THERMODYNAMIC TOOLS FOR OPTIMISATION OF THE HIGH-TEMPERATURE DECOMPOSITION OF PCB IN THERMAL PLASMA

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Abstract: Methodology using the thermodynamic equilibrium composition calculation to optimise the high-temperature organic waste treatment in thermal plasma was developed. The methodology was demonstrated in the case of high-temperature treatment of polychlorinated biphenyl (PCB) in a thermal plasma reactor using two different working media, air and water vapour. Calculated results obtained in this analysis show composition of combustible and harmful chemical species formed in the PCB oil decomposition process. Optimisation of the composition of the combustibles can suggest the most economical parameters of the process. Toxic substances emissions are one of the limiting factors in the process of optimization. This analysis can be considered as the first step towards defining optimal operating parameters of the pilot-plasma facility for waste treatment from the ecological and economical aspects.

Keywords: Thermal plasma, thermodynamic equilibrium, PCB, optimization, ecological and economical aspects

1. INTRODUCTION

The economic growth of modern society is directly related to the growth in the industrial sector. One of the vital environmental issues that come with industrial growth is waste generation. According to the Waste Framework Directive, waste can be considered hazardous and non-hazardous. Hazardous waste can cause great danger to human health and the environment because of its origin, composition, and concentration of hazardous components. With increasing industrialization, urbanization, and population, the increase in a waste generation has been noted.

Waste volume in the Republic of Serbia is hard to estimate mostly because of the lack of information. It is considered that only around 60% of municipal solid waste is collected in Serbia. In 2015 it was generated 50 884 926 t of waste, while in 2019 that number increased up to 66 565 200 t, which is equivalent to an increase of 30% [1]. The average person generates around 318 kg of waste per year which is equivalent to 0.87 kg per day [2]. According to the Statistical Office of the Republic of Serbia, waste generation divided by sectors is presented in **Fig. 1.** Observed by sections, the highest share of the generated waste is noted in the Mining and quarrying sector with 80.8%, followed by Electricity, gas, steam, and air conditioning supply with 11.3% (**Fig. 1**). Since Serbia is considered a country in transition that is applying to become a member of the European Union, new obligations in waste management will be given. With the adoption of the Law on Waste Management (LWM) and the Law on Packaging and Packaging Waste (LPPW), certain progress has been made in Serbia regarding this problem but it is insufficient.

Even though strict regulations on waste management are in place, primitive disposal methods like open dumping and discharge into rivers have been used in many places in Serbia making landfill disposal the main waste treatment method [1], [3] (**Fig. 2**). This method causes problems regarding releasing toxins and occupying the land. Poor waste management may result in various sanitary and environmental issues such as the risk of explosions in landfill areas and groundwater contamination due to leachate percolation [4]. There are around 3 500 dumping sites in Serbia, out of which only 180 are official communal landfills [2], [5].

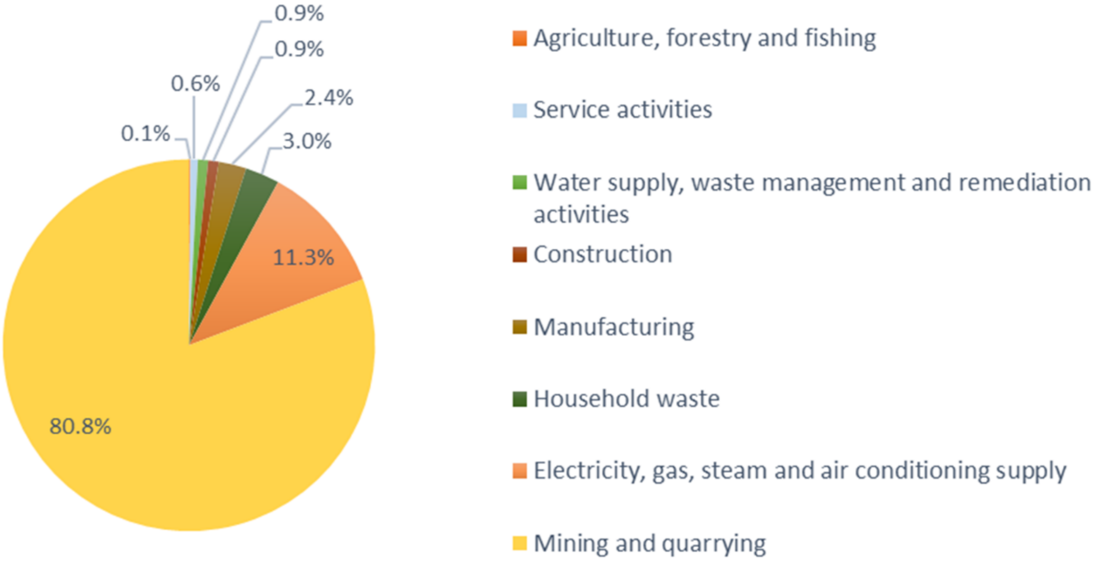

Figure 1: Waste generation by sectors, Serbia, (%), 2019 [1]

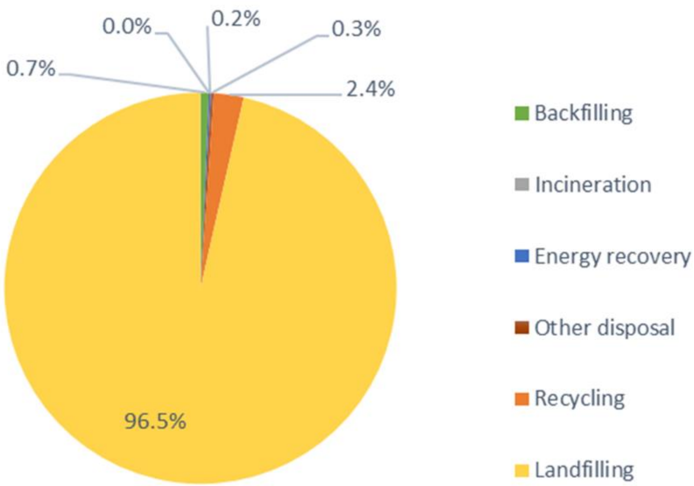

Figure 2: Waste treatment by treatment category, Serbia, (%), 2019 [1]

It is of the highest priority for the government and local authorities to come up with proper waste management technologies that are economically, environmentally, and socially sustainable. One of these technologies is considered to be waste-to-energy (WtE) which is a process of recovering energy in form of electricity, heat, or some alternative fuel from waste. Waste treatment technologies can be sorted into the following categories: thermo-chemical conversion technologies (incineration, pyrolysis, gasification, and liquefaction), biochemical conversion technologies (anaerobic/aerobic digestion and fermentation), and chemical conversion technologies (trans-esterification, etc.) (**Fig. 3**) [6].

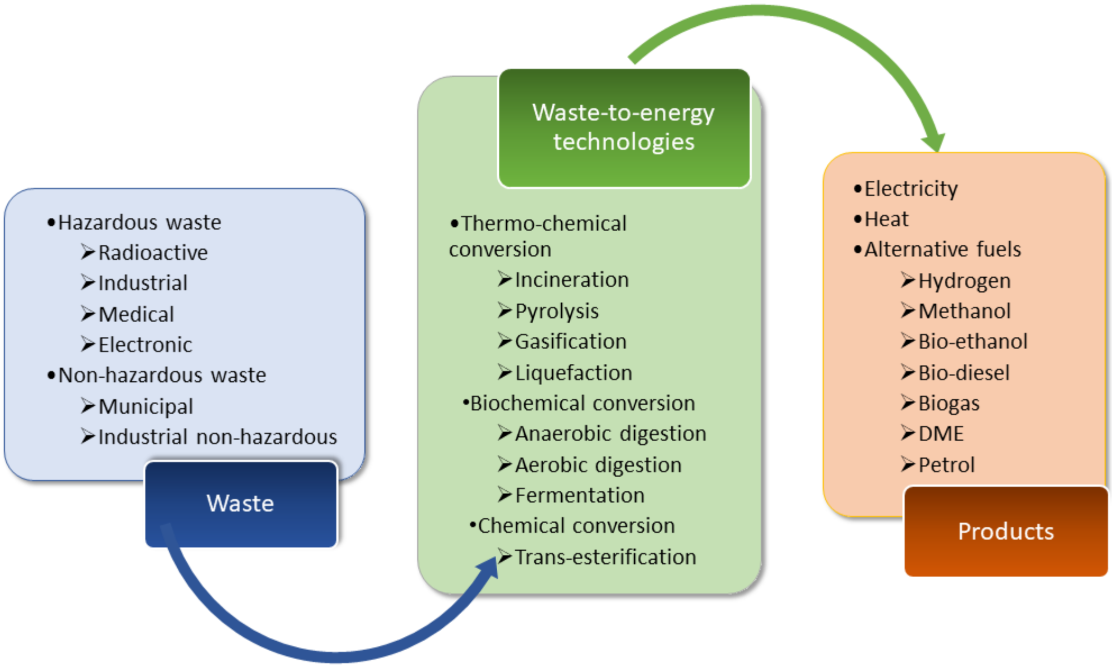

Figure 3: Graphical representation of WtE systems

Incineration is frequently used waste treatment technology and it is performed in the temperature range between 750 and 1000°C. It is divided into three segments: incineration, energy recovery, and air pollution control. Air pollution is a result of the emission of various air pollutants like sulfur, carbon and nitrogen oxides, and dioxins which are a generic term for dibenzo-para-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). Several studies have proven the presence of significant quantities of dioxins in municipal solid waste [7]. The problem also arises when talking about the incineration of hazardous waste such as chemicals, paints and other coatings, pesticides, pharmaceutical products, motor oils, poisons, herbicides, medical waste, etc. The vast majority of the hazardous waste in Serbia comes from the mining and quarrying sector which covered 99% of total generated hazardous waste in 2019 estimated at 15 800 650 t [1]. Incineration of municipal and hazardous waste is one of the main sources of dioxins, together with combustion sources (cement kilns, wood-burning, uncontrolled fires in landfill sites, etc.), industrial sources (pulp and paper mills, metals industry, chemical manufacturing, etc.) and reservoir sources (photochemical processes, accidental source, etc.) [7]. Dioxins are amongst most toxic chemicals on the earth, in particular, 2,3,7,8-tetrachlorodibenzo-para-dioxin (TCDD) and are unintentionally formed during waste incineration by two main pathways: a series of homogenous reactions at high temperatures between 500 and 800 °C and the other one is heterogeneous reactions in the post-combustion zone at low temperatures between 200 and 400 °C [8], [9]. PCDDs and PCDFs were never produced commercially, but are formed as unintentional by-products of waste incineration. On the other hand, PCBs or polychlorinated biphenyls are “dioxin-like” biphenyls and compared to PCDDs and PCDFs, they were synthesized by humans in large quantities from 1940 to 1970 when they were banned. Because of their persistent and bioaccumulative properties, today they can be found in every aspect of the global ecosystem causing environmental and health problems [10]. It can be concluded that in conventional incinerators where waste is combusted in excess air/oxygen at a temperature range between 750 and 1000 °C, not all waste components can be broken down at the molecular level.

Plasma gasification is a proposed technology of waste management. This process completely converts waste into its constituent elements such as carbon, hydrogen, oxygen, etc. which rapidly recombine into carbon monoxide and hydrogen known as syngas. The process offers flexibility, fast process control, and valuable end product, such as syngas which has a significant calorific value and can be further used for power and heat generation. Because of high temperatures (>1300 °C) that are reached during plasma gasification, produced syngas is cleaner compared to the conventional gasification process because tar, char, and dioxins are decomposed, while any inorganic components such as metals and silicates are melted and converted into vitrified slag [11], [12]. Plasma gasification is capable of treating a huge range of waste, independent of particle size, moisture content, and composition [13]. Also, this waste treatment method offers the possibility of extraction of pure hydrogen which is considered as a future alternative to fossil fuels [14]. The main two disadvantages of this process are the high energy consumption necessary for plasma generation and operating challenges regarding extremely high temperatures which ask for high-temperature-resistant materials.

This paper presents developed methodology using the thermodynamic equilibrium composition calculation as a tool for optimization of the high-temperature PCB oil treatment in thermal plasma. Calculated results show composition of combustible and harmful chemical species made by decomposing PCB-oil in the low-temperature plasma with two different working mediums, air and water vapor. This analysis can be considered as the first step towards defining optimal operating parameters of the pilot-plasma facility for waste treatment from the ecological and economical aspects.

2. FEEDSTOCK: PCB WASTE

PCBs used to be commercially produced products such as Aroclor, Clophen, and Phenoclor for a variety of applications like dielectric fluids for capacitors and transformers, heat transfer fluids, etc [15]. The most common examples of waste that is composed of/ contains/is contaminated with PCBs are graphically presented in **Fig. 4**.

According to the results of a detailed inventory of PCB-containing transformers in Serbia, it is estimated that around 7 % (4 480) of transformers and 585 tons of oils from transformers contain PCBs. Also, 5-8 % (4 200) of low voltage transformers are contaminated with PCBs which are residual after cleaning and recharging transformers with mineral oils. Contaminated oils with PCBs from low voltage transformers are estimated at 1 200-1 900 tons [16]. From 2010 to 2013 around 112.17 t of PCB waste was generated in Serbia [17]. Serbia doesn't have an appropriate location for storage and decontamination of this type of waste neither necessary plants for their degradation which just stresses the importance of dealing with this problem [18].

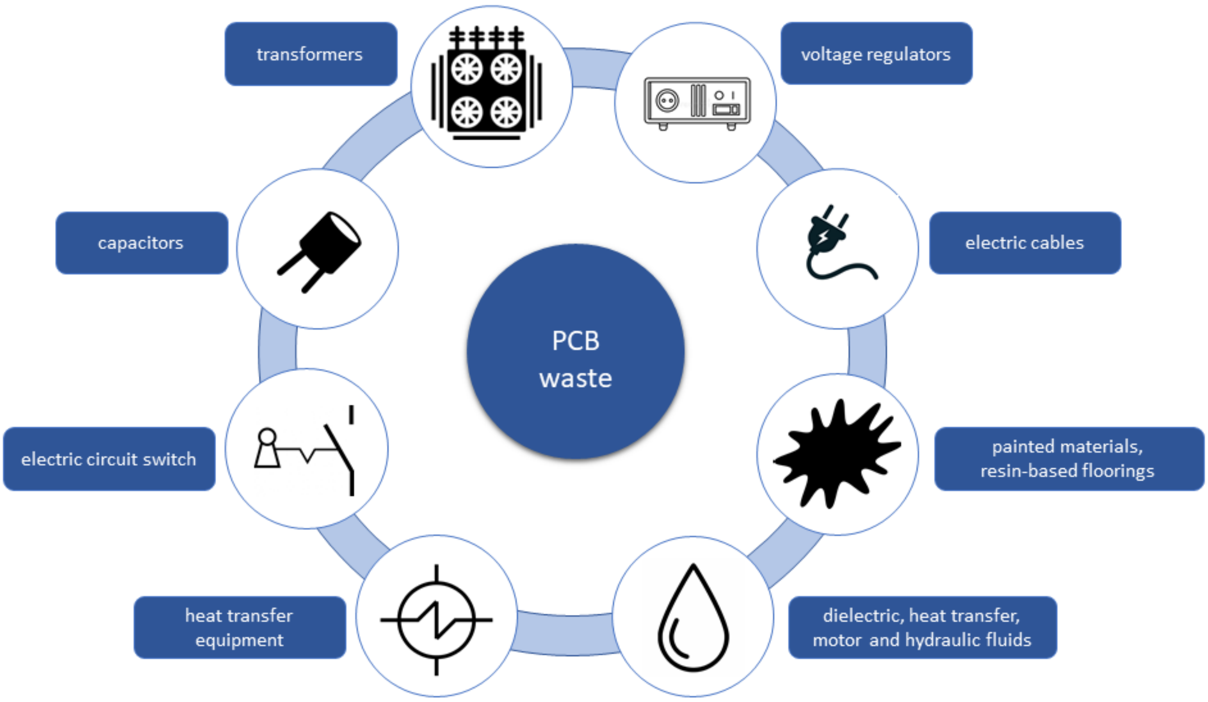

Figure 4: Graphical representation of PCB waste

From the aspect of chemistry, PCBs are synthetic aromatic compounds where hydrogen atoms from biphenyl molecules can be replaced with up to 10 chlorine atoms. The general chemical formula which represents PCBs is $C_{12}H_{10-n}Cl_n$ where n=1-10. Regarding chemical characteristics, PCBs have excellent dielectric properties, are chemically stable, non-flammable, and resistant to thermal and physical degradation. Also, the higher presence of chlorine in PCBs leads to water insolubility and higher chemical solubility. They tend to accumulate in lipid-rich tissues of organisms and the liver. Their bioaccumulative nature and resistance to thermal and physical degradation are responsible for their extremely high concentration in all aspects of the environment as well as in living beings. 12 congeners out of 130 of them which can be found in commercial products are proven to be cancerogenic [19]. Contamination of humans with PCBs causes skin changes, changes in the digestive tract, liver tumors, anemia, reproductive issues, leukemia, and cancer [19], [20].

3. TECHNOLOGY: THERMAL PLASMA GASIFICATION

Conventional gasification transforms solid or liquid waste into useful and convenient gaseous fuel like syngas, which is mainly composed of hydrogen and carbon monoxide, that can be burned to release energy or used for the production of value-added products such as hydrogen. The process involves the breakdown of the organic and inorganic elements into gases (CO, H_2, CH_4, H_2O, CO_2, etc.), liquids (tar), and solids (char). It is performed with the addition of gasifying mediums like air, steam, or oxygen. It operates at relatively low temperatures between 400 and 850°C [21]. Main problems regarding the use of conventional gasification in waste treatment refer to costly air separation units, much longer time to heat up, and products being dirtier compared to plasma gasification because of the presence of tars, chars, and soot. On the other hand, plasma gasification, because of very high temperatures and relatively long residence time for the gas in the gasifier, causes the tars to be cracked and dioxins like PCDDs and PPCDFs to be destroyed. Compared to conventional oxy-fuel flames, thermal plasma has 10-100 times higher power density [22]. Due to the high power density and achieved high temperatures, the small-scale reactor can treat a large amount of waste. The dominant method of generating plasma in waste treatment is using DC electric discharge because it is followed by more stable operation, better control, lower electrode and power consumption, and less noise [23]. The amount of gasifying medium has a major influence on the yield and composition of the product gas. The heating value and the composition of the gas produced in the gasifier depend on the nature and amount of the gasifying medium used. When air is the gasification medium, as is the case for 70% of all gasifiers, the nitrogen in it dilutes the product gas. Also, the formation of nitrogen oxides and cyanide compounds can arise due to the increased content of nitrogen in the air plasma system and achievable high temperature. When pure oxygen from an air separation unit is used, the heating value is higher, but a large amount of energy is spent on separating the oxygen from the air.

Plasma is defined as electrically neutral, partially or fully ionized gas. It consists of particles that are in permanent interaction and can be classified as “light” species, such as electrons and photons, and “heavy” species, such as positive and negative ions, atoms, free radicals, and excited and non-excited molecules [24]. It can be categorized into two groups, low and high-temperature plasma. Further division of low-temperature plasma is on thermal/hot/equilibrium and /non-thermal/cold/non-equilibrium plasma, while high-temperature plasma is stellar plasma. This division is based on the relative energetic levels of “light” particles-electrons, and “heavy” particles of the plasma. Thermal/equilibrium plasma implies that all the species of the plasma, ” light” and “heavy” (ions, electrons, and neutral species, atoms, etc.) retain the same temperature (2 000-20 000 K), meaning that local thermal equilibrium is achieved [25]. Non-thermal/Non-equilibrium plasma is characterized by high electron temperature (10^4 K) and low ion and neutral particles temperature (temperatures as low as room temperature), so local thermal equilibrium is not achieved [26]. From the aspect of pressure, thermal plasma is produced at higher pressure e.g. 10 kPa compared to a low-pressure cold plasma [27]. Thermal plasmas can be generated by different methods such as direct current (DC), alternating current (AC), radio-frequency (FC), and microwave electric discharge, while in the case of cold plasmas some of the possible techniques are glow discharge, gliding arc discharge, etc. (**Fig. 5**).

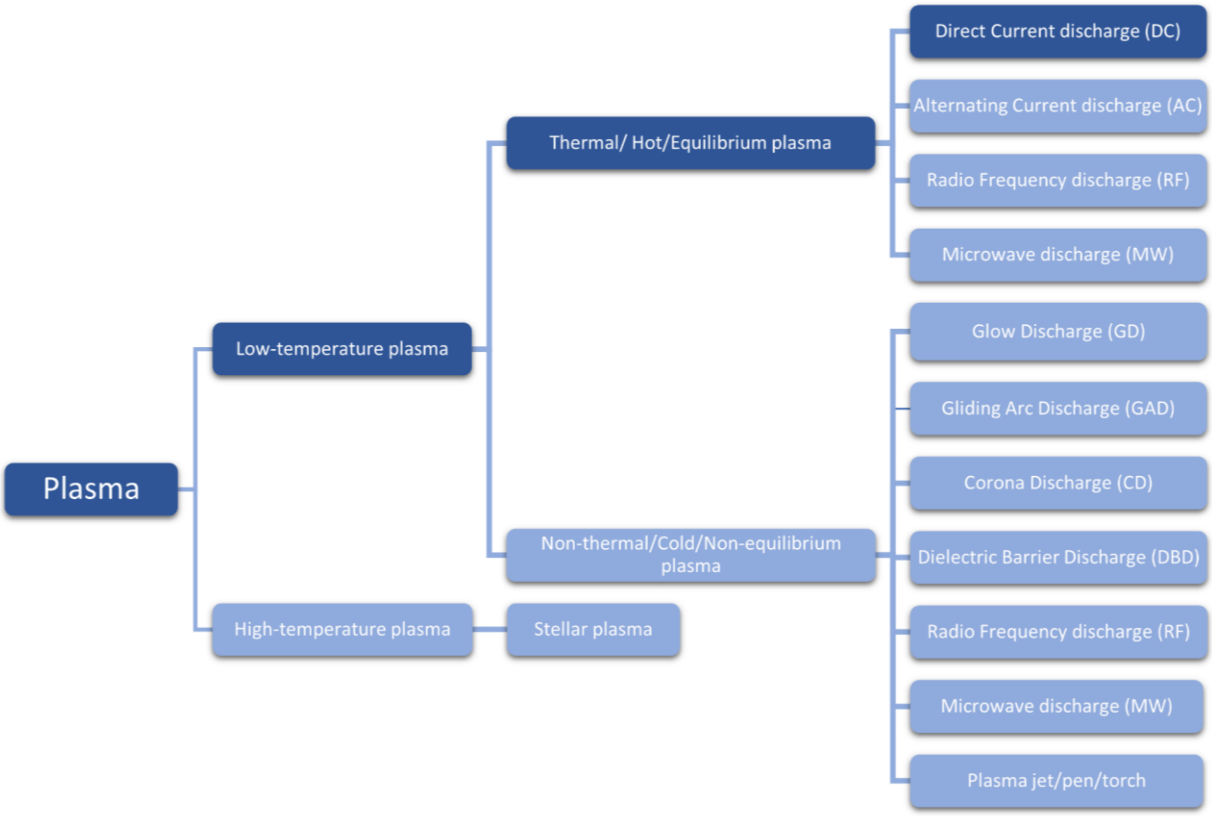

Figure 5: Classification of plasmas [26], [28]–[31]

4. THERMODYNAMIC EQUILIBRIUM MODEL AND ENTRY DATA

Mathematical modeling of plasma gasification process can provide an analysis of finding optimum operating and design parameters and can be divided into four model types: thermodynamic equilibrium, kinetic, computational fluid dynamics (CFD), and artificial neural network model [6]. This paper will present the thermodynamic equilibrium model which can predict the maximum achievable yield of desired products, H_2 and CO, if reactants, waste components, are left for an infinite time to react and reach equilibrium yield. Equilibrium models can further be divided into two categories: stoichiometric and nonstoichiometric. The main difference is that for the first group, one must know the reaction mechanism of the process. Here, it will be presented a non-stoichiometric equilibrium model that is based on the minimum of the total Gibbs free energy and the only input data are the compositions of the feedstock and gasifying medium.

Two systems will be analyzed, PCB-water and PCB-air in the temperature range between 1 000 and 6 000 K. The treated medium or feedstock is PCB oil (waste), tetrachlorinated biphenyl with the chemical formula $C_{12}H_6Cl_4$. The gasifying mediums are water and air, where water consists of H_2O molecules, while air is a mixture of the following gasses: 78 wt% N_2, 21 wt% O_2, and 1 wt% Ar.

It is assumed for analysis that the mass flow rate of gasifying mediums, water ($\dot{m}_w$) and air ($\dot{m}_a$); is constant and equal to 108 kg/h, the ratio of the mass flow rates of feedstock and gasifying medium ($f_a = \dot{m}_{PCB} / \dot{m}_a$ and $f_a = \dot{m}_{PCB} / \dot{m}_w$) obtains the values 0.50, 0.75, 1.00 and 1.50.

These two assumptions give the possibility to determine the mass flow rate of PCB oil as:

$$\dot{m}_{PCB} = f_a \cdot \dot{m}_w = f_a \cdot \dot{m}_a \tag{1}$$

The number of moles of each element that PCB oil consists of (C, H, and Cl) are defined as:

$$\dot{n}_{PCB,i} = \frac{\dot{m}_{PCB} n_i}{M_{PCB}} = \frac{\dot{m}_{PCB} n_i}{M_{C_{12}H_6Cl_4}} \tag{2}$$

where n_i is the number of atoms of the i^{th} type in a molecule and $M_{C_{12}H_6Cl_4}$ is a molar mass of PCB oil.

The number of moles of each element that water consists of (H and O) are defined as:

$$\dot{n}_{w,i} = \frac{\dot{m}_w n_i}{M_{H_2O}} \tag{3}$$

where n_i is the number of atoms of the i^{th} type in a molecule and M_{H_2O} is a molar mass of water.

The number of moles of each element that air consists of (N, O, and Ar) is defined as:

$$\dot{n}_{a,i} = \frac{\dot{m}_a g_i n_i}{M_a} \tag{4}$$

where n_i is the number of atoms of the i^{th} type in a molecule, M_a is a molar mass of air, and g_i is the mass fraction of the i^{th} type in a molecule of air.

Entry data into the model are the molar fractions of all chemical elements entering the gasifier in a unit of a time, defined as:

$$w_i = \frac{\dot{n}_i}{\sum_{i=1}^{j} \dot{n}_i} \tag{5}$$

where *j* is the number of components of the analyzed system and $\dot{n}_i$ the number of elements entering the gasifier in a unit of time (**Table 1**).

Table 1: The molar fractions of the components of the PCB-air and PCB-water systems

Ratio	PCB-air		PCB - water	Ratio		PCB-air	PCB - water
f_a	Molar fractions of the components of the system			f_a	Molar fractions of the components of the system		
0.50	C	1.1932	0.1006	1.00	C	0.2853	0.1699
	H	0.0966	0.5940		H	0.1427	0.5440
	Cl	0.0644	0.0335		Cl	0.0951	0.0566
	O	0.1363	0.2718		O	0.1007	0.2295
	N	0.5063	-		N	0.3739	-
	Ar	0.0032	-		Ar	0.0024	-
	S	-	-		S	-	-
0.75	C	0.2462	0.1382	1.50	C	0.3393	0.2205
	H	0.1231	0.5669		H	0.1696	0.5074
	Cl	0.0821	0.0461		Cl	0.1131	0.0735
	O	0.1158	0.2489		O	0.0798	0.1986
	N	0.4301	-		N	0.2963	-
	Ar	0.0028	-		Ar	0.0019	-
	S	-	-		S	-	-

5. RESULTS AND DISCUSSION

In the following diagrams, it will be presented the formation of possible combustible and harmful chemical species which are the result of the decomposition process of PCB oil with two different gasifying mediums, water and air, in the temperature range between 1 000 and 6 000 K (**Figs. 6-7.**).

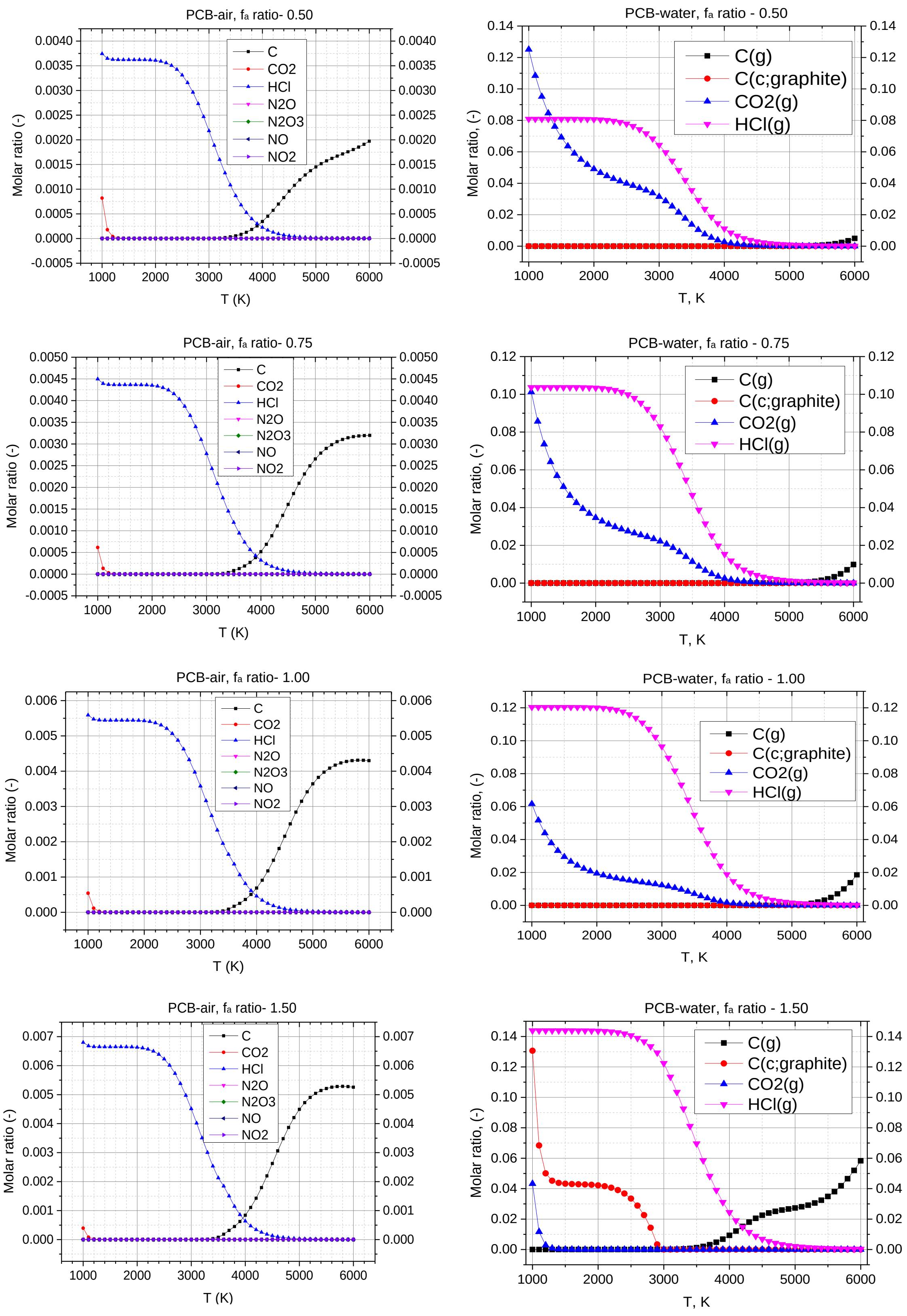

Figure 6: The molar ratios of harmful chemical species in defined systems

The main difference that can be seen in the formation of harmful chemical species during the decomposition of PCB oil with air, compared to the system that involves water is the presence of various nitrogen oxides that come from the air. The harmful species that can be seen in both systems are graphite C (c; graphite), gaseous carbon C(g), carbon dioxide CO_2, and hydrogen chloride HCl(g). Hydrogen chloride presented similar patterns of behavior in both systems. The highest molar ratio of hydrogen chloride is observed at the lower temperatures and with the increase of temperature, its presence is decreasing, while at the 6 000 K it is concluded that no hydrogen chloride is left. It is important to stress that while the highest molar ratio of hydrogen chloride in the system PCB-air is around 0.0045, in PCB-water that value is 0.10. Carbon dioxide showed different patterns - the higher molar ratio of carbon dioxide is formed in PCB-water with the maximum value of molar ratio at the lowest temperature of 1000 K. Carbon dioxide remains in products until the system reaches 4 000 K, after that its presence is negligible. In PCB-air, a negligible molar ratio of carbon dioxide is present at 1 000 K and below 1 500 K there is no carbon dioxide left. Regarding gaseous carbon, it is observed the increase of its molar ratio with the increase of temperature reaching the highest molar ratio at 6 000 K. The significant molar ratios of gaseous carbon start above 3 500 K. Comparing the values for different systems, it can be seen that the PCB-water system has significantly higher values of gaseous carbon. With the increase of f_a ratio, molar ratios of all harmful products increase in both systems.

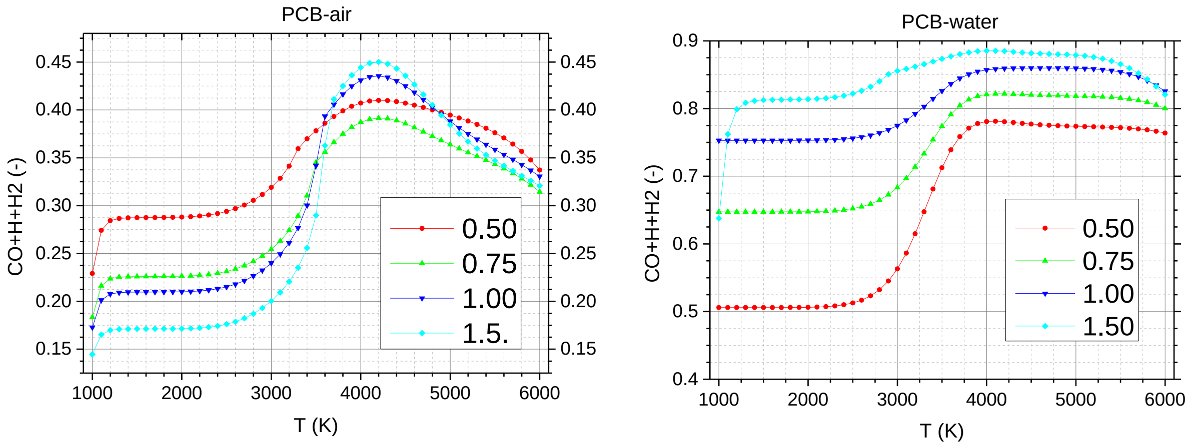

Figure 7: The molar ratio of combustible chemical species in defined systems

Analyzing the previous diagrams it can be seen that the highest molar ratio of combustible products C, H, and H_2 is around 4 000 K for both systems. The difference is in the behavior of species at higher temperatures than 4 000 K-while in the PCB-air system the molar ratio of combustible species is decreasing, in the PCB-water system the further increase in temperature doesn't significantly change the molar ratio of combustibles.

7. CONCLUSION

The research subject of this paper is the process of decomposition of polychlorinated biphenyls, the so-called PCB compounds, using low-temperature arc water or air plasma. The model is made in special software whereby entering molar fractions of the corresponding system's components it is possible to determine the equilibrium composition of harmful and combustible chemical species that can occur in the range of 1 000-6 000 K. The model is based on the principle of minimum Gibbs free energy. The treated material is PCB oil presented with the chemical formula $C_{12}H_6Cl_4$. The gasifying mediums which were considered were air and water steam. The paper analyses the change in molar ratios of harmful and combustible chemical species that can be formed during the decomposition process with the increase in temperature. Observed harmful species were gaseous and solid carbon, carbon dioxide, and hydrogen chloride. Also, in the PCB-air system, there were nitrogen oxides present which come from the air and are considered as harmful species, as well. Regarding combustible species, they are chemical species that have energy value and are part of syngas. The molar ratio of $C+H+H_2$ was analyzed. As a result of previously discussed results, it can be concluded that while the higher temperatures benefit the increase of the molar ratio of combustible species, they also cause the increase of the molar ratio of gaseous carbon as a harmful product. In the temperature range of 3 000-4 000 K, there is a negligible molar ratio of all the other harmful species, that is also the temperature range where it can be observed that the molar ratio of combustible species reached its maximum, and doesn't change significantly with the increase of temperature. This proposes the conclusion, that from the previously discussed aspects, the temperature range 3 000-4 000 K is the most beneficial one for decomposing the PCB oils. Comparing the gasifying mediums, air and water, it is important to say that while air causes the formation of nitrogen oxides, water yields significantly higher molar ratios of combustible as well as harmful species.

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