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Emission Models for Selected Harmful Substances Emitted During Low-Temperature Combustion of Wood Pellets

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ABSTRACT

The aim of the research was to develop mathematical models describing the emission of selected pollutants correlated with the residual oxygen content in the flue gas. The correlation was made for low-temperature combustion of wood pellets in biomass boilers and furnaces. The developed models can be used in modern control systems of boilers, furnaces or for precise calculation of emission factors for the discussed group of heating devices. The description was made for devices with a stationary wood pellet combustion process with a heat output range from 12 kW to 30 kW. The obtained models, not currently used in this group of devices, will allow controlling the operation of heating boilers in a sustainable and ecological way, taking into account the environmental burden.

Keywords: emission, CO_2, CO, NO_x, SO_2, C_xH_y, wood pellet, boiler.

INTRODUCTION

One of the phenomena accompanying the process of combustion of wood pellets is the emission of flue gas. The generated harmful substances are emitted to the atmosphere by an emitter (a heating device such as heater or furnace combined with a flue) [Ciupek and Bartoszewicz 2019; Ciupek 2020]. When dissipating in the atmosphere, the products of combustion may undergo secondary chemical transformations caused by other external factors such as UV radiation, the presence of radicals or solid air pollutants [Rogula-Kozłowska et al. 2019]. Eventually, part of the pollutants remains in the atmosphere and part precipitates on the ground and surface waters [Wierzbińska and Kozak 2023]. The flue gas, as a product of abrupt oxidation of a flammable particle in the oxygen environment is composed of a variety of chemical compounds including the harmful ones. Many of the combustion products have a negative impact on the human organism and the environment [Ciupek et al. 2019b]. Top research works in the area of harmful emissions analyze the energy system of a heating device as a technical object interacting with the environment and the human through its thermal parameters (heating power) but also harmful emissions [Janczak et al. 2019; Judt et al. 2020; Chłopek et al. 2021]. In [Ciupek et al. 2019a; Ciupek 2019], in reference to phenomenological thermodynamics, a solid fuel heater/furnace is perceived as a closed thermodynamic system (within the balance boundary established by the researcher) that is non-uniform and of heavy environmental impact (outside the balance boundary) depending on the operating parameters used by the researcher and the parameters of the fuel. From the literature analysis, it results that these properties significantly influence the changes of the system operation [Ciupek et al. 2019c]. This approach incorporates reflecting the scale of the problem in the form of real-world emissions but does not include the chemical aspects of the combustion process, which, in an in-depth analysis, allows approximating and describing the emission of a given flue gas component. In [Ciupek and Gołoś 2020; Ciupek et al. 2021a], the authors discuss the topic of emissions of harmful substances from non-environmentally controlled heat-generating devices. From the analyses, it results that many heat-generating devices located in close proximity to one another, have a significant share in the overall emissions on a given area. The concentrations of harmful components released to the atmosphere calculated by the researchers largely exceed the limits set forth in standards and directives, which hints a conclusion that emission parameters do not currently constitute an important aspect in operational adjustment of these devices. They are a mere result of the successful obtainment of the preset heating output. In [Rybak-Wilusz et al. 2020; Dong et al. 2022; Fagodiya et al. 2022], the authors analyzed the emission of gaseous and solid products of combustion. The most frequently analyzed components of flue gas and fuel are: carbon dioxide, carbon monoxide, nitrogen oxides, sulfur dioxide and hydrocarbons [Kurc et al. 2020; Rymaniak et al. 2023].

Carbon dioxide (CO_2) is not a directly harmful gas for humans. The symptoms of asphyxiation appear only when its share in the exhaled air exceeds 1.5%. The share of carbon dioxide in pure air amounts to approx. 340 ppm. The harmfulness of CO_2 has a secondary nature and exhibits through a significant impact on the greenhouse effect [Gonzalez-Salazar et al. 2018]. The significance of the emission of CO_2 is currently analyzed in terms of its share in the energy balance of combustion of fossil fuels not only in the emerging economies such as China or India but also traditional fossil fuel-based economies (USA), in which carbon dioxide is the main component of the flue gas [Kowalczyk-Juśko et al. 2023]. The current assumption and prospects for the coming years for the European Union is to limit global warming by maximum 1.5°C, which is directly related to the limitation in the emission of CO_2 to the atmosphere. CO_2 forms in the processes of stoichiometric combustion ($\lambda = 1$). Carbon monoxide (CO) is a colorless, scentless and extremely toxic gas. It displaces oxygen from the blood oxyhemoglobin and bonds with the hemoglobin resulting in reduced oxygen transportation in the blood system. When poisoned by CO, the greatest damage is done to the central nervous system, which distorts mental functions and impacts motor coordination. It forms in the processes of incomplete combustion (oxygen deficit $\lambda < 1$) [Ciupek and Bartoszewicz 2019]. Nitrogen oxides (NO_x) are understood as the sum of nitrogen monoxide (NO) and nitrogen dioxide (NO_2) in the proportion 1.533 NO + NO_2, where the multiplier 1.533 is the ratio of the particle mass of NO_2 to NO. The share of nitrogen dioxide (NO_2) in the total NO_x emitted by solid fuel heating boilers does not exceed 5% [Ahrens et al. 2022]. According to the current state of knowledge, we distinguish four different mechanisms of NO_x formation: thermal, instantaneous, N_2O-assisted and fuel-related. The amounts of emitted NO_x are influenced by the temperature in the combustion zone, the content of nitrogen in the fuel, the concentration of oxygen in the combustion chamber, the rate of combustion or the time of presence of nitrogen in the reagents zone. NO_x is the main component of the California Smog. It reacts with rain water forming acid rains [Polednik 2022]. Sulfur dioxide (SO_2) is the main pollutant (elemental sulfur) and constitutes min. 98% of the entire amount of emitted sulfur [Gai et al. 2018]. Sulfur dioxide is a pungent gas. Its presence leads to the formation of methemoglobin, which in turn, leads to the irritation of the blood producing organs. It is the main component of the Great Smog of London. It reacts with water forming acid rains. It is formed mainly from elemental sulfur originating from the combusted fuel. The emission of hydrocarbons (C_xH_y) is a secondary pollutant resulting from the process of combustion, in which C_xH_y do not undergo a full and complete combustion. The reason for this is an uneven mixture of the fuel and air at a simultaneous abrupt increase of the fuel temperature during its combustion and degassing of a high volume of volatile parts. The harmfulness of hydrocarbons lies in their carcinogenic and mutagenic properties resulting from their chemical activity [Ciupek et al. 2021b].

The authors performed emission-related research on a group of 30 mixed wood pellet-fueled heating boilers of the heating output from 12 kW to 30 kW. The results of this analysis served to determine the objective function for the selected approximated emission parameters, referred to the content of residual oxygen in the flue gas. The effect of the performed research was the linear regression-based approximation of the emission parameters for the averaged values, for which the uncertainty intervals were calculated with the 95% confidence. Within the research, the authors developed mathematical equations of emissions for carbon dioxide (CO_2), carbon monoxide (CO), nitrogen oxides (NO_x), sulfur dioxide (SO_2) and hydrocarbons (C_xH_y). These emissions were assumed to come from low temperature combustion of wood pellets in biomass-fueled heating boilers.

METHODS

The approximation of the mathematical models describing the emission of a given flue gas component in correlation with the content of residual oxygen in the flue gas from a low temperature combustion of wood pellets was carried out for the following components: carbon dioxide, carbon monoxide, nitrogen oxides, sulfur dioxide and hydrocarbons. The obtained equations apply to biomass-fueled heating boilers of the heating output from 12 to 30 kW. Each of the selected flue gas components is recorded and evaluated in terms of its emission not only during approval tests for the EU market but also while in regular operation. The emission tests were performed on wood pellet-fueled heating boilers fitted with automatic fuel supply. The diagram of the measurement stand with a description has been shown in Figure 1. The tests were performed for the nominal heating output of each of the boilers. The combustion quality analyzer (BRAGER BCA-02 ECO) was responsible for the measurement of the content of O_2 in the flue gas. Its measurement range is 0% to 22% of the O_2 volume and the measurement error is ±0.8%. The TESTO 350S and 330-2 LL analyzers served to measure the mass concentrations of CO_2, CO, NO_x, SO_2 and C_xH_y (Fig. 2). The analyzers were fitted with long-life photochemical measurement cells. The time of analysis was set at one sample per one second and the single test cycle for one boiler continued for at least 3h. The measurement accuracy for CO_2, CO, SO_2 and C_xH_y is ±10% and for NO_x - ±8%. The recording of the emission data was based on a state-of-the-art recording station equipped with measurement cards operating in a measurement chain developed in the LABview environment.

When performing the investigations, the authors observed that the emission of the harmful components during the combustion of low temperature wood pellets is described by functions, the form of which we do not know but can measure their values and their derivatives for given values of the argument such as the content of residual oxygen in the flue gas. Having the set of measurements of the emission values of individual flue gas components obtained at given times of the measurements, it is possible to interpolate the equations of these emissions for the assumed sensitivity of representation. For the development of the mathematical models, the authors used the method of linear interpolation. The calculations through statistical analysis of the emissions for a given emission parameter allowing for the

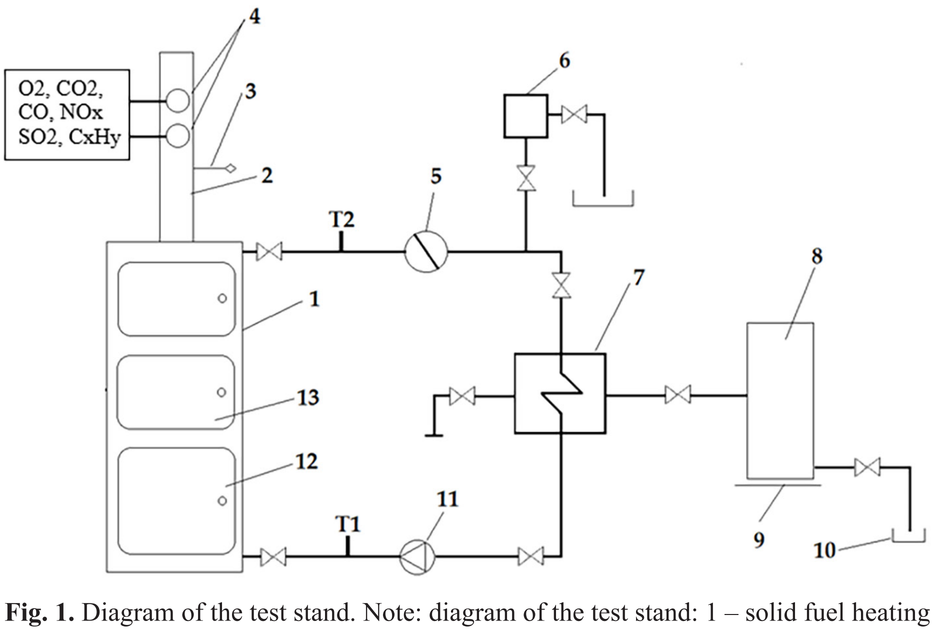

Fig. 1. Diagram of the test stand. Note: diagram of the test stand: 1 – solid fuel heating boiler, 2 – flue, 3 – measurement temperature, 4 – pollutant measurement points, 5 – water flow meter, 6 – expansion tank, 7 – heat exchanger, 8 – water tank, 9 – weight scale, 10 – outflow, 11 – water pump, 12 – combustion chamber, 13 – heater chamber.

Fig. 2. Flue gas analyzers used in the tests: Testo 350S (left), Testo 330-2 LL (right)

reference to the residual oxygen in the flue gas were made with the 95% level of confidence using the Microsoft Excel GNU Octave environment. Interpolation is what we refer to as action that leads to finding of a value of a certain function *f(x)* at a random point of the interval (x_0, $x_{1, \ldots,}$ $x_{n,}$ $x_{n+1,}$ x_{n+m}) where *n* and *m* are the consecutive expressions of the interval, based on the known values of this function at points characteristic of this interval:

$$x_0, x_1, \ldots, x_n \tag{1}$$

referred to as the nodes of interpolation, described with the relation:

$$(x_0 < x_1 < \ldots < x_n) \tag{2}$$

When mathematically describing the emission of selected flue gas components, the interpolation methods were applied, because, thus far, we do not know functions describing emission of a given flue gas component during low temperature combustion of wood pellets. The performed tests rendered the values of the emissions at given work points of the boilers describable by the content of residual oxygen in the flue gas. Hence, we may infer that the measurement of the emission of a given harmful component referred to a given work point of the heating device is represented by the residual oxygen level in the flue gas. The interpolation multinomial $W_n(x)$ is a multinomial of the maximum *n* level that, in points (x_0, $x_{1, \ldots,}$ $x_{n,}$ $x_{n+1,}$ x_{n+m}) meets the conditions of the interpolation:

$$W_n(x_i) = y_i \tag{3}$$

For the base points: i = 0, 1, 2,…, n. Hence, we may assume that there exists exactly one interpolation multinomial that, in points (x_0, $x_{1, \ldots,}$ $x_{n,}$ $x_{n+1,}$ x_{n+m}) meets the conditions of the interpolation as per the above formula. The interpolation nodes x_0, x_1,..., x_n can be distributed freely and the sought multinomial can be written in a general form:

$$W_n(x) = a_0 + a_1 x + a_2 x^2 + \ldots + a_n x^n \tag{4}$$

Using the definition of an interpolation multinomial, we obtain a system of equations *n+1*, with *n+1* unknowns and factors: a_0, a_1,..., a_n:

$$\begin{cases} a_0 + a_1 x_0 + a_2 {x_0}^2 + \ldots + a_n {x_0}^n = y_0 \\ a_0 + a_1 x_1 + a_2 {x_1}^2 + \ldots + a_n {x_1}^n = y_1 \\ \ldots \\ a_0 + a_1 x_n + a_2 {x_n}^2 + \ldots + a_n {x_n}^n = y_n \end{cases} \tag{5}$$

The matrix of the factors of this system is the Vandermonde matrix in the form:

$$A = \begin{bmatrix} 1 & x_0 & {x_0}^2 & \ldots & {x_0}^n \\ 1 & x_0 & {x_1}^2 & \ldots & {x_1}^n \\ \ldots & \ldots & \ldots & \ldots & \ldots \\ 1 & x_0 & {x_n}^2 & \ldots & {x_n}^n \end{bmatrix} \tag{6}$$

Assuming that $x_1 \neq x_i$. Therefore, such a matrix system has exactly one solution, and the values $a_{i,}$ as per the Cramer's theorem, are determined with the formula:

$$Aa_i = \frac{1}{A} \sum_{j=0}^{n} y_j A_{ij} \tag{7}$$

where: A_{ij}(j = *0, 1, 2,..., n*) are the consecutive algebraic complements of the elements of the i-th column of determinant A.

Table 1. Mathematical models together with representation accuracy for selected pollutants

Harmful substance	Mathematical model	Representation accuracy
CO	CO [mg/m³] = -4,733 · O_2 [%] + 312.478	94.49%
CO_2	CO_2 [mg/m³] = -0,890· O_2 [%] + 17.351	96.26%
NO_x	NO_2 [mg/m³] = 0,109 · O_2 [%] + 191.008	93.09%
SO_2	SO_2 [mg/m³] = -5,620 · O_2 [%] + 195.445	95.33%
C_xH_y	C_xH_y [mg/m³] = -6,180 · O_2 [%] + 147.633	94.14%

Using the methods of linear approximation, the authors obtained emission equations regarding the pollutants under analysis that allow calculating the emission of a given pollutant based on the oxygen level content in the flue gas. To this end, in order to calculate the emission of a given pollutant (marked 'y' in the equation), for 'x' we substitute the oxygen residual content in the flue gas. Then, we can easily calculate the assumed emissions of a given pollutant for the known residual oxygen resulting directly from the operation of a given heating device.

RESULTS

Table 1 presents the mathematical models and the sensitivity of their representation for the entire test sample, the selected five harmful components (CO, CO_2, NO_x, SO_2 and C_xH_y) for the final mathematical iteration and the approximation of the emission functions. Based on the developed mathematical models and estimated sensitivities of their representation (Table 1), the authors performed a simulation of the emission of a given pollutant (CO, CO_2, NO_x, SO_2 and C_xH_y) with a view to validating the developed models with the real-world operation of the wood pellet heating boilers. The simulation of the emissions was carried out in the GNU Octave environment using basic 'while' loops for the argument call in the form of the oxygen content in the flue gas (O_2 [%]). For each pollutant, an appropriate decision loop was prepared utilizing the approximated linear equation. Upon determining of the factors and the sensitivity of the representation against the obtained physical measurements on the level of ± 10%, corrections of the model were made in order to specify the emission results for each flue gas component. The emission simulations were carried out for the oxygen content in the flue gas in the range 1% to 19.5% with the measurement resolution of 0.5%, compliant with the measurement accuracy of the most commonly applied oxygen sensors. The maximum content of O_2 in the flue gas on the level of 19.4% results from the Siegert stoichiometric equations for the combustion of biomass [Ciupek and Gołoś 2020].

TECHNICAL APPLICATIONS

The calculation methods of the emission of pollutants generated by the combustion of wood pellets in automatic low heating power boilers, are used in a variety of technical applications. They are especially useful in solutions requiring on-going feed of information regarding individual emission components throughout an extended period of time. Combustion of solid fuels is one of the fundamental energy-related processes carried out for heating purposes in individual and urban heating. The accessibility to solid fuels and a wide range of their types ensure energy security and logistic independence from a single supplier or importer. From the viewpoint of the end user, such a solution eliminates the risk of energy supply monopoly, in which case, the end user is forced to accept services from a single fuel supplier. Currently in Poland, a variety of solid fuels marketed as wood pellets is available, the parameters of which are not verified and controlled at the stage of production and marketing. Only certified wood pellets, whose quality parameters are controlled according to the EN ISO 17225-2 standard ensure the repeatability of the operating parameters during combustion. It is noteworthy that during laboratory tests, the emission parameters of wood pellet-fueled boilers (calorific value, humidity and chemical composition) are verified with a high level of repeatability of the applied fuel. Such a situation guarantees the emission/energy parameters of a given type of heating boiler as declared by the manufacturer. In real-world operation, due to the varied parameters of the wood pellets resulting from the accessibility to uncertified products, whose origin is not limited to wood but extended to sunflower shells, agricultural waste (agropellets) or a variety of banned additives, we experience a significant deterioration of the emission-related parameters of the heating devices. This phenomenon not only leads to air pollution but also reduces the useful life of a heating device causing premature failure of the main device components (burners, heat exchangers). Owing to the application of simple mathematical models that do not require great computational power, the calculation method proposed by the authors of this paper allows its direct implementation in the heating boiler control units. Earlier research clearly indicates the need for emission analysis as a tool for diagnosing problems with the process of combustion and its subsequent improvement [Nygard at al. 2019; Urbaniak at al. 2013]. Detailed information (real time) pertaining to the emission of CO and CO_2 allows a correction of the air/fuel mixture ratio, which eliminates incomplete and non-full combustion. The proposed mechanisms of adjustments are particularly justified during reduced heat energy demand (autumn and spring months), where the boilers operate in the range 12% to 30% of their heating output [Ciupek at al. 2021a]. Another iteration of the process of emission reduction from wood pellet heating boilers is the use of calculations related to the emission of nitrogen oxides as a substance, the formation which we may control by adjusting the combustion process and applying aftertreatment. Another area necessitating the calculations of the emissions of all identified pollutants is environment protection. In the years 2021–2023, within the area of the city and municipality of Pleszew, a pilot program was carried out with the following assumptions:

- registration of all heating devices divided into classes and sources of power,
- monitoring of the upgrade/renewal of the heating devices in real time,
- recording of the operation of wood pellet and coal-fueled heating boilers troughout the calendar year,
- fitting of 30 sensing devices for the measurement of the dustiness level in the area of the said city and municipality
- detailed analysis of the air quality in relation to the flue gas emissions from solid fuel heating boilers.

Following the completion of the program, a comprehensive view was obtained of the correlation between the air pollution and the number of solid fuel heating devices and their quality of operation in the above-mentioned area. Owing to the recording of all the operating parameters of the heating boilers, the developed calculation methods will be used to calculate all individual pollutants in the period of the last 3 years, thus providing a full view on the impact of heating devices on the natural environment.

CONCLUSIONS

Upon the analysis of the performed research and the obtained results of the numerical simulations of the individual emissions in correlation with the main objective of the research, the following conclusions were drawn. It is possible to determine the range of emission of individual pollutants generated during a combustion of individual types of solid fuel currently used for individual heating applications, particularly wood biomass representing a popular source of renewable energy in Poland. One may observe a heavy impact of the types of heating devices and the types of fuel on the quality of the combustion process. When applying numerical iteration methods, it is possible to approximate equations describing the emission of individual pollutants (e.g.: CO, CO_2, NO_x, SO_2 and C_xH_y) in reference to the residual oxygen in the flue gas. It is possible to correlate individual pollutants with the residual oxygen level in the flue gas as a diagnostic parameter allowing the comparison of individual emissions for the technical objects under analysis. The performed research has been justified by technical implementations in pellet-fueled heating boilers and the use of its findings by public administration. While still maintaining the ongoing recording of the investigated parameters, the calculation methods, despite their lesser accuracy, are characterized by a lower cost of acquisition as opposed to direct measurements.

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