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INFLUENCE OF RUBBER POWDER MASS FLOW RATE ON PLASMA PYROLYSIS

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Abstract

The paper describes an experimental examination of the thermal utilization of used rubber. The research was carried out to examine the influence of rubber powder mass flow rate on the plasma pyrolysis of rubber. An arc plasma generator was applied. Ar and a mixture of Ar and H₂ were used as plasma gases. The composition of gaseous products was analyzed by infrared absorption spectroscopy. All of the rubber introduced to the plasma jet was decomposed. The outgoing gas did not contain any toxic chemical compounds such as NO_x or HCN.

Introduction

Even though, that there have been applied numerous methods to utilize rubber wastes (LIANG et al. 2020, SOVJAK et al. 2019, HUILIN et al. 2019) the problem of used rubber has not been solved satisfactorily yet (WOJCIECHOWSKI, DOLIŃSKI 2014). Almost every landfill in Poland has a special spot covered with used tires well beyond the landfill capacity. This situation seems to be proved by devastating fires of the landfills in 2018 in Skawina, Siemianowice Śląskie,

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Trzebinia (PAWŁOWSKA 2018) and many others. Criminal issue of setting fires is involved to cover cases of used tires illegally brought to Poland. The fires are set out to cut investigations and lawsuits. The examples multiply in the whole country.

In year 2018 in Poland almost 190 thousand (BIAŁASZ 2018) tons of tires were brought out to the domestic market (Fig. 1). The weigh of used tires annually seems to grow almost linearly. It has to be highlighted that used tires make up approximately 75% of all the mass of rubber waste. Additionally, close 20% of tires get worn out during regular exploitation (BIAŁASZ 2018).

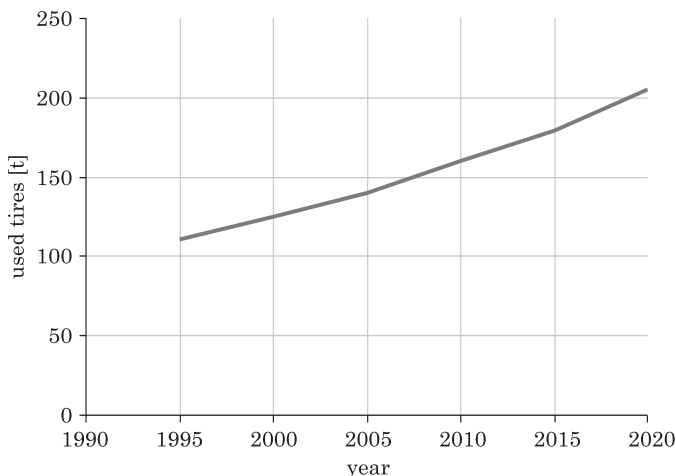


Fig. 1. Estimated annual amount of used tires in Poland

Thermal utilization of rubber waste can be not only a solution to the environmental problem but it might be a profitable activity as well. Decomposition of rubber in the plasma jet may be a source of hydrocarbons and syngas which exquisitely can reduce costs of utilization process of rubber by using those gases as fuel. Moreover, plasma pyrolysis of rubber might reduce amount of side products comparing to other thermal utilization methods of rubber waste.

Amongst the physical and thermal methods of utilization of rubber waste plasma pyrolysis brings a special attention and interest. That kind of pyrolysis is applied more seldom than thermal pyrolysis (maximum process temperature – 1,000 K). Plasma pyrolysis has much bigger potential though (WIELGOSIŃSKI 2011, TANG, HUANG 2004). Plasma pyrolysis offers very high process temperature, which can range from 6,000 K to 18,000 K (MAJEWSKI, DĘBSKI 2012). So high process temperature provides plasma pyrolysis with huge advantage over thermal pyrolysis or ordinary incineration, especially considering decomposition of any substance or purity of gaseous products of rubber thermal utilization. That high temperature is able to decompose almost every substance and material.

The process of plasma pyrolysis has already been examined (SZUSZKIEWICZ et al. 2001) considering plasmatron power influence on the course of plasma pyrolysis of rubber. However, a dependence of rubber powder mass flow rate on plasma pyrolysis of rubber has not been investigated yet.

The aim of the research described in the paper was to determine the influence of amount of rubber delivered to the plasma jet on the composition of the gaseous products in the process during plasma pyrolysis. The secondary aim of the research was to investigate the influence of mass of rubber on concentration changes of the gaseous products as a potential source of energy.

Experimental set-up

The research on plasma pyrolysis of rubber has been carried out using a plasmatron (Fig. 2). The source of the heat is a plasma jet generated with plasma gas ionized by electric arc in the plasmatron (MAJEWSKI 2011, MIKOŚ 1987). The maximum electric power of the plasmatron in all the experiments equalled up to 30 kW.

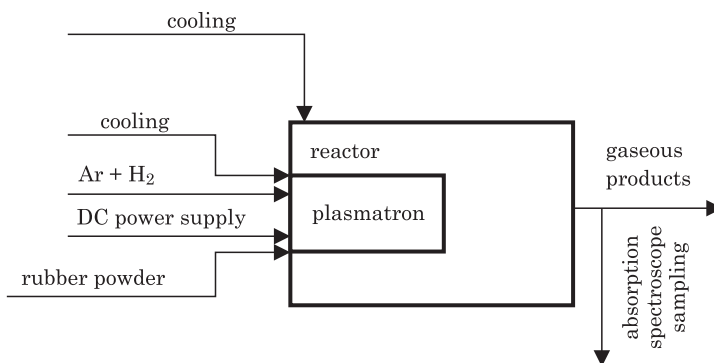


Fig. 2. Scheme of test – stand of plasma pyrolysis of rubber waste

Ar was used as a primary plasma gas. It is an inert gas and so it does not influence composition of the gaseous products. Also, H₂ was used as a secondary plasma gas. On the basis of the additional experiment (SZUSZKIEWICZ 2007) it was found that in the mixture of plasma gas H₂ should not exceed rate of 3.6%. Higher amount of H₂ in the plasma gas could be dangerous. H₂ is a molecular gas and it plays the role of a gas stabilizing the plasma jet. Additionally, it increases the power of a plasmatron.

The plasma generator is a PN-120 type plasmatron (Fig. 3). It has been produced by the Institute of Nuclear Research in Świerk, Poland. The cathode (3)

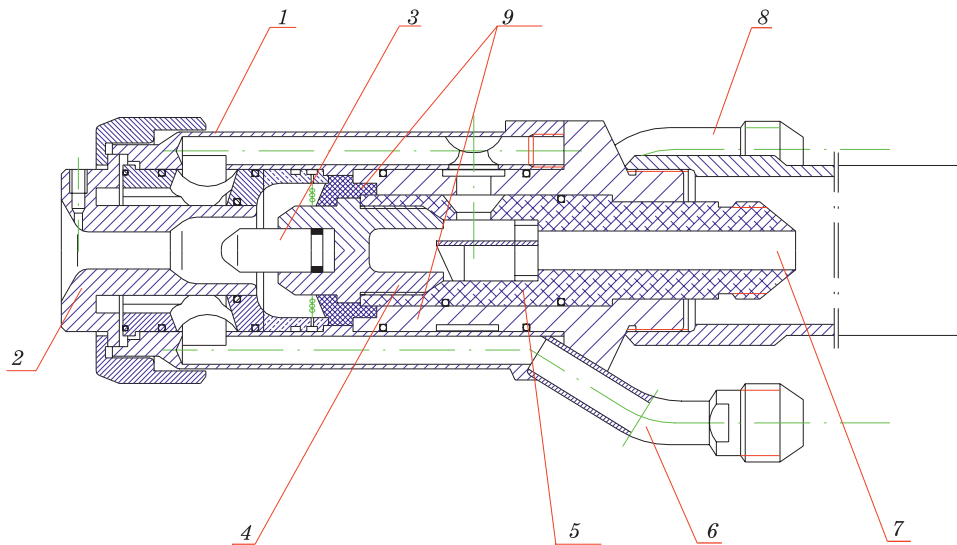


Fig. 3. Cross-section of PN-120 plasmatron 1 – casing, 2 – anode-nozzle, 3 – cathode tungsten rod, 4 – cathode holder, 5 – cathode core, 6 – water drainage; positiver pole of electric supply circuit, 7 – water supply; negativer pole of electric supply circuit, 8 – intake of plasma gas, 9 – isolator

of the plasmatron is made of tungsten. The anode (2) is made of pure copper. The anode also serves as a nozzle, which shapes the plasma jet. The plasma generator is electrically DC supplied. That construction is the most popular amongst all the plasmatrons. The DC electric supply (6, 7) simplifies the electric arc initiation and makes the plasmatron construction easier, less expensive and more dependable.

Since the temperature in the axis of the plasma jet is very high and equals up to 18,000 K (CHAMOLLO et al. 2018), both electrodes of the plasmatron have to be cooled intensively. This task is obtained by the water cooling system (6, 7).

To ensure that the reactions of decomposition and synthesis processes of the rubber are not influenced by the ambient air, the plasmatron was placed in the reactor (Fig. 2). The reactor is an open tank, so the outgoing gas will not cause the overpressure inside of it. The temperature of the reactor is lowered by the separate water cooling system (Fig. 2). The other task of the reactor cooling system is to provide intensive quenching inside of it.

The source of the examined rubber was used tires. The rubber was obtained from the vulcanization plant. The exact chemical composition of rubber mixtures has never been revealed by the tires company. It always is kept as

a secret. Nevertheless, CHANG et al. (1996) revealed the elemental composition of rubber mixture used in the USA: C – 86.84%, H₂ – 7.17%, incinerated mineral compounds – 3.78%, S – 1.89%, N₂ – 0.3%, O₂ – 0.02%.

The rubber was delivered to the plasma jet by a fluidal feeder (Fig. 2). The size of the rubber particles have to be amounting up to hundreds micrometers to get entirely decomposed. Granulation of rubber powder was investigated in SZUSZKIEWICZ et al. (2007).

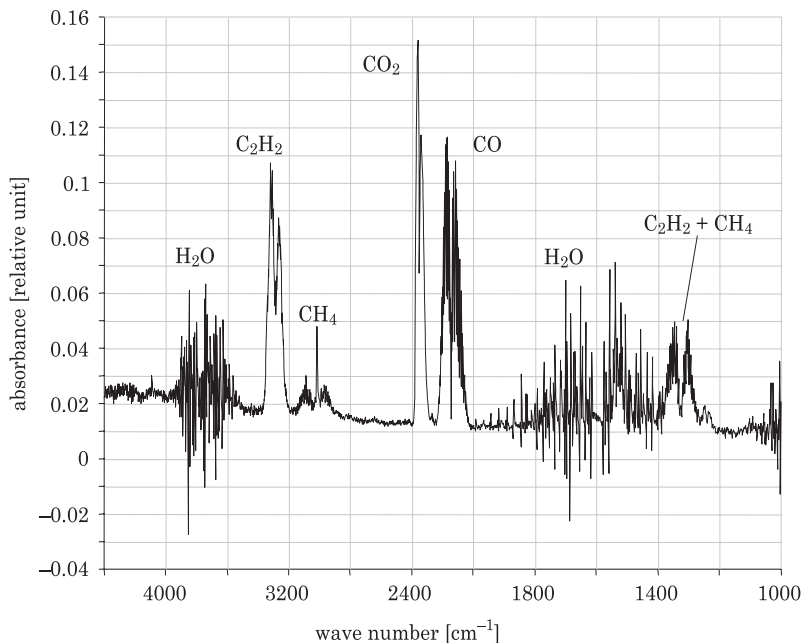


Fig. 4. Absorption spectrum of outgoing gas for pyrolysis of rubber powder in Ar plasma ($P = 21.45$ kW)

The investigation of plasma pyrolysis of rubber was carried out in two separate series of experiments. In the first experiment plasma pyrolysis was carried out in the Ar plasma gas, at constant plasmatron power and constant plasma gas flow rate (Tab. 1). The rubber powder mass flow rate was a variable and ranged from 0.05 kg/h to 8.04 kg/h. In the second experiment plasma pyrolysis was carried out in the Ar + 3.6% H₂ plasma gas (Tab. 2). Also the rubber powder mass flow rate was a variable and ranged from 0.05 kg/h to 8.04 kg/h.

Table 1

Parameters of pyrolysis of rubber powder in Ar plasma

Sample	Voltage U	Current I	Plasmatron power P	Plasma gas flow rate Q	Rubber powder mass flow rate g
	[V]	[A]	[kW]	[l/h]	[kg/h]
1	39	550	21.45	5,325	0.05
2	39	550	21.45	5,325	0.16
3	39	550	21.45	5,325	1.8
4	39	550	21.45	5,325	2.29
5	39	550	21.45	5,325	3.46
6	39	550	21.45	5,325	4.39
7	39	550	21.45	5,325	5.76
8	39	550	21.45	5,325	8.04

Table 2

Parameters of pyrolysis of rubber powder in Ar + 3.6% H₂

Sample	Voltage U	Current I	Plasmatron power P	Plasma gas flow rate Q	Rubber powder mass flow rate g
	[V]	[A]	[kW]	[l/h]	[kg/h]
1	49	550	26.95	5,453	0.05
2	49	550	26.95	5,453	0.16
3	49	550	26.95	5,453	1.8
4	49	550	26.95	5,453	2.29
5	49	550	26.95	5,453	3.46
6	49	550	26.95	5,453	4.39
7	49	550	26.95	5,453	5.75
8	49	550	26.95	5,453	8.04

Results and discussion

The rubber powder was decomposed in two different plasma gases: in pure Ar and in mixture of Ar and 3.6% H₂. Plasma pyrolysis was carried out in the function of the rubber powder mass flow rate.

The exemplary absorption spectrum for pyrolysis of the rubber in the Ar plasma was presented in the Figure 4. There were identified infrared bands of C₂H₂, CH₄, CO, CO₂ and the common band of C₂H₂ and CH₄. The band of H₂O is also visible in the absorption spectrum but it is only a residue of water vapor present in the reactor prior to the experiment.

The presence of H_2 in the outgoing gas could not be verified because of the technical specification of the absorption spectrometer. Although, according to the literature (CHANG et al. 1996), H_2 is present in the outgoing gas.

No bands of NO_x are present in the absorption spectra. No bands characteristic for HCN were found in the spectra, neither. The analysis of the spectra (Fig. 4) confirms that there are no toxic chemical compounds. It means that pyrolysis of rubber in the Ar plasma brings about no undesirable compounds in the outgoing gas. So, there is no need to install any filtration or cleaning systems in the test-stand set-up.

The analysis of the gaseous products in the outgoing gas lets find out that the concentration of C_2H_2 is increasing as the rubber powder mass flow rate grows (Fig. 5). The concentration function of C_2H_2 is monotonically increasing.

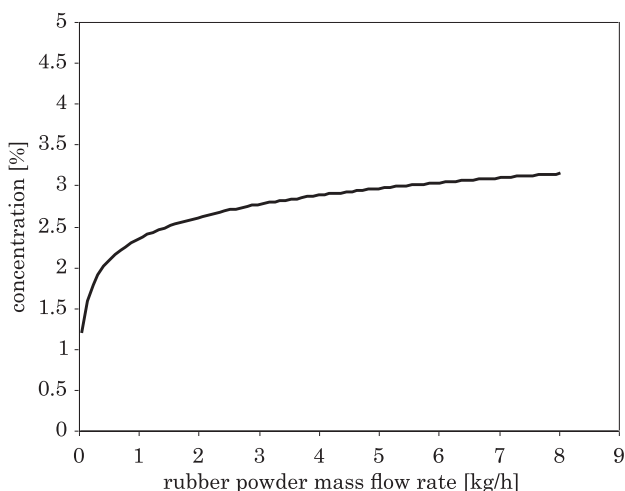


Fig. 5. C_2H_2 in outgoing gas for pyrolysis of rubber in Ar plasma in the function of rubber powder mass flow rate ($P = 21.45$ kW)

The concentration functions of the other identified gaseous products (CH_4 , CO and CO_2) of pyrolysis of the rubber in the Ar plasma (Figs. 6, 7, 8) are not monotonic.

The analysis of the absorption spectra of pyrolysis of the rubber in the Ar + 3.6% H_2 plasma shows off more numerous products of the process. Except for C_2H_2 , CH_4 , CO and CO_2 , identified also in the spectra of pyrolysis of the rubber in the Ar plasma, also C_3H_8 (propane) and C_4H_{10} (butane) were found for pyrolysis in the Ar + 3.6% H_2 plasma (Fig. 9). Obviously, presence of H_2 in the plasma gas stimulated synthesis of bigger number of hydrocarbons.

According to the analysis of the spectra (Fig. 9), neither toxic nor harmful gases have been identified in the outgoing gas for pyrolysis of the rubber in the Ar + 3.6% H_2 plasma.

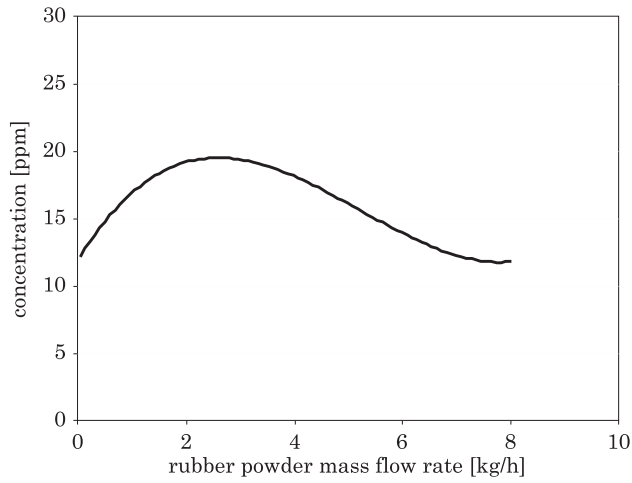


Fig. 6. CH_4 in outgoing gas for pyrolysis of rubber in Ar plasma in the function of rubber powder mass flow rate ($P = 21.45$ kW)

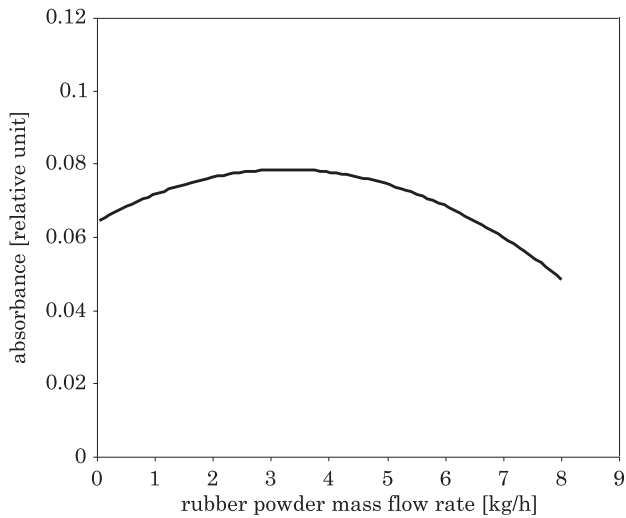


Fig. 7. CO in outgoing gas for pyrolysis of rubber in Ar plasma in the function of rubber powder mass flow rate ($P = 21.45$ kW); CO concentration is proportional to measured absorbance

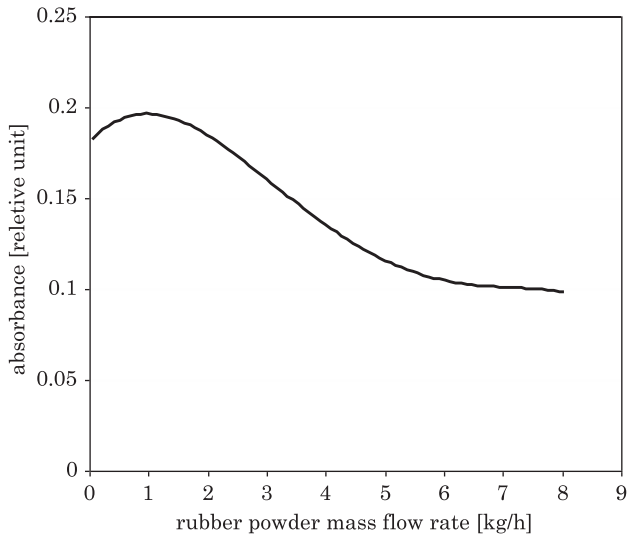


Fig. 8. CO₂ in outgoing gas for pyrolysis of rubber in Ar plasma in the function of rubber powder mass flow rate ($P = 21.45$ kW); CO₂ concentration is proportional to measured absorbance

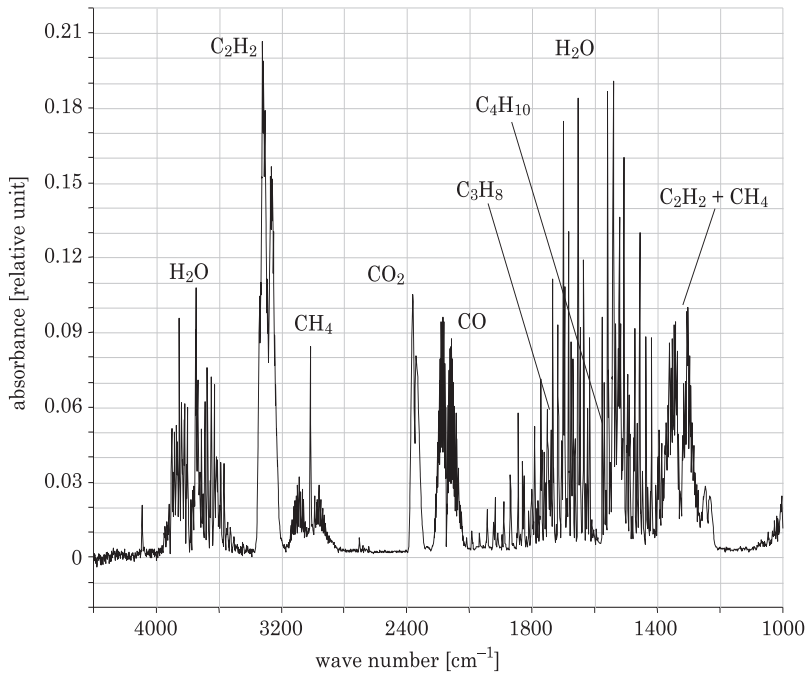


Fig. 9. Exemplary absorption spectrum of outgoing gas for pyrolysis of rubber powder in Ar + 3.6% H₂ plasma ($P = 26.95$ kW)

During the experiment the rubber powder mass flow rate was being changed in the range from 0.05 kg/h up to 8.04 kg/h. The increase of the rubber powder mass flow rate caused the concentration of C_2H_2 , C_3H_8 and C_4H_{10} was increasing (Figs. 10, 11, 12). The concentration functions of the three gases were monotonic.

While the rubber powder mass flow rate was increasing the concentration functions of CH_4 , CO and CO_2 were changing non-monotonically (Figs. 13, 14, 15).

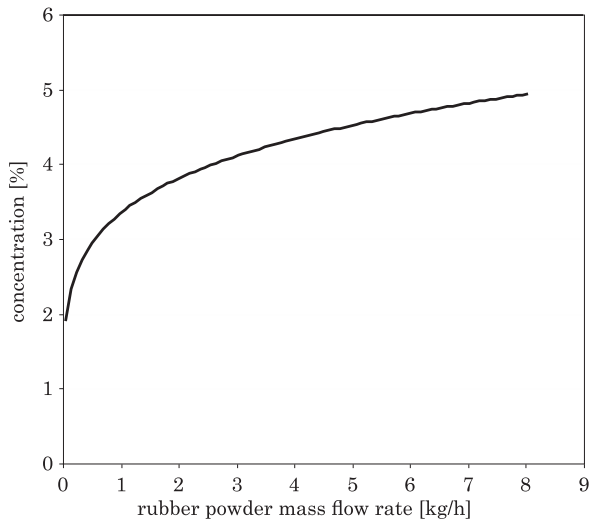


Fig. 10. C_2H_2 in outgoing gas for pyrolysis of rubber in Ar + 3.6% H_2 plasma in the function of rubber powder mass flow rate ($P = 26.95$ kW)

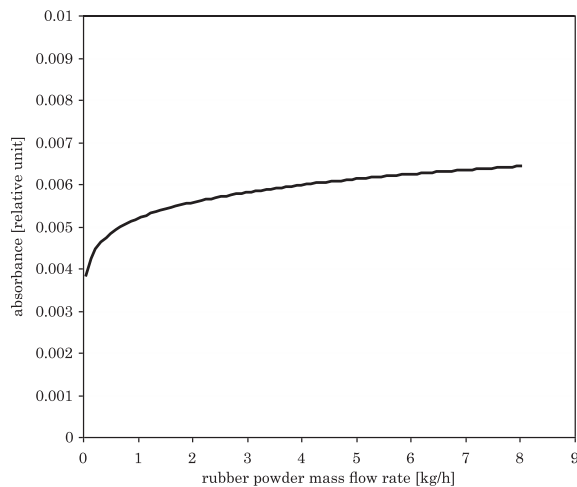


Fig. 11. C_3H_8 in outgoing gas for pyrolysis of rubber in Ar + 3.6% H_2 plasma in the function of rubber powder mass flow rate ($P = 26.95$ kW); concentration of propane is proportional to measured absorbance

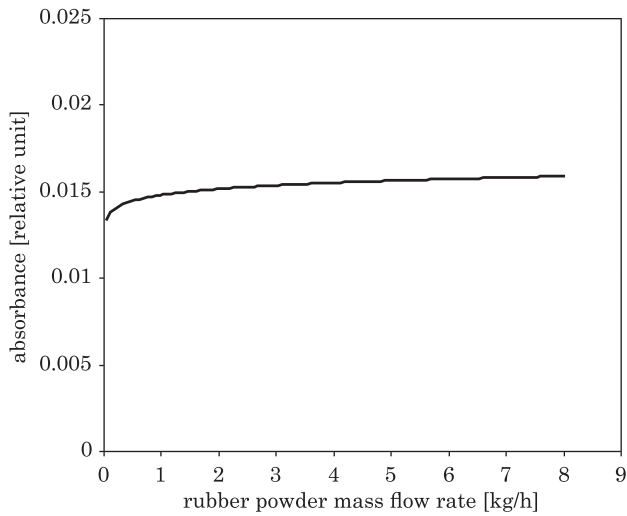


Fig. 12. C_4H_{10} in outgoing gas for pyrolysis of rubber in Ar + 3.6% H_2 plasma in the function of rubber powder mass flow rate ($P = 26.95$ kW); butane concentration is proportional to measured absorbance

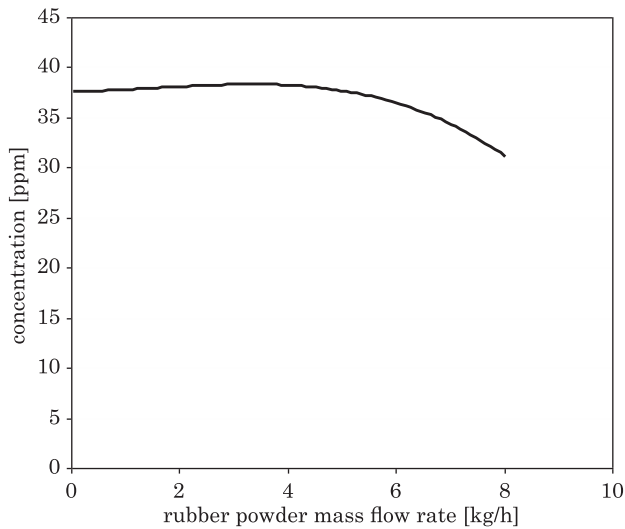


Fig. 13. CH_4 in outgoing gas for pyrolysis of rubber in Ar + 3.6% H_2 plasma in the function of rubber powder mass flow rate ($P = 26.95$ kW)

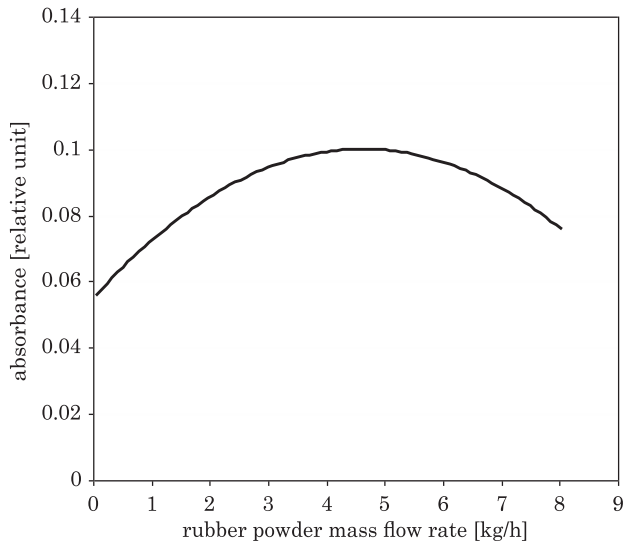


Fig. 14. CO in outgoing gas for pyrolysis of rubber in Ar + 3.6% H₂ plasma in the function of rubber powder mass flow rate ($P = 26.95$ kW); CO concentration is proportional to measured absorbance

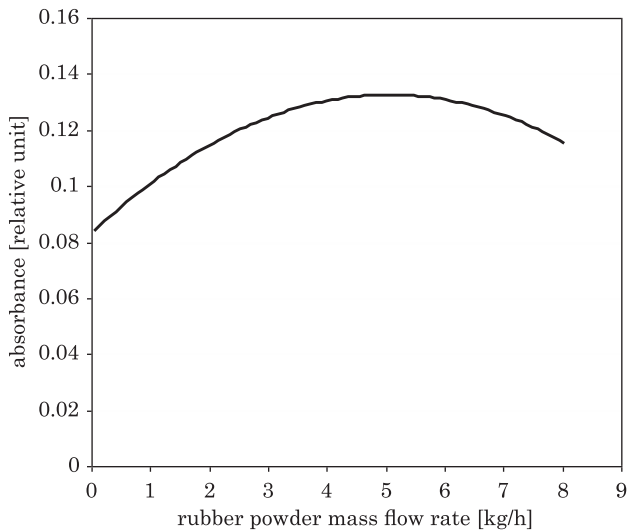


Fig. 15. CO₂ in outgoing gas for pyrolysis of rubber in Ar + 3.6% H₂ plasma in the function of rubber powder mass flow rate ($P = 26.95$ kW); CO₂ concentration is proportional to measured absorbance

Vast majority of the products of plasma pyrolysis of rubber is gas (over 99%) (SZUSZKIEWICZ et al. 2001). The analysis of the solid state products has been done by the atomic absorption spectrometry, flame photometry and absorption spectroscopy FTIR. The analysis revealed that solid state products were mainly soot. It contained: chemical compounds with SO₂ group and twelve elements, namely Pb, Zn, Cu, Fe, Mn, Cd, Cr, Ni, Ca, Mg, Na and K.

Conclusions

The carried out research positively verified all the assumed aims. Plasma pyrolysis seems to be a successful method for utilization of rubber waste. Generally, the increase of the rubber powder mass flow rate brings positive effect concerning the increase of production of most of the gaseous products, especially hydrocarbons.

The detailed conclusions according to the experimental research reveal all the benefits of the plasma pyrolysis of rubber. All the rubber powder introduced to the plasma jet was entirely decomposed. As the products of plasma pyrolysis mainly gas and small amount of soot were obtained.

Despite no filters had been applied no toxic compounds, like NO_x, HCN or SO₂, were identified in the gaseous products of plasma pyrolysis of the rubber.

Application of H₂ as a secondary plasma gas had not only a chemical importance for plasma pyrolysis. H₂ contained in the plasma gas influenced the increase of the electric current in the plasma jet. Also, H₂ stimulated numerical amount and amount of hydrocarbons coming into being during plasma pyrolysis. Concentration of the gaseous products for the Ar + H₂ plasma is bigger than for the Ar plasma.

The increase of the rubber powder mass flow rate did not increase the production of all the gaseous products. The increase of the rubber powder mass flow rate for the Ar plasma made the concentration of C₂H₂ monotonically increased. The concentration of all the other identified products of plasma pyrolysis changed non monotonically.

The increase of the rubber powder mass flow rate in the Ar + H₂ plasma resulted in the monotonic increase of the concentration of C₂H₂, C₃H₈ and C₄H₁₀ and non-monotonic concentration change of all the other identified gaseous products of plasma pyrolysis.

References

- BIADASZ S. 2018. *Practical Use of Rubber Recyclates as Approach to Environmental Protection*. Ecological Engineering, 19(5): 63–74.
- CHAMOLLO GARCES J.C., PREVOSTO L., CEJAS E., KELLY H. 2018. *Quantitative Schlieren Diagnostic Applied to a Nitrogen Thermal Plasma Jet*. IEEE Transactions on Plasma Science, 99: 1-10.
- CHANG J., GU B., LOOY P., CHU F., SIMPSON C. 1996. *Thermal Plasma Pyrolysis of Used Old Tires For Production of Syngas*. Journal of Environmental Science and Health. Part A: Environmental Science and Engineering and Toxicology, 31(7): 1781-1799.
- HUILIN L., XIAOPING W., DEMIN J. 2019. *Recycling of waste rubber powder by mechano-chemical modification*. Journal of Cleaner Production, 245: 118716. <https://doi.org/10.1016/j.jclepro.2019.118716>.
- LIANG M., CHANGJUN S., ZHANYONG Y., HONGUANG Y. 2020. *Utilization of wax residue as compatibilizer for asphalt with ground tire rubber/recycled polyethylene blends*. Construction and Building Materials, 230: 116966.
- MAJEWSKI T. 2011. *Investigation of Plasma Modification Processes of W and Re Powders and W-Re Mixtures*. Biuletyn WAT, LX(2): 231–237.
- MAJEWSKI T., DĘBSKI A. 2012. *Investigation of Plasma Balling Processes of W-Re-Ni and W-Re-Ni-Fe Powder Mixtures*. Inżynieria Materiałowa, 5: 464–468.
- MIKOŚ M. 1987. *Low Power DC Plasmatron for Spraying*, Patent PL 13990281. Urząd Patentowy Polskiej Rzeczypospolitej Ludowej, Warszawa.
- PAWŁOWSKA M. 2018. *Potężny pożar w Trzebini. Pali się składowisko gumowych odpadów*. Dziennik Polski 24. Region. Małopolska Zachodnia <https://dziennikpolski24.pl/potezny-pozar-w-trzebini-pali-sie-skladowisko-gumowych-odpadow-zdjecia/ar/13209477> (access: 22.08. 2019),
- SOVJAK R., PEŠKOVÁ Š., ŠMILAUER V., MARA M., KONVALINKA P. 2019. *Utilization of crumb rubber and FBC-based ternary binder in shotcrete lining*. Case Studies in Construction Materials, 11: e00234.
- SZUSZKIEWICZ J. 2007. *Application of Thermal Methods for Utilization of Used Tires*. Motrol, 9: 178–183.
- SZUSZKIEWICZ J., MIZERACZYK J., DORS M. 2001. *Plasma treatment of rubber waste*. High Temperature Material Processes, 5(3): 345-348.
- TANG L., HUANG H. 2004. *An investigation of sulfur distribution during thermal plasma pyrolysis of used tires*. Journal of Analytical and Applied Pyrolysis, 72(1): 35-40.
- WIELGOSIŃSKI G. 2011. *Review of Technologies of Wastes Thermal Transformation*. Nowa Energia, 1.
- WOJCIECHOWSKI A., DOLIŃSKI A. 2014. *Diversification of Energy Sources from Material Recovery/Organic Materials Recycling*. International Scientific Conference Energy and Environment Production-Logistics-Management, p. 11278-11288.