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Influence of fuel mixture and catalyst on the ethanol burner flue gas composition

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Abstract

The aim of this study is to determine the influences of chosen primary (mixing of standard recommended fuel with additives) and secondary (oxidation catalyst) measures on the composition of flue gas, produced by combustion of liquid fuels in a standard ethanol burner. The total amount of emitted NO_x was decreased by 63%, after replacing the fuel by pure methanol, while the total amount of emitted CO was the lowest during pure ethanol usage. The implementation of chosen catalyst significantly affected the total amount of emitted CO with high conversion rates (82%–90%). The amount of emitted water vapour was increased by 0%–25% (per 1 kWh of released energy) when additives were used. Overall, applying the mentioned measures together proved to be very beneficial from the total amount of emitted CO and NO_x point of view.

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1. Introduction

With the declining stability of the European energy market, the demand for appliances and fuels for decentralized household heating is growing significantly as was mentioned by Canepa [1]. Consequently, increasing prices of the fuel and combustion appliances together with the lack of production materials started a discussion about alternative pathways for household heating. According to Mikkell [2], one of the alternatives for local heating could be bioethanol (hereafter just ethanol) combustion in the EB. Šproho [3] describes EB as uniquely designed containers filled with ceramic wool usually equipped with a regulation flap. The real utilization of EB as the source of HE was described by Hajamalala [4]. The heat output parameters of different construction types of EB were described by Ryšavý et al. [5]. EB is usually installed in an EF. EF usually does not affect the combustion process itself [6].

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Nomenclature

$A_{r,C}$	relative atomic mass of carbon, [-]
$A_{r,H}$	relative atomic mass of hydrogen, [-]
$A_{r,O}$	relative atomic mass of oxygen, [-]
EB	ethanol burner
EF	ethanol fireplace
FG	flue gas
HE	heat energy
HHV	higher heating value
k	heat of vaporization considering the volumetric work done by the water formed from the hydrogen during combustion at 25 °C, 2.37 MJ kg ⁻¹
k_1	specific heat of water evaporation at constant pressure at 25° C, 2.44 MJ kg ⁻¹
LHV	lower heating value, MJ kg ⁻¹ ; kWh kg ⁻¹
\dot{m}_{fuel}	fuel mass flow, kg min ⁻¹
m_{H_2O}	specific mass of emitted water vapour in the flue gas, kg kWh ⁻¹
\dot{m}_i	pollutant mass flow, mg min ⁻¹
M_i	molar mass, kg mol ⁻¹
\dot{V}_{fg}	volume flow of flue gas at STP conditions, m ³ min ⁻¹
$V_{m,air}$	molar volume of air, dm ³ mol ⁻¹
V_{m,CO_2}	molar volume of CO ₂ , dm ³ mol ⁻¹
$V_{m,i}$	molar volume of pollutant, dm ³ mol ⁻¹
λ	excess air ratio, [-]
φ_i	pollutant volume fraction in the flue gas, ppm
$\varphi_{O_2,air}$	volume fraction of oxygen in the air, [-]
$\varphi_{O_2,fg}$	volume fraction of oxygen in the flue gas
ω_C	mass fraction of carbon in the fuel, kg kg ⁻¹
ω_H	mass fraction of hydrogen in the fuel, kg kg ⁻¹
ω_N	mass fraction of the nitrogen in the fuel, kg kg ⁻¹
ω_O	mass fraction of the oxygen in the fuel, kg kg ⁻¹
ω_w	mass fraction of the water in the fuel, kg kg ⁻¹

The main purpose of the appliances for ethanol combustion (EB and EF) nowadays, is to enable the feeling of a real, open fire in the houses without a chimney. However, due to the high heat output of the burners, they should be considered as the source of HE, especially in case of their usage in the houses with low HE demand [3].

Denatured ethanol, as the only allowed fuel for EB (according to users manuals), can be produced by many ways, by using many different types of input feedstock for example: molasses [7], palm empty fruit bunch [8], corn stems [9], waste food [10], sugar or starch crops [11] and rice straw [12]. Input materials are usually connected with local production which significantly reduces dependence on imported products. The quality of the standardly sold fuels differs especially in the water content within the fuel, as was presented by Nozza et al. [6], Martinka et al. [13] and Ryšavý et al. [5]. According to a study of Breaux [14], water/ethanol mixture is cheaper and much easier to manufacture since the distillation energy used for ethanol dehydration (for reaching more than 99% of the mass fraction of ethanol in the fuel) could cost approximately 1/3 of the overall energy consumed. The mass fraction of the water in the fuel is strongly related to its LHV and, consequently, to the released heat, quality of combustion process and FG composition.

Ethanol combustion is a complicated process, consisting of many chemical reactions which were described by Millán-Merino et al. [15]. Estimated products of ethanol combustion are CO₂ and H₂O. The European standard: EN 16647 Fireplaces for liquid fuels [16] is considering the EF, especially from the technical and safety point of

view, including the limits for the volume fraction of CO and CO₂ in the FG. Since the mentioned standard is not harmonized for the EU, the stated limits are not mandatory. Due to the real combustion process of ethanol, pollutants such as CO and NO_x could occur in the FG in not negligible concentrations as was proved by previous studies of Höllbacher et al. [17] and Schripp et al. [18]. EB and whole EF usually do not have any air flow control system, air staging control system nor automatic actual heat output regulator for the combustion process improvement.

Apart from fuel conversion [19], nitrous oxide decomposition [20] or CO₂ reduction [21], catalytic substances can be also applied for FG purification from the products of incomplete combustion as well, making any burner design improvement unnecessary. As was proved by Ryšavý et al. [22], conversion rates of the CO by Pt and Pd based catalyst in real FG formed during the wood combustion are strongly connected with FG parameters, especially the FG temperature. According to results achieved by Ryšavý et al. [5], there is a presumption that the FG temperature of the EB will be sufficient for an oxidation catalyst operation.

This study is aimed at the composition determination of the FG formed during the ethanol combustion in commonly sold EB, especially from the CO and NO_x points of view. Consequently, the study is aimed at quantification of the influence of mixing ethanol with water and methanol and of the utilization of oxidation honeycomb catalyst on flue gas composition.

The novelty of the research is in the application of primary (fuel treatment) and secondary (oxidation catalyst) measures, commonly used during the small solid fuel combustion appliances usage, during the EB usage.

2. Materials and methods

2.1. Experimental equipment

The experimental stand was equipped with the XS balance BL 30K1 scale. The scale was laid on the wooden pallet to ensure the freest possible combustion air flow to the burners, which were placed on the scale. Between the burners and the scale, there was a heat resistant plate. Above the burners, a hood (its position was set according to visible flame to avoid touching the hood by the flame) was situated. At the top of the hood, there was a place for catalyst installation (just for the cases when the catalyst was used). At the catalyst outlet, there was a place for FG temperature monitoring. FG then flowed through the duct out to the ambient air. The chimney draught was regulated by the regulation flap to avoid the excessive flow of the air around the burner and the flame, which did not occur during the standard burners' usage in case of their installation in the fireplace. Due to the negligible pressure drop of the catalyst, the regulation flap was at the same position during all measurements. FG sampling took place approximately 500 mm above the FG outlet from the catalyst, to ensure steady–laminar FG flow. Sampled FG flowed through a heated ceramic filter and a heated hose to the analyser Horiba PG-350E. The simplified scheme of the experimental device is shown in Fig. 1 below.

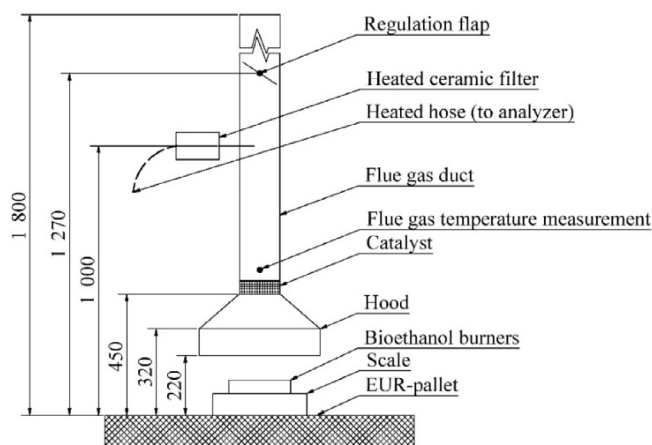


Fig. 1. Simplified diagram of the experimental device.

Two pieces of single-chambered EB were chosen for the experiments. The construction of single-chambered EB was described by Ryšavý et al. [5]. The expected heat output of each burner according to the manufacturer was 1.5 kW. The burner opening area was 3464 mm². The maximal fuel dose for one combustion period (for one EB) was 200 ml according to the manufacturer's instruction.

2.2. Catalyst

For the experiments, the oxidation honeycomb catalyst with palladium (Cat A) as an active element was chosen. The producer of the catalyst was Whitebeam d. o. o. Detailed information about the catalyst is shown in Table 1.

Table 1. An example of a table. (nd — not defined; pressure loss was defined for FG flow 20 m³ h⁻¹).

Cat A	Unit	Value
Body material	–	cordierite
Carrier	–	Al ₂ O ₃
Washcoat material	–	Pd
Weight of active element per cubic metre	g m ⁻³	475
Inlet surface area	m ²	0.0163
Height	m	0.051
Effective volume	m ³	8.306 · 10 ⁻⁴
Cell density	cells cm ⁻¹	4.14
Cell shape	–	quadratic
Pressure loss	Pa	1

The producer of the catalyst provided the regular operating temperature but did not provide the conversion efficiency diagram. Only the physical and chemical properties provided by the manufacturer were known, while no additional testing to determine active layer thickness or carrier material were performed. The catalyst with its diameter of 0.144 m fits perfectly into the FG duct (thin free space around the catalyst was filled by a sealing cord). Before the main experiments, the catalyst was used for several wood log combustion tests (less than 50 h of operation), after which the catalyst was calcinated at a temperature of 550 °C, so the catalyst was without any damage by poisoning, fouling, clogging, coking or sintering.

Description of reactions occurring on the catalyst's surface is not the aim of this study. These reactions were described for the chosen type of catalyst by Langmuir et al. [23]. This research is aimed only at already produced catalysts usage for FG purification.

The main reason for choosing the Pd-based catalyst is because of positive results obtained during the previous tests with similar catalysts aimed at the flue gas purification formed during the solid biomass combustion. Previous tests with the chosen catalyst have also shown remarkable effectiveness even at relatively low temperatures usually reached during the ethanol combustion in EB. Other reasons for choosing this catalyst are easy availability on the market and low pressure drop.

2.3. Fuel

Basic information about the used fuels are listed in Table 2. Commonly sold high-quality ethanol called BIOSpirit 100 was chosen as the basic fuel for these experiments. The manufacturer declared zero mass fraction of the additives, especially water, in the ethanol, exclusive of the denaturants. LHV listed in the safety documents was 26.9 MJ kg⁻¹. As the additives for the decrease the mass concentration of pollutants in the FG, distillate water and methanol were used. The methanol HHV listed in the safety data sheet was 22.884 MJ kg⁻¹.

The real HHV of the fuels was determined using calorimeter LECO AC600. Each sample was measured at least five times and the averages were taken into the consideration. The obtained values of LHV were recalculated from the HHV, according to the following formula Eq. (1) mentioned in the standard DIN 51900-1 [24]:

$$LHV = HHV - (k \cdot 8.94 \cdot \omega_H + 0.8 \cdot (\omega_N + \omega_O) + k_1 \cdot \omega_W) \quad (1)$$

Densities of the liquids were obtained from safety data sheets. Densities of the mixtures were calculated as the weighted arithmetic mean. Fuel mixtures are stated as the weight ratios of each substance.

Table 2. Basic information about the fuel and fuel mixtures.

Fuel	HHV [MJ kg ⁻¹]	LHV [MJ kg ⁻¹]	Density [kg dm ⁻³]
ethanol	29.38	26.57	0.789
methanol	22.88	19.93	0.792
ethanol/water (90/10)	26.86	24.08	0.810
ethanol/water (80/20)	23.88	21.13	0.831
ethanol/methanol (90/10)	28.73	25.91	0.789
ethanol/methanol (70/30)	27.39	24.26	0.790
ethanol/methanol (50/50)	26.06	23.02	0.791

2.4. Process of the testing and data evaluation

Every testing mode started by zeroing the scale with a heat resistance plate and the burners on it. Consequently, the fuel was prepared and filled into the burners. For each test, 400 ml (2 × 200 ml) of the fuel was used, which was the maximal allowed capacity of the burners, according to the manufacturer. After the fuel loading, 5 min delay was made for ensuring maximal and uniform fuel saturation into the ceramic wool inside the burner. Consequently, the fuel weight was recorded and fuel was ignited. Weight loss of the fuel was monitored during the burning process and recorded every minute until the weight dropped back to zero.

From the weight loss and the LHV of the fuel, the actual heat output of the burners was determined for each minute. FG sampling started at the exactly same time as the ignition. All monitored FG parameters were measured continuously with data recording as one-minute averages.

The actual dry FG flow (at STP conditions) was obtained by recalculation of the theoretical FG flow (given by the C, H and O composition of the fuel) according to the real volume fraction of oxygen in the FG according to the Eq. (2):

$$\dot{V}_{fg} = \left(\frac{V_{m,CO_2}}{A_{r,C}} \cdot \omega_C + \frac{\frac{V_{m,air}}{A_{r,C}} \cdot \omega_C + \frac{V_{m,air}}{A_{r,H} \cdot 4} \cdot \omega_H - \frac{V_{m,air}}{A_{r,O} \cdot 2} \cdot \omega_O}{\varphi_{O_2,air}} \cdot (\lambda - \varphi_{O_2,air}) \right) \cdot \dot{m}_{fuel} \quad (2)$$

Pollutant mass flow was calculated from the measured volume fraction, molar mass and molar volume of the pollutant and from the flue gas flow at STP conditions according to the Eq. (3) [25–28]:

$$\dot{m}_i = \varphi_i \cdot \frac{M_i}{V_{m,i}} \cdot \dot{V}_{fg} \quad (3)$$

The total mass of the emitted pollutant was calculated as the sum of the values for each minute of the testing period. Detailed information about the measuring equipment, the measured range of each equipment, the principle of the measurement and the accuracy are shown in Table 3.

Table 3. Detailed information about the measuring equipment, measured range of each equipment, the principle of the measurement and the accuracy.

Device and measured component	Range	Principle	Accuracy
Horiba PG-350E			
CO	0 – 200 ppm	NDIR	±1%
CO ₂	0%–10%	NDIR	±1%
O ₂	0%–25%	Paramagnetic	±1%
SO ₂	0 – 200 ppm	NDIR	±1%
NO _x	0 – 50 ppm	Chemiluminescence Detection Method	±1%
Thermocouple, Type K, class 1			
FG temperature	- 50 to 1000 °C	Thermoelectric effect	±1 °C

3. Results and discussion

Ten combustion tests were performed in total. The most important results from each test are listed in Table 4. As can be seen, in Table 4, LHV of the fuels or fuel mixtures significantly affected the energy input to the burner,

Table 4. The most important results from combustion tests.

Fuel	Fuel loading [g]	Energy input [kWh]	Catalyst [-]	Maximal heat output [kW]	Average heat output [kW]	Total emitted amount of CO [mg]	Total emitted amount of NO _x [mg]	Emitted CO per 1 kWh of HE [mg]	Emitted NO _x per 1 kWh of HE [mg]	Average FG temperature [°C]
ethanol	322.3	2.38	–	4.43	2.50	194.6	314.1	81.8	132.0	280
methanol	317.1	1.76	–	3.29	2.39	280.1	96.8	159.6	55.1	232
ethanol/water (90/10)	324.9	2.17	–	2.97	2.21	311.9	215.8	143.5	99.3	256
ethanol/water (80/20)	335.8	1.97	–	2.36	1.77	452.7	148.3	229.6	75.2	229
ethanol/methanol (90/10)	319.6	2.30	–	3.63	2.46	193.2	282.9	84.0	123.0	283
ethanol/methanol (70/30)	318.4	2.15	–	2.85	2.30	187.7	239.6	87.5	111.7	276
ethanol/methanol (50/50)	318.4	2.04	–	2.71	2.18	176.7	225.5	86.8	110.8	276
ethanol	327.7	2.42	Cat A	4.25	2.74	34.8	321.2	14.4	132.8	263
methanol	316.7	1.75	Cat A	2.39	1.81	46.1	94.4	26.3	53.9	197
ethanol/water (80/20)	332.8	1.95	Cat A	3.03	2.06	39.5	147.1	20.2	75.3	219

while during both combustion tests with methanol, the lowest energy input was reached. The highest energy inputs were reached during the tests with ethanol. The highest value of maximal heat output and the average heat output were reached also during the combustion of ethanol, while the lowest values were reached during the methanol and ethanol/water (80/20) mixture (fuels with the lower LHV). These results comply with the previous study of Sileghem et al. [29] and the fuel properties described by van Loo and Koppejan [25] and Zink [26]).

Considering the total amounts of emitted CO, there are several obvious trends. The lowest amount of emitted CO per 1 kWh of HE was reached during the combustion test with the ethanol (reference fuel recommended by the producer of the burner). Mixing the ethanol with methanol in 90/10, 70/30 and 50/50 ratios slightly increased the value (by 2 – 6%). The significant increase was observed in the case of methanol (by 95%) and ethanol/water mixtures (90/10 — by 75%; 80/20 — by 180%) combustion. According to Zink [26], the main reasons for mentioned deterioration of FG composition were decreasing temperature of the flame caused by its cooling due to evaporated water, which flows through the flame (in the case of ethanol/water mixtures) and by combustion of fuel with lower LHV (in case of combustion and co-combustion of methanol with ethanol). Similar results were obtained during the ethanol/water mixtures combustion in spark-ignition engines by Sileghem et al. [29]. Adiabatic flame temperatures of ethanol/water mixtures presented by Breaux [14] also confirm the statement.

After the application of the catalyst, the amount of emitted CO has significantly decreased regardless of the used fuel. In the case of ethanol, ethanol/water (80/20) mixture and methanol combustion the reached values were decreased by 82%, 75% and 68%, respectively, in comparison to the situation of ethanol combustion without the catalyst. Catalyst caused a decrease by 83% in comparison to the methanol combustion (with and without the catalyst) and also caused a decrease by 90% in comparison to the ethanol/water mixture (80/20) combustion (with and without the catalyst). The reached values of so-called conversion rates, caused by the presence of the catalyst, corresponds with the conversion rates obtained by similar, precious-metal-based catalysts application in the FG formed during the solid biomass combustion purification, as was present by Ryšavý et al. [22], Klausner et al. [30] and Reichert et al. [31].

The highest emitted amount of NO_x per 1 kWh of released HE was reached during the combustion of ethanol. The lower results were reached during the methanol combustion (decrease by 60%) followed by ethanol/water mixtures (80/20 — decrease by 43%; 90/10 — decrease by 25%). Co-combustion of methanol with ethanol also caused lower values than in the case of ethanol combustion (50/50 — decrease by 16%; 70/30 — decrease by 15%; 50/50 — decrease by 7%). Comparable results were obtained during the ethanol/water mixtures combustion described in studies of Breaux [14], Sileghem et al. [29] and Li et al. [32]. Catalyst usage did not affect the values of emitted

NO_x. Only thermal and prompt NO_x formation can occur during nitrogen-free fuel combustion. The possible paths of their formation were described by Kiang et al. [33].

From the specific mass of emitted water vapour in the flue gas per 1 kWh of released energy into the ambient air point of view, the reached values of the fuels ranged, between 0.16 and 0.2 kg, while the lowest values were reached during the ethanol combustion and the highest values were reached during the methanol and ethanol/water mixtures combustion (including just water vapour formed during the combustion process or by evaporation of water in the fuel, combustion air moisture was not counted). Mentioned results were calculated according to the following equation Eq. (4):

$$m_{\text{H}_2\text{O}} = \frac{(8.94 \cdot w_H + w_w)}{LHV} \quad (4)$$

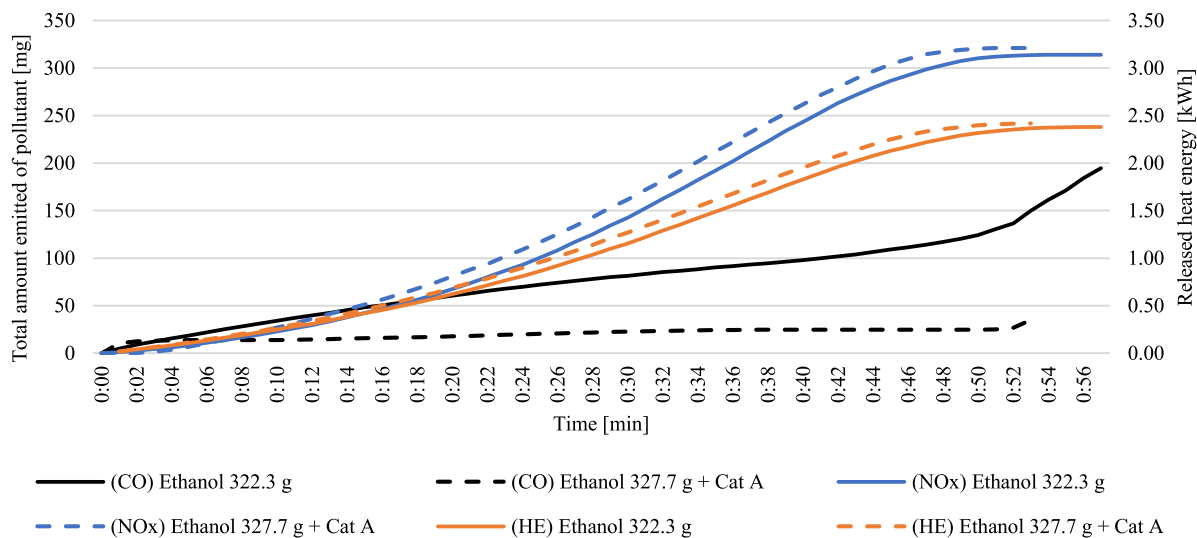


Fig. 2. The courses of increments of emitted CO depending on the burning time.

There are courses of the increments of emitted pollutants depending on the burning time for ethanol combustion (both regimes — with and without the catalyst) as well as the courses of increments of released energy during the combustion process in Fig. 2. Most of the emitted amount of CO was formed during the pre-heating and burnout phases (the phases are more detailed described in the study of Ryšavý et al. [5]). Oppositely, the most of the emitted amount of NO_x was formed during the main — heating phase, when, simultaneously, most of HE was released. There is an obvious influence of the catalyst on the total emitted amount of CO when primarily heating and burnout phases were significantly, positively affected. Pre-heating phase was not affected due to low FG temperature, inadequate for the proper functioning of the catalyst.

4. Conclusions

The main influence of different fuels based on ethanol, methanol and water mixtures was described from the total amount of emitted CO and NO_x. Fuels with the highest LHV represented the lowest emitted amount of CO and, on the contrary, fuels with the lowest LHV represented the lowest emitted amount of NO_x. Obtained results were connected with measured FG temperature. The positive influence of precious metals-based oxidation honeycomb catalyst on the FG composition from the total amount of emitted CO point of view was confirmed. The influence of individual combustion phases on the formation of pollutants has been described in connection to the amount of released energy during these phases.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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