REDUCTION OF CARBON MONOXIDE EMISSION FROM A SOLID-FUEL THERMO-ACCUMULATION FURNACE

by

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Many households in Serbia, using electric thermo-accumulation furnaces for heating, have been forced to find an alternative solution, due to a significant in crease in electric ity prices during the last de cade. A pos si ble so lu tion is replacing electric heating appliances with the solid fuel-fired ones. A proto type of a new concept of thermo-accumulation solid fuel-fired furnace has *been developed to meet these growing needs, providing electricity saving* together with considerable environmental benefits. Two strategies for reduction of carbon monoxide emission are examined in the paper: applica*tion of Pt/Al*² O_3 *catalyst, in the form of 3* \pm *0.3 mm spheres, providing further combustion of flue gases within the furnace, as well as an additional emission reduction by means of the air excess control. Experimental investigations of the catalyst influence on the conversion of carbon monoxide have* been done for different operation regimes and positions of the catalyst. The *pa per pres ents se lected re sults re gard ing car bon mon ox ide emis sion dur ing wood and coal combustion. Investigations suggest a considerable effect of the catalyst and a strong influence of the catalyst position within the furnace on carbon monoxide emission reduction. In addition, experimental tests have been conducted to assess the effect of the air excess control in the furnace on carbon monoxide emission. The amount of combustion air, the flue gas flow rate, and the fuel feed ing re gime have been ad justed in or der to keep the flue gas oxygen content in a relatively narrow range, thus obtain*ing controlled combustion conditions and lower carbon monoxide emission. In this way, the furnace has been made able to respond to the changes in *heating needs, fuel quality and other parameters, which is advantageous in comparison with similar solid-fuel fired furnaces.*

Key words: *thermo-accumulation furnace, solid fuel, emission reduction, catalyst, air excess control*

Introduction

Significant number of households in Serbia are heated with electric thermo-accumulation stoves, with individual power of $2-6$ kW. Due to the increase of electricity prices and limited electric power production capacities, electric heating is becoming very expensive for many households. The reserves of coal, which is the principal source of primary energy for electricity production in Serbia, are being exhausted gradually. These facts, as well as general trends in the society, give quite a few incentives to replace this type of heating with an alternative one. One of the possible solutions is replacing these electric appliances with solid fuel-fired thermo-accumulation furnaces. Utilization of solid fuel-fired thermo-accumulation furnaces, compared with electric stoves, has certain advantages. The possibility of electricity saving that can arise from replacing electric stoves with solid fuel-fired ones is of immense importance, since it could decrease the load of the power production system in Serbia. This is in accordance with energy saving tendencies in space heating [1], as well as with a requirement imposed by the measures of the European Union for achieving energy savings in households. For instance, $4,000.000 \text{ kWh/day}$ could be saved, and power production load lowered, especially during peak hours, by replacing 100.000 electric stoves, with 4 kW of electric power each, working 10 hours per day each. If all of these electric stoves are turned on at the same time (since the heat is accumulated usually during the low-tariff electricity period), their power would correspond to the power production of the 400 MW_e unit. Electricity saving implies significant indirect pollutant emission reduction from power production facilities, the most important being reduction of greenhouse gases emissions, above all carbon dioxide. Based on total emissions from Serbian power plants data $[2]$, by saving $4,000.000$ kWh of electricity per day, indirect carbon dioxide emission reduction of 3,228 t/day is achieved. Indirect emission reductions for nitrogen oxides, carbon monoxide $(2.56 t/day)$, and sulphur dioxide are also important [2].

Usual problems found in solid fuel household heating systems are low combustion efficiency, small heat transfer intensity, and high emission. The problems in residential heating are treated now adays from various aspects $[1, 3-5]$, with pollutants emission being one of the most important $[1, 4, 5]$. By proper organization of the combustion process, the efficiency of heating devices can be improved considerably [6]. Solid fuel-fired furnaces can utilize, apart from coal, various kinds of biomass and waste, which is a renew able energy source. The biomass potential of Serbia is very important. Using biomass, as a renewable energy source, is of wide social importance, both for the savings that may arise from it, as well as from environmental reasons, which include complying with Kyoto Protocol requirements.

For all these reasons, a prototype of an innovative concept of solid fuel-fired thermo-accumulation furnace, aimed for residential heating purposes, has been developed and tested in Laboratory for Thermal Engineering and Energy of the VINČA Institute of Nuclear Sciences in Belgrade. The furnace provides high efficiency, considerable energy savings, emission of pollution that meets European standards and possibility of firing both low- and high-rank solid fuels, (biomass, coal, *etc.*) with simultaneous firing of different solid fuels, when needed. Development efforts have been focused on three concepts of the thermo-accumulation solid fuel-fired furnace – the basic concept, the furnace with water heating and the high-efficiency concept with the catalyst and a simple system for air excess control, with the aim to find the influence of different flow and combustion conditions on the furnace efficiency and emission. The basis for development of the furnace was a registered patent $[7]$. The furnace is presented and described in details

in [8]. Furnace design enables good conditions for complete combustion – the heat losses due to chemical incompleteness of combustion and the heat losses with the flue gases are kept low, as well as flue gases emissions. For the basic concept of the furnace and the furnace with the heat exchanger, the efficiency coefficient exceeds 70% [9, 10], and for the high-efficiency concept of the furnace it exceeds 80%, for both wood and coal as a fuel. The efficiency coefficient of the household solid fuel-fired thermo-technical devices produced in domestic factories rarely exceeds 50%. The furnace design, with the special convective air ducts between furnace chamotte walls and the outside wall made of ceramic tiles, makes heating much more pleasant, due to increased convective heat transfer and lower outside wall temperature (lower radiative heat transfer to the heated room). This contributes to diminishing the unpleasantries linked to this type of heating facilities [11], and also enables uniform space heating - a problem very commonly addressed nowadays $[12-14]$ – by combining radiative and convective heat transfer.

Thorough tests of the furnace basic version [15] and the version with the heat exchanger in the third draft [16] were previously carried out. These tests proved that furnace design enabled excellent combustion conditions and an efficient transformation of toxic carbon monoxide into carbon dioxide. The results obtained in the experiments with the heat exchanger served as a basis for development of a heat transfer and flow model in the heat exchanger zone [8].

Two strategies for further reduction of carbon monoxide emission are examined in this paper: application of Pt/Al_2O_3 catalyst, in the form of 3 0.3 mm spheres, providing further combustion of flue gases within the furnace, as well as an additional emission reduction by means of the air excess control. The tests were done for different operation regimes to evaluate the catalyst influence on the conversion of carbon monoxide during wood and coal combustion, and also to assess the influence of air excess control on furnace performance and its environmental impact. The oxygen content in the flue gases was continuously monitored with the help of a data acquisition system, and combustion air flow rate was adjusted accordingly, so as to keep the oxygen content in a relatively narrow range (approximately 13%). These tests were done in accordance with the European Standard for solid fuel-fired furnaces (EN 12815) [17].

Experimental set-up and operating conditions

The furnace operating at different regimes, firing wood and coal, with and without catalyst, has been experimentally tested. The Pt/Al_2O_3 catalyst is used, in the form of 3 0.3 mm spheres. The support used for the catalyst is commercial ($\gamma + \theta$) Al₂O₃ from Rhone Poulenc in the form of spheres. The platinum catalyst has been prepared by impregnation of a dry support with aqueous solution of chloroplatinic acid [18]. The furnace and the experimental set-up are shown in fig. 1. The flue gases flow through the three drafts $(1a, 1b, 1c)$, while combustion of solid fuel takes place on a horizontal grate (2) . The air enters the furnace through the opening under the grate. With respect to the catalyst efficiency diagram (the temperature dependence of the catalyst's capability for removing certain flue gas components) [19], pre-tests have been performed to find the place for op-

Figure 1. The solid fuel furnace with the catalyst and the experimental set-up

timal catalyst installation. The pre-tests have shown that the temperature range of the catalyst optimal efficiency for carbon monoxide conversion corresponds to the flue gas temperatures measured in the central draft (1b), and therefore this draft has been chosen as the most appropriate place for catalyst positioning. The first draft (1a) could have not been selected in any case, since the combustion with intensive flame takes place here. In order to examine the influence of catalyst position on its ability to reduce carbon monoxide emission, the catalyst (3) has been placed in two vertical positions (denoted as *upper* and *lower*), within the central draft (1b), with flue gases flowing downward and around the catalyst. The flue gases leave the furnace through the stack (4), used for flue gas sampling and temperature and flow rate measurements. The airflow rate is controlled by changing the flue gas fan speed. This fan is connected to a variable $(0-220 \text{ V})$ AC supply (5). During the experiments, when the furnace reached a steady-state (after the initial firing), the AC voltage has been kept constant at 110 V. This has enabled the whole amount of fuel (about 1.5 kg) in the combustion chamber (1a) to burn out until the next fuel feeding, in one hour.

Gas samples have been taken from the stack (4) and continuously analyzed. The gas sampling probe, made according to EN 12815 Standard [17], has been water-cooled on the outside of the stack, in order to dry the flue gas samples. The gas sampling line has been also equipped with vessels for collecting the condensate and filters for protection of gas analysers from particles and tar in the flue gases. The gas analyzers are controlled by a PC, memorizing measured concentrations data every three seconds. Flue gas temperatures have been measured with type K, class I thermocouples, at a number of points, like at the top of the central draft (t_1) , at the catalyst entrance and exit sections $(t_2$ and $t_3)$ and at the furnace $ext{exit}(t_4)$. Thermocouples signals have been measured with a digital voltmeter on the HP3852a data acquisition system, and memorized every 5 seconds. The furnace as

a whole has been placed on a high-precision balance $(10 g)$, in order to monitor the weight loss between consecutive fuel feedings, *i. e.* the combustion dynamics.

Experimental tests have been done also in order to examine the influence of air excess control on the furnace environmental impact, primarily the emission of carbon monoxide. In order to additionally decrease carbon monoxide emission, the Pt/Al_2O_3 catalyst (3) in the upper position has been used in one of the experimental regimes. For precise air flow rate control, an additional fan has been applied, which has blown the air through the ash bin opening, under the grate (2) . This fan has been connected to a variable DC source $(0-12 \text{ V})$. Measurements have been performed in three different operating regimes of the furnace, with wood used as a fuel. During experiments in the basic regime without air excess control (Regime W1), when the furnace reached a steady-state (after the initial firing period), the AC voltage has been kept constant, which has enabled the whole amount of fuel in the chamber to burn out until the next fuel feeding, in one hour, with only incandescent char left on the grate. In the regime with air excess control (Regime $W2$), the air flow rate has been adjusted primarily by changing the AC voltage, supplying the flue gas fan, in a very wide range, in accordance with the values of oxygen content in the flue gases, measured by the gas analyzers. A smaller, additional fan, blowing the air under the grate, has been used for fine control of the air flow rate. The furnace has been operating without the catalyst in regimes W1 and W2. In Regime W3 (air excess control with the presence of the catalyst in the central draft), the air flow rate control has been performed in the same manner as in Regime W2. The catalyst has been put in the upper position, as depicted in fig. 1. The average fuel consumption in the three regimes has been $B = 1.5$ kg/h. In order to obtain a basis for the planned development of an automatic air excess control system, the λ -Probe (zirconia probe) signal, in volts, has been also measured with a digital voltmeter on the HP3852a data acquisition system and memorized every 5 seconds. The λ -Probe has been installed at the top of the central draft. In all regimes of operation, the fuel has been fed to the furnace in shorter intervals – one hour or 30 minutes, found to be optimal from the pre-tests of the furnace. Furnace operation has been continuously monitored for several hours with the data acquisition system.

Results and discussion

Measured carbon monoxide concentration values in dry flue gases are converted to concentration values at reference oxygen content in the flue gases, according to eq. (1):

$$
CO_{\text{ref}} \quad \frac{20.9 \quad O_{2\text{ref}}}{20.9 \quad O_{2\text{meas}}} CO_{\text{meas}} \tag{1}
$$

The reference oxygen content in the flue gases, according to the EN 12815 Standard, is $O_{2ref} = 13\%$. The carbon monoxide concentration at reference oxygen content is converted to mg/ Nm^3 as follows:

$$
CO_{\text{ref}}[\text{mg}/\text{Nm}^3] \quad CO_{\text{ref}}[\text{vol.}\%]10,000 \quad 125 \tag{2}
$$

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Released carbon-monoxide in time is calculated as:

$$
\dot{m}_{\rm CO} \quad \dot{V}_{\rm fg} \,[\rm m^3/s] \quad CO_{\rm ref} \,[\rm mg/Nm^3] \tag{3}
$$

Mass of carbon monoxide released in 3 seconds is.

$$
m_{\rm CO}(3\,\mathrm{s}) \quad 3\dot{m}_{\rm CO} \tag{4}
$$

Since gas samples analysis data are memorized every three seconds, the cumulative carbon monoxide emission from the beginning of the test until the moment observed $\tau - E_{\text{CO}}(\tau)$ is calculated by the expression in eq. (5):

$$
E_{\rm CO}(\tau) \frac{1}{1000} \int_{0}^{\tau} m_{\rm CO}(3 \,\mathrm{s}) \tag{5}
$$

The furnace with the catalyst – wood combustion experiments

The furnace was examined in three operation regimes, with fuel consumption of about 1.5 kg/h in each. For the basic one (without the catalyst), wood was supplied to the furnace every 30 minutes (0.750 kg) in the beginning, but it turned out that this amount of fuel was optimal for 40 minutes of operation and fuel feeding was adjusted accordingly. In the other two regimes, the catalyst was mounted in the upper and the lower position, respectively. The flue gases temperature at the catalyst entrance section and carbon-monoxide concentration in the outlet flue gases at 13 vol.% O_2 , changing in time, are shown in figs. 2 and 3, for both operation regimes with the catalyst. Experimental results for the effect of the catalyst on carbon-monoxide concentration in the flue gases at 13 vol.% O_2 (averaged over fuel feeding intervals) and on cumulative carbon-monoxide emission, are given in figs. 4 and 5. Compared to the basic one, both regimes with catalyst are obviously advantageous. After a quick and very intense increase immediately after fuel feeding, carbon monoxide concentration decreased very soon to the value of 0.8 vol.% (figs. 2) and 3), permitted by the EN 12815 Standard. Average carbon monoxide concentration (fig. 4) in the case of the catalyst in the upper position is around 0.4 vol. $\%$, *i. e.* much lower than the permitted value. Further enhancement was obtained by mounting the catalyst in the lower position, when presence of the catalyst completely removed carbon monoxide for significant period of time between two fuel feedings (fig. 3) and average carbon monoxide concentration was about and bellow 0.2 vol.% (fig. 4). Positive effects of catalyst on carbon monoxide emission reduction are even more obvious when considering the cumulative carbon monoxide emission during the experiments (fig. 5). Cumulative emission after 200 minutes of operation for the catalyst in the lower position was 5

 -1.0 Gas temperature [°C] 400 $-0.8\frac{1}{5}$ 350 ition 0.6 300 $\begin{bmatrix} 1 & 1 \\ 0 & 0 \\ 0 & 0 \\ 0 & 0 \end{bmatrix}$ concentrate 250 200 -0.0 150 50° 100 200 250
Time [min.] $\dot{0}$ 150

 \bullet CO

Temperature

 450

Figure 2. Time dependence of the flue gases temperature at the entrance section of catalyst in the *upper* **position and CO concentration in the outlet flue gases, for wood combustion**

Figure 3. Time dependence of the flue gases temperature at the entrance section of catalyst in the *lower* **position and CO concentration in the outlet flue gases, for wood combustion**

Figure 4. The catalyst effect on the average CO concentration in the outlet flue gases, for wood combustion

Figure 5. The catalyst effect on the cumulative CO emission, for wood combustion

times lower compared to the basic regime, and for the catalyst in the upper position was cut to one half.

The catalyst proved to be more efficient when placed in the *lower* position than in the *upper* one (in the zone with higher temperatures). The temperature interval of maximum catalyst efficiency for carbon-monoxide conversion is at temperatures above $200-225$ °C [19], while figs. 2 and 3 show that for both regimes with catalyst the temperatures at the catalyst entrance section have been also higher than these, most of the time.

Thus, better efficiency for the catalyst in the *lower* position can not be assigned to temperature, but most probably to the flow conditions. In this regime, the flue gases enter the catalyst zone and flow over catalyst beads uniformly, so the catalyst is utilized more efficiently. The catalyst in the *upper* position disturbs flow and temperature field in the central draft considerably and it is likely that an intense recirculation zone exists above the catalyst zone.

The furnace with the catalyst – coal combustion experiments

The experiments were carried out for two operation regimes, the basic one (without the catalyst) and the regime with the catalyst in the *upper* position. The amount of air supplied, in both regimes, was sufficient for burning out 1.5 kg/h of coal. In the basic regime 1.5 kg of coal was supplied once an hour. It was noted during the experiment that in time the grate became covered with a huge amount of unburnt fuel and ash. In order to avoid this in the regime with catalyst, the furnace was supplied with 0.750 kg of coal every 30 minutes.

Carbon monoxide concentration was low, almost zero, for most of the time between fuel feedings (fig. 6). The flue gas temperature at the catalyst entrance section did not change considerably, in the range of 320-365 \degree C, and probably did not affect carbon monoxide catalytic conversion much. The results for cumulative carbon monoxide emission (fig. 7) showed that presence of the catalyst, even in the upper position (less suitable for reducing carbon monoxide), significantly contributed to lowering carbon monoxide concentration levels in the flue gases. After 120 minutes of operation, carbon monoxide emission was 20% lower with the use of the catalyst.

Figure 6. Time dependence of the flue gases temperature at the entrance section of catalyst in the *upper* **position and CO concentration in the outlet flue gases, for coal combustion**

 Figure 7. The catalyst effect on the cumulative CO emission, for coal combustion

The furnace with the air excess control – wood combustion experiments

In the basic operation regime – W1, less air than needed was supplied into the furnace deliberately, in order to show that an already proved efficient furnace, if not operated the correct way, can give non-satisfactory results, regarding carbon monoxide emission and combustion quality. In the beginning of this regime, wood was supplied to the furnace every 30 minutes (0.750 kg) , in order to avoid the grate to be completely covered with fuel and "choked". It turned out during the experiment that this amount of fuel was optimal for 40 minutes of operation, and fuel feeding was adjusted accordingly. Measured (fig. 8), as well as average values (fig. 9) of carbon monoxide concentration in this regime of operation, where the furnace was deliberately supplied with lower amounts of air than needed, were at times somewhat higher than $1 \text{ vol.}\%$ (0.8 vol.% is permitted by the EN 12815 Standard [17]). This points to the importance of correct furnace operation (guiding of the combustion process). Concentration values shown in fig. 9 were averaged over periods between fuel feedings.

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For assessing the influence of combustion air flow rate control on carbon monoxide emission from the furnace, investigations were done in the Regime W2 (air excess control without the catalyst) and the Regime W3 (air excess control with the catalyst). During these experiments, the aim was to keep the oxygen content in the flue gases in a relatively narrow range (11-14%). This equals to air excess coefficient values of $2.1-3$, which were during the pre-tests found to be optimal for furnace operation, with respect to combustion quality and carbon monoxide emission. For most of the time between two fuel feedings, flue gas oxygen content was about 13% (equal to 2.33 for air excess coefficient). In both regimes, average carbon monoxide concentration in the flue gases, converted to 13% O_2 , was lowered to the values significantly under 1% (fig. 9), which proved that air excess control contributed to the furnace performance. Previous investigations $[20, 21]$ have also shown that furnace, if operated properly, completely fulfills European environmental norms and requirements.

Figure 10. Cumulative CO emission in different regimes of operation

The comparison of the influences of air excess control and catalyst installation is easier to be carried out by observing the cumulative carbon monoxide emission during the experiments (fig. 10). The air excess control itself considerably reduces carbon monoxide emission, when compared to Regime W1, and this influence becomes more and more obvious during the experiment. The presence of the catalyst improves the effects of air excess control and reduces carbon monoxide emission further. After 200 minutes of operation, the cumulative carbon monoxide emission in Regime W2 was 30% lower than in Regime $W1$. By placing the catalyst in the central draft, an ad-

ditional 10% decrease of carbon monoxide emission was achieved, at the same moment in time.

Conclusions

A prototype of an innovative concept of efficient and environmentally friendly thermo-accumulation solid fuel-fired furnace, aimed for household heating purposes, has been developed to meet the growing needs for electricity savings together with environmental benefits. The furnace enables the possibility of firing both low- and high-rank solid fuels.

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Two strategies for reduction of carbon monoxide emission are examined in the paper: application of Pt/Al₂O₃ catalyst, providing further combustion of flue gases within the furnace, as well as emission reduction by means of the air excess control. A series of experiments considering the influence of the catalyst have been performed on the furnace prototype firing wood and coal. Measured and average values of carbon monoxide concentration, as well as cumulative carbon monoxide emission, have shown that the presence of the catalyst considerably reduces carbon monoxide emission, for both fuels used. Investigations also suggest a strong influence of catalyst position on carbon monoxide emission reduction, as well as the importance of the flow conditions within the furnace. The furnace has been experimentally tested also with the aim to assess the effect of the air excess control on the combustion efficiency and carbon monoxide emission reduction. The tests have shown that air excess control significantly contributes to carbon monoxide emission reduction, by optimizing combustion conditions in the furnace. The presence of the catalyst in the central draft helps in decreasing carbon monoxide emission further. These investigations can also help in development of an automatic air excess control system, by using the signal of the zirconia probe, installed in the furnace.

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Nomenclature

Greek letters

 τ – time, [s]

Subscripts

 $-$ flue gases fg - measured meas

 ref - reference

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