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Numerical and experimental analysis of pyrolysis process of RDF containing a high percentage of plastic waste

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The current COVID-19 pandemic situation and the associated restrictions have increased the amount of generated waste. It results from the necessity to wear personal protective equipment. Thus, the disposal of masks and gloves is a topical issue and requires immediate investigation. The main aims of this work are management and environmental studies of municipal solid wastes (MSW), which have been generated during the COVID-19 pandemic time. Effective waste management in relation to a circular economy is presented. A sample of refuse derived fuel (RDF) with a high content of plastics was used for the experimental and calculation studies. Pyrolysis was selected as the best thermal decomposition process for this kind of wastes. Proximate and ultimate analyses were performed for RDF and its products. Pyrolysis was carried out using a pilot-scale reactor with a continuous flow of 250 kg/h at 900 °C. Thermogravimetric analysis was applied during the pyrolysis investigation and showed that the main decomposition of RDF took place in the temperature range of 250–500 °C. The pyrolysis gas contained combustible compounds like CO (19.8%), H\textsubscript{2} (13.2%), CH\textsubscript{4} (18.9%) and C\textsubscript{2}H\textsubscript{4} (7.1%), giving a high calorific value – 24.4 MJ/m\textsuperscript{3}. The experimental results were implemented for numerical calculations. Chemkin-Pro software was applied to predict the chemical composition of the pyrolysis gas. The performed computer simulations demonstrated very good agreement with the results obtained during the experiments. They also indicated that there is a strong relationship between the chemical composition of the pyrolysis gas, the process temperature and residence time in the reactor.

1. Introduction

As indicated by the latest reports, the COVID-19 pandemic is a challenge not only from a medical and economic point of view, but also an environmental one, due to the enormous increase in the demand for personal protective equipment (e.g. surgical masks, protective gloves, gowns, goggles and disposable bags and foil gloves) and, as a consequence, an increase in the amount of generated medical waste [1]. The record waste growth of 370% was reported in Hubei Province in China [2]. Fearing the spread of the virus, some countries, such as the US, have partially stopped recycling programs, while restoring the widespread use of single-use plastic bags [3]. Moreover, pandemic restrictions (closed restaurants) have led to an increase in the amount of plastic packaging waste from food delivery and takeaway [4]. The greatest threat is posed by medical waste from quarantined persons from households, which is not subject to restrictive storage and disposal procedures [5]. This waste is most often disposed of as mixed waste in municipal landfills, posing a huge threat to the natural environment and human life [6]. Taking into account all these facts, effective management and disposal of the above-mentioned waste is a global challenge, forcing a review of the technologies and solutions used so far, enabling their safe and effective management [7]. The development of municipal waste management services, especially in urban areas, is not able to keep up with the fast pace of urbanization despite numerous initiatives taken by governments [8]. In addition, the rising costs of disposal, which, according to Aleluia and others, may account for as much as 50% of the city authorities’ budget [9] do not favour proper waste management. Furthermore, more and more stringent regulations in European
legislation, aimed at reducing the environmental burden associated with the storage of MSW [10] determine the directions of waste management, i.e. material (recycling) [11] and energy recoveries using thermal waste conversion methods (combustion, gasification, pyrolysis) (Fig. 1) [12,13].

Pyrolysis and gasification allow the recovery of chemical energy in the form of secondary fuels or provide raw materials for other processes [15], while incineration enables the recovery of electricity and heat [16]. As indicated by the research conducted by Wang et al. [17], undesirable by-products resulting from waste conversion may also be a valuable carbon material at the nanoscale. An example is soot from the high-temperature thermal-chemical conversion of plastic waste. The most common technology of thermal conversion of RDF in Poland is its incineration, while in the face of the constant reduction of pollutant emissions in European legislation, other, more environmentally friendly methods of managing alternative fuels from waste are sought. It seems that the improvement of the overall energy recovery from waste, by combining the efficiency of installations currently operating in Poland, using rotary kilns of cement plants and new pyrolysis and gasification installations, e.g. located in mechanical–biological waste treatment (MBT) plants, will allow for further reduction of the role of landfilling in the Polish waste management. Firstly, it will ensure compliance with the increasingly demanding European regulations [18], and secondly, it will contribute to the rational use of natural resources, while increasing the level of environmental protection [19].

The large-scale implementation of plants for energy recovery from waste may allow in the long term reduce the independence on fossil fuel import. Average estimates suggest that each ton of chemical municipal waste replaces 0.4 tonnes of coal used to generate electricity in the US per year [20]. As indicated by numerous literature sources, one of the most beneficial strategies for municipal solid waste management (MSWM) is the conversion of waste into Refuse Derived Fuel (RDF) [21,22]. RDF can be used as a substitute for conventional fossil fuel in the cement industry [23] and for electricity generation [24]. The efficiency of the methods of thermal processing of MSW is varied, which results from the variable morphology of waste, depending on e.g. from the place of their occurrence [25]. Due to the high flexibility of the obtained products [26], lower emission of pollutants into the air, e.g. SO$_2$ and NO$_x$, compared to incineration and better quality of the solid residue, the pyrolysis process deserves attention [27]. Pyrolysis is a
thermochemical process carried out without oxygen, resulting in the formation of three main products [28], namely: solid (solid residue rich in carbon (char)) [29], liquid fraction (from the condensation of released volatile matter (pyrolysis oil)), and gaseous (including CO, CO₂, CH₄, and H₂), suitable for direct use [30]. The share and quality of products depend on type of waste, pellet packing density, process temperature, type of reactor, residence time and heating rate [31,32]. Most often, RDF undergoes the pyrolysis process at a temperature of 400–900 °C [33] and depending on the set parameters of the process, variable proportions of the obtained products are obtained [34]. Noteworthy is the high calorific value of gas from the pyrolysis of RDF fuel, ranging from 15 to 30 MJ/m³ [35], encouraging its use for energy purposes [36,37].

1.1. Management of waste

The COVID-19 pandemic had a negative impact not only on the global economy, but also on the efficient functioning of local governments and the proper management of municipal waste [38]. Municipalities and cities around the world are struggling with increased amounts of solid waste and medical waste, with less budget inflows due to the economic slowdown [39]. Insufficient forces and resources at the disposal of local governments make them unable to deal with the growing amount of hazardous waste deposited in municipal landfills [40]. The material specificity of this waste significantly differs from the composition of typical municipal waste, due to the presence of a considerable amount of medical waste and an above-average amount of plastic waste [41]. Therefore, the currently operating waste management systems based on landfilling, due to the negative impact on the environment and people, should be replaced with innovative solutions based on thermal neutralization [42]. At present, incineration is the most frequently used method of thermal conversion of municipal solid waste [43]. Other methods of thermal conversion (pyrolysis, gasification) are still in the research phase and they meet with some problems in large scale implementation in Poland [44]. The reasons for the above-mentioned situation can be seen in the lack of sufficient data characterizing municipal waste, poor quality of the raw material, or inadequate structure of the facility [45,46].
The analysis carried out by Vanapalli et al. [2,7] shows that the most widely used techniques for the management of plastic waste are mechanical recycling, incineration and landfilling. At the same time, the authors point out that these techniques are far from perfect in tackling the entire problem of plastic waste. In addition, the development of new solutions should be intensified all the more in connection with the increased amount of waste generated during the COVID-19 pandemic. Trends in plastic waste generation and consequences of pandemic for existing waste management systems and likely solutions to challenges in post-covid-19 waste management systems are shown in Fig. 2 [2,7].

The motivation to undertake research within the above-mentioned issues was the dynamic growth of global interest in the pyrolysis of waste, especially plastics, as well as the current situation on the waste market in Poland and the related problem of the effective management of waste generated during the pandemic. It is worth noting that pyrolysis is a very complex process in terms of chemistry; hence, the possibility of predicting the physical and chemical properties of pyrolysis products requires systematic experimental and numerical tests. An additional argument supporting the above-mentioned issues is that there are not many studies concerning chemical composition of gaseous products of waste pyrolysis, in particular plastic waste based on experimental and numerical investigations. The results of computer calculations presented in this paper complement the current one and provide new knowledge on the chemical mechanisms of the formation of gaseous products of waste pyrolysis process. Moreover, the use of a computational tool such as the ANSYS CHEMKIN-PRO software, which enables a detailed analysis of the chemical composition of gaseous pyrolysis products, is innovative.

The study investigated RDF with a high proportion of plastics as the raw material in the production of potentially valuable products obtained by pyrolysis. Detailed and extensive instrumental analyses of the pyrolysis products were carried out. The obtained results proved the usefulness of employing computer simulations to forecast the gaseous products of thermal conversion.
2. Materials and methods

2.1. RDF sample

Samples of RDF pellets and pyrolysis products, i.e., pyrolysis oil and char, used for the tests, came from the Polish Waste Management Plant. The input material, in the form of an over-sieve fraction of municipal waste, from which RDF pellets were made with the dimensions 2 cm in length and 6 mm in diameter, contained 60% plastic waste, including waste generated during the pandemic (e.g., gloves and protective masks) in addition to paper, cardboard, and foil. The pyrolysis process was carried out under oxygen-free atmosphere at 900 °C.

Owing to the energy efficiency (i.e., the percentage of energy contained in the mass unit of the obtained pyrolysis product in relation to the energy contained in the mass unit of the feedstock) of the obtained pyrolysis products (80% pyrolysis gas, 15% char, 5% pyrolysis oil), the pyrolysis gas and char were subjected to detailed analysis.

2.2. Pyrolysis reactor scheme

The pyrolysis process was carried out using a pilot pyrolysis Auger reactor with a continuous flow of 250 kg/h at 900 °C. The reactor was equipped with an electrically heated spiral heating coil that also acted as a conveyor along the axis of the reactor. A schematic diagram of the process is shown in Fig. 3.

Due to the residence time of the batch in the reactor, i.e., about 20–25 min, the process is classified as an intermediate process, between slow pyrolysis used in batch reactors (4 h) and fast pyrolysis in fluidized bed reactors (~1 s) [47]. The pressure in the reactor was close to atmospheric or slightly under pressure. The raw hot gas left the reactor and went to a vertical and insulated gravity settling tank, enabling the initial separation of dust from the raw gas. The process gas from pyrolysis flowed through the pipeline to the modular shell-and-tube recuperator. Process air was fed in counter-flow through the shell, which was heated to a temperature of 200 °C, was directed to the gas launderer or was used in the process of drying waste. Cooled process gas with a temperature of approx. 550 °C fed the dedusting and desalting system. The gas then went to a system of two shell and tube coolers (condensers), where a condensing liquid fraction and gas at a temperature below 20 °C were formed. When the gas was cooled, residual water and organic condensate (mainly BTEX and naphthalenes) condensed. Additionally, the condensed liquid was collected in the condensate separator. Then the gas was sent to the last stage of purification, which was a series of two filters with an input of active carbon in the form of pellets and other additional sorbents to absorb residual impurities of tar substances, sulphur compounds, chlorine, siloxanes, and others. Additional sorbents to absorb impurities were mineral sorbents in the form of granules with an absorbency of up to 120%. The adsorption mechanism made it possible to keep the guaranteed concentrations, especially of naphthalene and derivatives, and thus ensure the safe operation of the incineration generator [48]. All condensates, liquids or suspensions were collected by gravity through runoff to pallet containers [49]. The composition of the pyrolysis gas was analyzed by gas chromatography.

The scope of the conducted studies and research is presented schematically in Fig. 4.

2.3. Proximate and ultimate analysis

The elemental analysis of the studied materials was performed by means of a LECO CHN628 elemental analyser. The carbon (C) hydrogen (H) and nitrogen (N) contents were determined. Determination of the elements is based on the combustion of a sample under pure oxygen at 950 °C. Helium carrier gas sweeps the combustion gas to separate infrared cells utilized for the detection of H2O and CO2, while a thermal conductivity cell is used to detect nitrogen. The oxygen (O) content was calculated by difference.

2.4. TGa

Thermal analysis was performed to study the thermal behaviour. The sample was subjected to a temperature program (heating rate – 10 K/min) in order to measure the pyrolysis process (atmosphere – nitrogen, flow rate – 40 mL/min). As the output, TGA and DSC curves were obtained. TGA reflects the change in mass of a sample as a function of temperature. The TGA curve was used to detect changes in the sample mass. The DSC signal is the energy absorbed by or released by the sample. The DSC curve enables the detection of endothermic or exothermic effects, and temperatures that characterise a peak. DTG is the first derivative of TGA.
2.5. SEM and EDS analysis

Microstructure examinations were performed for the char sample by means of a JEOL JSM-6610LV scanning electron microscope. In order to obtain the most complete microstructural characteristics of the char, both places representative of the analysed sample and areas showing a different microstructure and morphology were examined. Additionally, in the case of particles with a distinctly different microstructure and morphology from the fraction dominating in the char, EDS studies were carried out to identify their chemical composition and, consequently, to identify the type of waste that was the source of these particles. The EDS examinations were conducted using an X-ray microanalyser by Oxford Instruments working with the JEOL JSM-6610LV scanning microscope.

2.6. Numerical method

Calculations were made based on experimental (process) data. The methodology is described in papers [32,35] and there was used licensed Ansys Chemkin-Pro software [50–53]. The data was applied for the calculations: i) 2–30 s residence time in highest temperatures zone; ii)
800–900 °C temperature range; 250 kg h\(^{-1}\) fuel stream. The range of temperature was done based on TGA results, which showed that no significant changes took place above the temperature of 800 °C.

A detailed chemical mechanism developed by The Creck Modeling Group, involving 167 compounds and 4,656 chemical reactions, was implemented for the calculations. The mechanism took into account the detailed mechanism of high-temperature chemistry of HCl and Cl\(_2\) presented in [54].

A diagram of the modeling procedure is shown in Fig. 5.

### 3. Results and discussion

3.1. Experimental results

3.1.1. Elemental analysis

In Table 1 the elemental analysis as well as calorific values of the studied RDF, obtained char and oil are presented. The studied RDF was typical with contents c.a. 50% carbon and 6% hydrogen and a calorific value of 20 MJ/kg. Such properties allowed successful pyrolysis to obtain high-quality pyrolysis oil and gas. The solid residue can find applications as well.

Fig. 6 presents the TGA, DSC and DTG curves of the pyrolysis process of the studied RDF sample. The first noticeable DTG peak and mass loss reflect the moisture content release (up to c.a. 200 °C). Next, based on TG, one main mass loss is observed in the temperature range from 200 to 425 °C, with the maximum DTG peak at 382 °C. At this stage, the endothermic DSC be assume that main decomposition takes place is detected. This stage is mainly related to the degradation of plastic components, as well as cellulosic components. It can be stated that the main decomposition takes place at a high rate. Additionally, two small peaks on the DTG curve are detected at 457 and 669 °C, respectively. They can be connected with the decomposition of more thermally stable plastic components (LDPE) or lignin [55]. The peak above 700 °C was found for cotton by [55]. Above 750 °C a slight change in the mass is observed. Similar tendency was detected in RDF studied by others [56,57]. Also Sipra A.T. et al. [58] reported that above 500 °C some peaks can appear during RDF decomposition under N\(_2\) and CO\(_2\). At 900 °C solid residue was 20% of the initial mass of the sample.

![Fig. 8. Results of EDS analysis.](image)

### Table 2

Pyrolysis gas composition from RDF pellet pyrolysis tests.

| Chemical compound | Test 1 | Test 2 | Test 3 | Test 4 | Test 5 | Average value | Unit |
|-------------------|--------|--------|--------|--------|--------|---------------|------|
| CH\(_4\)          | 18.9   | 22.9   | 20.4   | 24.6   | 24.3   | 22.22         | %    |
| H\(_2\)           | 13.2   | 18.5   | 16.4   | 22.8   | 21.3   | 18.44         | %    |
| CO                | 19.8   | 12.6   | 19.6   | 23.6   | 23.1   | 19.74         | %    |
| CO\(_2\)          | 21.1   | 14.8   | 16.2   | 17.5   | 16.2   | 17.16         | %    |
| C\(_2\)H\(_4\)    | 7.1    | 10.9   | 13.0   | 5.4    | 6.8    | 8.64          | %    |
| C\(_2\)H\(_6\)    | 2.3    | 1.3    | 1.9    | 0.7    | 0.9    | 1.42          | %    |
| C\(_2\)H\(_8\)    | 6.1    | 1.5    | 2.9    | 0.5    | 0.9    | 2.38          | %    |
| C\(_2\)H\(_10\)   | 3.7    | 0.7    | 0      | 0.2    | 0.4    | 1.0           | %    |
| O\(_2\)           | 1.4    | 3.8    | 2.4    | 1.2    | 1.4    | 2.04          | %    |
| N\(_2\)           | 6.9    | 13.1   | 7.1    | 3.5    | 4.7    | 7.06          | %    |
| HHV               | 26.5   | 21.5   | 23.4   | 19.0   | 20.0   | 22.08         | MJ/m\(^3\) |
| LHV               | 24.4   | 19.7   | 21.5   | 17.3   | 18.4   | 20.26         | MJ/m\(^3\) |

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Fig. 9. Effect of residence time on concentration of selected gaseous pyrolysis products.
3.2. Pyrolysis solid products – Char analysis

3.2.1. SEM analysis

SEM investigations of the solid residue (char) revealed strong differentiation of the material both in terms of morphology as well as the size of the particles present in it (Fig. 7 a-f). Particles ranging in size from tenths of a micrometre to a few millimetres were present in the char, with the dominant fraction being 10 µm to 200 µm. These particles were characterized by an irregular shape and a high degree of surface topography. On the other hand, particles with dimensions above 1 mm were less numerous (Fig. 7 c,d). These particles were characterized by similar morphological features and surface topography as the dominant fraction; therefore, it should be assumed that at least some of these particles could have arisen as a result of the mechanical connection (interlocking) of smaller particles during movement of the char in the furnace space, as well as after the completion of pyrolysis, for example in during the dumping operation. Considering the extensive geometric structure of the surface and the irregular shape of the particles, such a situation was considered very likely. Moreover, there were not many flake-like particles in the char, characterized by a lower degree of surface development, and probably a residue of the starting material with a similar morphology, which did not decompose during thermal treatment, thus maintaining its original shape and microstructure (Fig. 7f). Additionally, numerous particles of a regular, polyhedral shape and more compact structure than the dominant fraction were found (Fig. 7e). In general, the degree of development of the surface of the char components was very diverse, which can be explained by the diversity of the material subjected to thermal conversion and the different chemical compositions of the individual components of the analysed specimen.

In the case of particles whose shape or surface topography were clearly different from the dominant fraction, EDS chemical composition was conducted in order to identify the type of waste that was their source. Based on the performed EDS analyses, it was found that SiO$_2$ silica particles and aluminium waste were present in the char. An example of the EDS analysis of the SiO$_2$ particles is shown in Fig. 8a, and of the aluminium waste in Fig. 8b. The analysed area is marked with a frame. Compared to the dominant fraction, the silica particles are characterized by a clearly smaller specific surface and mostly regular polyhedral shape. In turn, the aluminium waste is in the form of flat particles with a small thickness and a small specific surface. The aluminium waste present in the char is most likely a residue of multi-material waste containing aluminium in the form of thin layers or foils.

3.3. Pyrolysis gas products from pilot installation

The results of the analysis of the gas composition from RDF pyrolysis from the pilot plant are summarized in Table 2. Due to the heterogeneous nature of RDF, this table lists 5 measurements, carried out for the same process conditions, i.e. the temperature of 900 °C in an oxygen-free atmosphere.

Measurements were carried out at time intervals for the same process conditions and the same batch material. The differences in the gas composition are the result of the different morphology of the municipal waste from which the pellets were made.
3.4. Calculation results

3.4.1. Numerical calculations of pyrolysis gas products

As part of the computer simulations, the detailed chemical composition of the pyrolysis gas was determined for the process conditions, and the influence of the temperature and residence time in the reactor in the zone of the highest temperatures on the share of the main gaseous products of pyrolysis determining the calorific value of the pyrolysis gas was analysed, namely CO, H₂, CH₄, C₃H₈, C₄H₁₀, C₅H₁₂, C₆H₁₄, C₇H₁₈, and C₃H₆ (Fig. 9).

The calculations show that the concentrations of all the analysed compounds strongly depend on the residence time in the highest temperatures zone. A shorter residence time, i.e. less than 5 s leads to a higher concentration of most of the analysed compounds, i.e. CO, H₂, C₃H₆, C₆H₁₂, C₇H₁₈ and C₄H₁₀ for the three pyrolysis temperatures. The concentrations of the above-mentioned compounds decline with an increasing residence time. The opposite situation is observed for the remaining compounds, i.e. CH₄ and C₃H₈, the proportions of which increase with an increasing residence time. The largest share of the combustible compound in the pyrolysis gas is observed for CO for the entire analysed range of residence time, with the highest concentration at the temperature of 900 °C and amounts to 40.08%. There are also significant proportions of hydrogen and methane in the pyrolysis gas. The highest concentration of hydrogen was recorded for 900 °C, i.e. 15.3% within 30 s, while methane for the longest time at 800 °C was 14.5%. Noteworthy is the high share of C₆H₆ at the level of 7.5%, which was not determined in the experimental conditions. The share of the other compounds for the analysed residence time and temperatures did not exceed 3%.

The obtained modelling results were compared with the results of experimental studies obtained from RDF pyrolysis carried out at the temperature of 900 °C (Fig. 10). The maximum and minimum analysed values of the residence time in the reactor, i.e. 2 and 30 s, were taken into account.

Comparing the results obtained by modelling with the results from the experiment, good agreement is observed as evidenced by the high proportion of flammable compounds such as CO, H₂, CH₄ and C₂H₄, for all the analysed cases. The modelling results clearly indicate a high proportion of benzene in the pyrolysis gas in the range of 6.23–7.58%. It should be emphasized that in the experiment the concentrations of

Fig. 10. Comparison of calculated and experiment results for selected concentrations gaseous pyrolysis products: a) residence time 2 s; b) residence time 30 s.
compounds with longer carbon chains, i.e. heavy hydrocarbons, as well as their aliphatic and cyclic derivatives which were subject to condensation, were not determined. The concentrations of flammable compounds presented in Fig. 10 translate into a high calorific value of pyrolysis gas, amounting to even 24.4 MJ/m$^3$ for the experiment, while for the modelling it was 23.58% 24.4 MJ/m$^3$ (at the temperature of 800 °C and residence time of 30 s) (Fig. 11). The calorific value was calculated based on the calorific values of the combustible gases and their share in the pyrolysis gas [59]:

$$LHV = [H_2]*LHV_{H_2} + [CO]*LHV_{CO} + [CH_4]*LHV_{CH_4} + [C_2H_2]*LHV_{C_2H_2} + [C_2H_6]*LHV_{C_2H_6} + [C_3H_8]*LHV_{C_3H_8}$$

(1)

It should be noted that the high calorific value is significantly influenced by the presence of heavy organic compounds and their derivatives in the wet pyrolysis gas, even up to 30% of the gas volume. These compounds condense, as a result of which pyrolysis oil is obtained. As the preliminary research carried out by the authors of the article shows, there is a possibility of direct management of wet pyrolysis gas (i.e. a mixture of pyrolysis gas and gaseous oil) in high-temperature heating chambers, where the temperature reaches 1300 °C. Such a high temperature favors the disposal of the problematic liquid fraction, i.e. pyrolysis oil, leading to measurable economic benefits. The proposed solution is described in detail in the paper [60].

4. Conclusion

Based on the conducted research and the obtained results, the following conclusions and statements were formulated:

1. The implementation of pro-ecological solutions in the field of municipal waste management is currently one of the priorities of waste management strategy. It is also an important marketing asset in the market activity of enterprises dealing with the management and processing of municipal waste.

2. RDF made from personal protective equipment and other wastes using during the pandemic (e.g. plastic packaging) has a great energy potential because it consists of plastic, paper and textiles. RDF
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