium pentaoxide (V_2O_5) and borax ($Na_2B_4O_7$) and their mixtures with calcium carbonate ($CaCO_3$). The choice of additives was determined by their characteristics. In the temperature of the fuel combustion process, divanadium pentoxide forms vanadates with heavy metals – stable and thermal persistent forms. Borax is a flux, which immobilizes metals in the residue stage, whereas $CaCO_3$ bounds chlorine, which limits the possibility of volatile heavy metal chlorides formation. Understanding the behavior of metals in high temperature processes and the ability to increase their immobilization in the residue scraper, will limit the emissions of heavy metals along with dust and fly ash. This type of metal emissions is the most difficult to eliminate, and the most dangerous and disruptive to the environment.

2. EXPERIMENTAL

The study was conducted in a boiler with a capacity of 225 kW with a grate retort (Fig. 1).
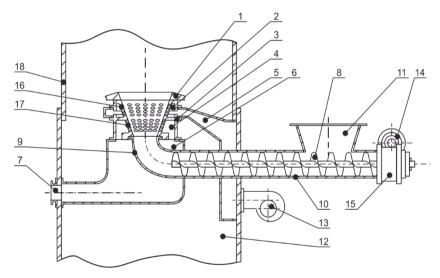

Fig. 1. Scheme of the grate: 1 – the upper part of the grate boiler, 2 – air combustion chamber, 3 – housing bottom grate boiler – trough retort, 4 – gas reversing chamber, 5 – combustion air supply duct, 6 – reversing exhaust duct, 7 – gas inlet reversible, 8 – fuel feeding, 9 – knee fuel feeder, 10 – fuel channel, 11 – fuel tank, 12 – the bottom part of the boiler, 13 – combustion air blower fan, 14 – electric motor drive fuel feeder, 15 – gear reducer drive tray fuel, 16 – upper side wall of the trough retort air blast nozzles, 17 – the bottom side wall of the trough retort gas blowing nozzles reversing, 18 – the furnace casing

Research samples weighed 30 kg. Immobilising additives – V_2O_5 and $Na_2B_4O_7$ – were dozed in amounts of 2% to the weight of the charge (waste rubber and fuel Ecomat). To Pas-r and Pas-i fuel were added: 2% of V_2O_5 and 2% of additive mixtures: and

2% of V_2O_5 + 2% of $CaCO_3$ and 2% of $Na_2B_4O_7$ + 2% of $CaCO_3$. Samples of the fuels and fuel additives were burned in the boiler with retort grate (Fig. 1). Fuel was mechanically fed onto the retort grate from the tray with a spiral transporter. The application process was taking place steadily. Combustion chamber temperature was 1203 K. Concentrations of carbon monoxide CO in the exhaust gases of all samples during the combustion were within the scope of 500–1000 ppm. Excess air ratio λ was 2.3–2.6. Emissions of heavy metals (Cr, Ni, Pb) with the exhaust gases was determined by determining their amount in incombustible residue scraper. For this purpose, after finishing each combustion trial the ash was removed. The ashes were homogenized, where afterwards the concentration of hevy metals was determined. Each combustion test was repeated three times, then the three determinations were made of metal in the ashes.

The study involved the following waste and fuel formed from waste: waste rubber, fuel Ecomat formed from selected combustible fraction of municipal waste, fuel Pas-r and Pas-i formed from industrial waste. The fuel Ecomat is a separated and appropriately processed combustible fraction of municipal and municipal-like waste of defined physicochemical properties (waste was produced by a Swiss-Spanish-German company ECOMAT, located in Moudon in Switzerland).

Fuel Pas-i (produced by SITA STAROL in Chorzów, Poland, SITA SUEZ Group) was formed on the basis of waste sawdust from carpentry and furniture plants. Sawdust was contaminated by various substances to impregnate wood such as paints, varnishes and adhesives. The composition of the fuel Pas-i also included waste petroleum substances, mainly in the form of used oils. Fuel Pas-r was formed from the municipal solid waste such as waste furniture (they included such materials as wood, textiles, plastics, sponge), packaging waste, plastic, wood and cardboard. SITA STAROL company produces fuel Pas-r in a Finnish installation BMH.

Testing of fuel and the concentration of heavy metals. For the preparation of representative samples, fuels were collected in accordance with the standards of solid sampling, and then crushed and homogenized. According to Polish Standards fuel properties of waste and fuels formed from waste were determined: moisture content (ISO 9029:2005), flammable and nonflammable parts (ISO 1171:2002), heat of combustion (PN-ISO 1928:2002), the elemental composition of flammable substance. Coal, hydrogen, nitrogen and sulfur were determined using an elemental analyzer CHNS model 2400 Series II, by Perkin Elmer, chlorine was measured according to ISO 587/2000.

To determine the concentrations of heavy metals (Cr, Ni, Pb) mineralizates were prepared. Fuels were mineralized by the wet method in the microwave mineralizer Speed Four by Berghoff Products + Instruments GmbH. Concentrations were measured by the flame method of spectrometric atomic absorption (FAAS – Spectra AA 880 Varian with deuterium background correction).

3. RESULTS AND DISCUSSION

In order to eliminate big errors, each determination was repeated three times, and the table provides only average values. The results of determinations of fuel properties and concentrations of heavy metals in the fuels are shown in Table 2.

Table 2

Fuel properties and content of heavy metals Cr, Ni, Pb in fuels

Parameter	Kind of fuel			
	Rubber waste	Ecomat fuel	Pas-i fuel	Pasrr fuel
Humidity, %	0.9	19.4	13.5	5.8
Combustible parts, % dw	89.9	72.6	78.7	80.7
Ash, %	10.2	27.4	21.3	19.3
Heat of combustion, MJ/kg dw	32.8	16.7	19.2	20.1
Elemental composition (dry ash, free state – daf)				
C, %	85.0	53.8	76.9	66.7
H, %	7.5	8.3	6.9	7.6
N, %	3.9	0.9	5.1	3.5
S, %	1.6	1.2	0.5	1.2
Cl, %, %	0.0	0.9	0.4	0.5
O and others	2.0	35.8	10.2	20.5
Heavy metals				
Cr, ppm dw	48	238	61	73
Ni, ppm dw	57	44	97	242
Pb, ppm dw	47	159	341	359

The properties of given fuels do not depend only on composition of initial waste. Considerable influence have mainly: kind of separated combustible components, method and degree of processing and addition of auxiliary and stabilising substances. Important is also the method of forming of fuel mixtures, their homogenisation, as well as their final form (flakes, pellets or briquettes). The properties of fuels made of waste are contained in German Standards DIN, in regulations issued by the International Organization for Standardization (ISO). Fuels formed from waste are classified according to the Solid Recovered Fuel (SRF) classification system, elaborated by the European Committee for Standardization [17]. The system classifies the Ecomat fuel and other fuels from municipal waste as *solid fuel produced from waste other than hazardous, used to recover energy in combusting or co-combusting installations and meeting the requirements of the classification and specification mentioned in CEN/ TS 15359*.

Moisture content in the fuel significantly affects the caloric properties of fuel. The scale of the problems associated with fuel storage, transport and feed into the combustion chamber, the amount of gas produced, the size of the outlet losses and efficiency of the furnace.

The study of elemental composition of the combustible substance of fuels formed from waste shows that the fuel contains a large share of the carbon from 53.8 to 85% of dry ash-free mass, reflecting the possibility of efficient combustion. In the process of combustion of fuels formed from waste, the addition of alkali metal compounds is advisable, and this is due to the high share of chlorine in these fuels (Ecomat – 0.9% dry wt, Pas-i – 0.4% dry wt, Pas-r – 0.5% dry wt). It should be noted that the alkalization treatment could slightly affect their fuel properties, as they are characterized by large gram shares of carbon.

Many researchers dealing with waste and fuel combustion point out the risks associated with emissions of trace elements, including heavy metals. Akiko et al. [18] studied 18 heavy metal emissions from the incineration of municipal solid waste (MSW). They presented it in the form of concentration expressed in mg Me/Nm^3. Total emission amounts of heavy metals in Japan were also calculated using the estimated emission factors.

The research on division of Cr, Ni and Pb between solid scraper phase and ash residues from the combustion of fuels was performed in a real object, which is a boiler with a capacity of 225 kW with grate retort. The difference between the amount of metals placed on the grate with the fuel and the amount of residual scraper is the size of their emissions. The results are illustrated in Figs. 2–5.

Cr, Ni and Pb, which are waste pollution, during combustion are taken away from the combustion chamber in a stream of exhaust gases. Chromium and nickel emissions from the combustion of fuels tested were similar. They were observed in the range of 60–80%. Lead passed into the gas phase in amounts ca. 90%. Such a large portion of heavy metals discharged from the combustion chamber results in a threat of atmospheric air pollution with toxic metals. Belevi et al. [19, 20] and Sørum et al. [7] confirmed the transfer of heavy metals from municipal waste (during combustion in the plant) into the gas phase in their studies. Also, the authors of the review [21], on the basis of the cited publications, inform about the emissions of trace elements in the exhaust gas stream. They presented an analysis of behavior and mobility of trace element during coal combustion. Also metal vapors condense on the particles of submicron fly ash and are being carried from the combustion chamber with the combustion gases.

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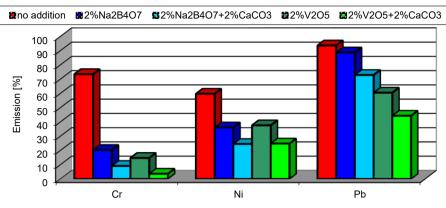
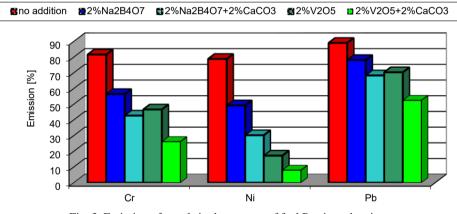Fig. 2. Emission of metals in the process of fuel Ecomat combustion

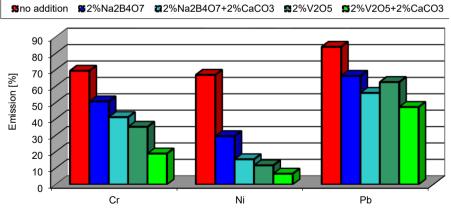Fig. 3. Emission of metals in the process of fuel Pas-i combustion

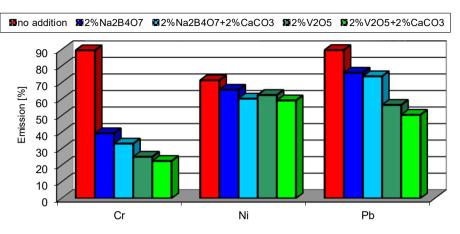Fig. 4. Emission of metals in the process of fuel Pas-r combustion

Fig. 5. Emission of metals in the process of rubber waste combustion

It is possible to immobilize trace elements in bottom ash by using immobilizing additives. Fuel additives in the form of V_2O_5 and $Na_2B_4O_7$ decreased emission of tens of percent. In most cases, a greater immobilization effect in residue scraper was observed in samples of V_2O_5. Immobilizing activity of $Na_2B_4O_7$ and V_2O_5 action amounted to the creation of thermally more stable forms of heavy metals. $Na_2B_4O_7$ serves as flux, V_2O_5 at high temperatures creates vanadates with metals which, being thermally stable, are not mobile and remain in the slag.

$Na_2B_4O_7$ and V_2O_5 added to fuels, immobilized significantly heavy metals in the slag. Their activity was selective with respect to the studied metals. And so Cr emission was limited to the level of 10–20% in research of fuel Ecomat, and 30–45% of the fuel Pas-i, Pas-r and rubber waste. In the process of thermal destruction of rubber , the impact of the above mentioned immobilizing additives, with respect to nickel, reduced its emission by a few percent. However, in the slag formed from the combustion of fuels from waste (Ecomat, Pas-i and Pas-r), borax immobilized Ni by 30% more effectively than the sample without this additive. V_2O_5 reduced emissions of nickel even up to 10% (fuel Pas-r and Pas-i) and 30% (fuel Ecomat).

Lead least subjected to the action of borax in the process of all fuel combustion (reduction of emission by up to 20%). V_2O_5 stopped Pb by 20–30% more effectively than in trials to burn only fuels.

It is worth mentioning that the additives were mixed with the fuel, which is a simple application method. Xu et al. [21] suggest the possibility of using mineral sorbents to remove metals from the gas phase. However, here the application of sorbents was performed by them being blown into the gas space of the combustion chamber.

The presence of chlorine in the waste has a high impact on the volatility of heavy metals. This is confirmed by Belevi and Langmeier [20], stating that not only the higher temperature of the combustion process increases MSW metal emissions but also the

presence of chlorine. So removing the chlorine released (as HCl or Cl_2) from temperature transformed waste, we can influence indirectly the mobility of metals. In this work it was done with chalk ($CaCO_3$). The lime sorbent bound chlorine to form calcium chloride.

In a series of studies, where in addition to $Na_2B_4O_7$ and V_2O_5, $CaCO_3$ was added, an additional effect of metal retention in the residue scraper was obtained. Chalk ($CaCO_3$) binds chlorine released from the fuel combustible material, which in turn prevents the transformation of heavy metals to form chlorides. Chlorides have greater volatility than oxides, resulting in higher emission. Binding of hydrogen chloride from the gas atmosphere of the boiler to calcium chloride, eliminates the ability to create heavy metal chlorides. In the trials, where apart from fuels and immobilizing additives chalk was added, emission of all determined metals (Cr, Ni, Pb) decreased further by a few to several percent.

4. CONLUSIONS

• During the incineration of waste rubber and fuels formed from waste chromium, nickel and lead are leaving the combustion chamber in a stream of exhaust gases. These are the emissions of 60–80%, for lead even about 90%.

• Fuel additives in the form of V_2O_5 and $Na_2B_4O_7$ decreased this emission by tens of percent. In most cases, a greater immobilization effect in residue scraper was observed in samples of V_2O_5.

• Immobilizing activity of $Na_2B_4O_7$ and V_2O_5 action amounted to the creation of thermally more stable forms of heavy metals. $Na_2B_4O_7$ served as flux, V_2O_5 created vanadium.

• $CaCO_3$ binds chlorine contained in the fuel, which indirectly helps to increase the amount of Cr, Ni and Pb in the slag.

• Additives used strongly reduce emissions of Cr, Ni, Pb, addition of chalk reduces also its HCl emission and thus minimizes corrosion of the boiler components.

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