Toxic substances formation in co-incineration process for food production

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Abstract

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Introduction. The research aim is to define the influence of the using hydrogen combustion technologies on existing or upgraded boiler equipment in food technologies and the choice of rational ways to use the modernized fuel and energy complex equipment for operating on new fuel, in compliance with the industrial and environmental safety requirements.

Materials and methods. General scientific methods and special methods were used. The laboratory unit for experimental process at the semi-lamina front follow-up is created for research the role hydrogen in co-firing with natural gas, depends on concentrations of oxide and nitrogen dioxide in combustion products.

Results and discussion. Separate NO and NO₂ measurements showed that the dependence of the NO₂ fraction on the H₂ content in the solution, supplied to the laminar methane flare, is ambiguous and significantly depends on the sampling location at the flame front cut section of the.

An intensive increasing the nitrogen oxides concentration is observed in the initial cut flame front sections from the moment where the mixture start the heating to the maximum temperature.

Moreover, the higher the H₂O₂ solution concentration is injected into the flame front (similarly, the smaller the hydrogen proportion in the solution), the more intense the increase in the nitrogen oxides content.

When a 30% H₂O₂ solution injected into the flame front, the NO concentration practically reaches the initial one, which obtained without the drip moisture injection (90–95 mg/m³). At the same time, the H₂O injection into the flame front leads to a decrease in NO concentration by 35–40%, up to 65 mg/m³. This can be explained by the fact that hydrogen peroxide forms the OH radical, which is actively involved in the hydrocarbons and CO oxidation, more intense than water.

The OH radical forms hydroperoxide-radical HO₂ from H₂O₂, which dissociates with the superoxide ion O₂-formation and gives the H-radical from H₂O₂.

Conclusions. With increasing hydrogen content in the solution the NO_x formation decreases, which can be explained by the oxygen consumption in fast reactions that require less energy.

Introduction

There are a variety of ways for the food processing industry to reduce their manufacturing costs with energy saving methods and technologies.

While many lab-scale investigations have been carried out on the thermochemical conversion, commercial production and utilization of biofuels and or chemicals are still at an early stage (Chengfang Song *et. al.*, 2020; Myronchuk V. *et. al.*, 2019). Several projects have been launched to demonstrate the potential of hydrogen at an industrial scale (Yue M. *et. al.*, 2021). Although the role of hydrogen in the industry has been widely assessed over the last two decades, and considering that the heat sector is highly energy consuming, significant adaptations in production processes are necessary, requiring a longer study period. (Guilbert Damien *et. al.*, 2021)

In the work (Jaruwan Wongthanate *et. al.*, 2020) it was describe of producing hydrogen from food and beverage processing wastewater by anaerobic microflora enriched of starch versus in a batch reactor. Besides authors (Sang Eun Oh *et. al.*, 2005) showed that hydrogen production from a food processing wastewater high in sugar can be linked to electricity generation using a microbial fuel cell to achieve more effective wastewater treatment.

Some food processing companies are using organic waste from their facility and convert them into fuel for the phosphoric acid fuel cell at their facilities. Many of these companies use proven technology such as anaerobic digestion to convert organic waste and manure into hydrogen to power the fuel cells (S. Curtin *et. al.*, 2017) Therefore, converting food waste into biofuels, bio-based fertilizers, bio-based enzymes, chemicals, proteins and other bio-based molecules and materials will accelerate the sustainable development goals. (Tsegaye B. *et. al.*, 2021)

Fuel cell technology is a maturing technology that has been implemented in the food industry for several years (Ping Wu *et. al.*, 2018).

It well known that hydrogen is combined with compounds such as oxygen, natural gas, coal, and oxygen-carbon from plant biomass (Guilbert Damien *et. al.*, 2021). Consequently, as part of strategy to use hydrogen, need to gradually change the composition of existing gas infrastructure by blending hydrogen and natural gas. For instance, is targeting 20 to 30% blends of hydrogen and natural gas in the short term. (Abbas A.J. *et. al.*, 2021). Therefore, in addition to the determination of the amount of hydrogen gas that would deliver equivalent energy contents as natural gas, accurate investigation of the effect of fluid and flow properties such as the compressibility factor, and velocity on the safe and efficient transportation of hydrogen and hydrogen blends is important as they influence gas flow in the pipelines (Andre J. *et. al.*, 2014).

While current appliances may be compatible with natural gas that is enriched with up to 20 per cent hydrogen, higher percentage blends of hydrogen will require new appliances or significant upgrades to existing appliances specifically in relation to burner designs (Orana Sandri *et. al.*, 2021).

The research aims to define the influence of the using hydrogen combustion technologies on existing or upgraded boiler equipment and the choice of rational ways to use the modernized fuel and energy complex equipment for operating on new fuel, in compliance with the industrial and environmental safety requirements.

To achieve this aims it is necessary to solve the following tasks:

1. Develop a concept and laboratory unit for the experimental investigation the hydrogen and natural gas mixtures combustion.
2. Conduct the experimental research with the aim of determining the effect of changes in hydrogen content by adding hydrogen peroxide drops with different concentrations to

laminar flame front of natural gas and compare with the same volume, number and technology for using distilled water.

3. Determine the dependences that characterize the hydrogen combustion in a mixture with natural process gas and propose the recommendations for the technology of possible hydrogen mixtures in boilers combustion adaptation.

Materials and methods

The object of study was the process of combustion of the hydrogen and natural gas mixtures. The subject of the research was determination of the nitrogen oxides formation by the hydrogen effect on the combustion process.

Chemicals

The hydrogen peroxide solution by the various concentrations from 0% up to 30% was used for experimental investigations.

Gas and gaseous fuel was used for experimental investigations.

Water and steam was used for experimental investigations.

Experimental installation

An experimental installation is designed and manufactured for studying the moisture effect on the toxic substances formation during fuel combustion and described in detail in (Sigal A. *et. al.*, 2021) and several experiments series conducted with it. The unit described (Sigal A. *et. al.*, 2021) was taken as a base, and a device for hydrogen peroxide solutions preparation and a peristaltic pump for supplying moisture to the flame front was installed.

The experimental installation (Figure 1) consists of a gas duct, mounted on the frame, equipped with air supply systems, gaseous fuel and steam supply systems, heating system that can maintain the required temperature, gas burner device, drip moisture to the flame front supply system, gas sampling probe precision movement system, thermocouple and capillary for drip moisture supply and combustion process control system. The frame provides the experimental unit rigidity and stability in general.

The flue is made from a heat-insulated cylindrical pipe that made heat-refractory material with 16.0 mm diameter and 4.0 mm wall thickness, and has an attached fan to supply air for combustion. A diaphragm with a micro-manometer for measuring airflow is installed in the horizontal section. A nozzle for gaseous fuel and steam supply and an electric heater to heat the mixture to the desired temperature are mounted in the vertical section. A gas burner device is mounted at the end of the vertical flue section. The gas supply was carried out through a 10.0 mm diameter copper tube in which a measuring diaphragm with a micro-manometer (type INN-1) is mounted for measuring gas flow. Precision needle valves are installed in the appropriate communications to regulate air and gas flow. Steam generator connected to a flue by 10 mm diameter copper heat-insulated tube is used for steam production. The gas distribution device, with holes 0.8-1.0 mm diameter, provided gas and air complete mixing.

Figure 1. Experimental installation:

1 – gas duct; 2 – peristaltic pump; 3 – flame; 4 – flowmeter;
5 – drip moisture to the flame front supply system; 6 – combustion process control system

Research method of nitrogen oxides formation

The hydrogen effect on the nitrogen oxides formation researched directly in the flame front at the laboratory on a stand with precision measuring instruments. The measurements scheme is shown in Figure 2.

In experimental studies, the main attention was paid to the maximum local temperature and nitrogen oxides concentrations that was measurement in the "trace" of the drop, which contained hydrogen peroxide different concentrations from 0 to 30%. The experimental research of the normal combustion front was carried out in a stationary "reverse" flame, which is formed by continuous mixture supply to the burner after a steel cylindrical stabilizer ($d = 3.0$ mm). Previous research, carried out by authors, showed that within the stabilizer diameters from 1 to 5 mm, the flame front elementary structure did not depend on the one's diameter. When changed up α to 1.8, the flame front width did not exceed 2.5 mm, the mixture flow parameters corresponded to $u = 1500$ mm / s, $Re = 250$.

Control of the main parameters in the process experimental research included: measuring the position of the microprobe, thermocouple and capillary for the droplets injection, the all components of the mixture for combustion flows, and the combustion products gas analysis.

Figure 2. Measurements scheme:

- 1 – Flame front; 2 – Combustion products; 3 – Drop “face”;
4 – Cold mixture; 5 – Stabilizer; 6 – Burner cut; 7 – Drop.

Research methods of the measuring the position of the microprobe thermocouple and capillary for the droplets injection

Micromanipulators arrow indicators of stereotactic unit with value of division 0.1 mm controlled the microprobe, micro-thermocouple and capillary for moisture supply geometric location. The manipulators device allowed moving the probe and the micro-thermocouple fixed in them, in three planes and provided angular movements in two planes. The remote control minimized the vibration possibility in the micro-sensors movement controlling process.

Therefore, stereotactic unit allowed selecting a step equal to 0.05 mm in the direction, perpendicular to the flame front, for probing the flame front. The coordinate's equality in this direction for the micro-thermocouple junction and the probe enclosure of the (subject to a correction of 0.1 mm through the perturbation zone) was not violated. The flame front was flat, which precluded measurement errors due to mismatch between the probe and thermocouple coordinates in a parallel to the front plane direction.

Research method of the all components of the mixture for combustion flows

Peristaltic pumps, which were connected to the moisture source and capillary by silicone rubber hoses, were used to supply drip moisture to the flame front. Moisture-capillary capillaries were made of steel X10CrNiTi18-10 and had 0.8 mm outer diameter. Peristaltic pumps were equipped with a flow regulator, which stabilized the moisture flow at a given value. The flow was chosen to compensate the moisture evaporation in the flare, and to keep surface volume and evaporation of the drop injected into the laminar flame front.

Statistical data processing

Data were expressed as means $\pm$ standard deviations for triplicate determination. The measurement error value due to inaccurate probe and thermocouple relative to this point of flame fixation did not exceed 5% of the flame front width. The flams' opening angle was taken into account during the experimental data processing. Thus, data on the temperature and concentration fields' distribution over the flame front width were obtained. All statistical procedures were computed using the Microsoft Excel 2007 software. Differences were considered to be significant at a validity of $\alpha = 0.95$.

Results and discussion

Investigation of the hydrogen peroxide injection

The various concentrations (up to 30%) hydrogen peroxide solution was fed in the drop of liquid form into the flame laminar methane flame front. The obtained results testified to the equidistant character of the concentrations distribution dependences during the distilled water injection.

The moisture influence in the fuel combustion process was previously studied at the experimental stand developed by the authors (Klimenko V.M. *et. al.*, 2008). In those studies, the various volumes of distilled water drops injected into the methane combustion zone (laminar flame). According to the conclusions, the moisture added to the combustion process with the initial products can be divided into moisture, which is involved in the chemical fuel oxidation process and which is not involved.

Fig. 3 shows the following dependence between the nitrogen oxides concentration and the hydrogen content in an aqueous hydrogen peroxide solution: with decreasing hydrogen content in the solution, and, accordantly, increasing the oxygen content, nitrogen oxides concentration increase. Authors (Soroka B.S. *et. al.*, 2020) attempted to describe household burners on the possibility of their operation with a methane-hydrogen mixture in the range of H_2 from 0 to 50%.

The obtained results and conclusions show the possibility of applying the studied methane-hydrogen ratios on domestic gas stoves. However, these results could hardly be scaled either for industrial burners or for burners installed in existing boiler units in general, due to differences in technical characteristics.

Investigation of the nitrogen oxides formation

The increasing the nitrogen oxides formation in atmosphere during the combustion hydrogen and hydrogen mixtures is well known problem (Castilla-Martinez C.A. *et. al.*, 2021).

This can be explained by the assumption that the growth of the thermodynamic component for these ratios dominates over the kinetic. In Figure 3 dashed line (beginning at point A') shows the probable expected nitrogen oxides concentration value flame front cut for the $\delta = 0.511$ mm.

However, a series of experiments performed in this section showed a point A that is deviates. At the same time, it lowers the temperature in the combustion zone, which leads to a decrease the nitrogen oxides concentration. In addition, the water molecule disintegrates into hydrogen (H_2) and oxygen (O_2), if the energy of flame is sufficient. The released hydrogen is involved in combustion reactions, that is, in fact the process we are studying (Klimenko V.M. *et. al.*, 2008).

Figure 3. The NO_x content in the combustion products depending on the hydrogen content in the solution

The authors suggest that this area, approximately the middle of the flame front, is a relevant area for further study with the aim of more accurately explains the deviations. The difference between the obtained and expected nitrogen oxides values is 10.5% and is equal to 6–8 mg/m^3 .

In the results of (Sigal O.I. *et al.*, 2020), the authors found that at temperatures, less than 1000 K the nitrogen oxides concentration is close to 1011 $\text{cm}^3/\text{mol}\cdot\text{s}$, but at temperatures more than 1000 °K increases sharply, the CO combustion intensifies and the reaction thermal effect increases. That is, increasing the H_2O_2 solution concentration to 30% and, accordingly, reducing the hydrogen content in the solution to 9.44% led to increase in the nitric oxide total yield by 35–45% for these experimental conditions. This may be due to more intensive OH and OH_2 radicals' formation and an increasing the temperature in the CO afterburning.

For chain radical reactions in a hydrogen-oxygen mixture at $T > 1000$ K, the branching mechanisms are relatively independent of the nature of the fuel. However, at temperatures below 1000 K, that is traditional for most boilers, they become much more complicate and depend on the fuel nature (Warnatz, J. *et al.*, 2006).

Experimental studies expectedly showed that with increasing hydrogen content, higher temperatures are reached. However, it is impossible to estimate the effectiveness of the additional substance (ballasts) to the combustion process by calculating their thermodynamic characteristics only. It is also necessary to take into account the process kinetics. For example, in (Klimenko V.M. *et al.*, 2008) the ballasts, which are often added during staged

combustion, such as excess air and steam are compared. The authors showed that with increasing maximum local temperatures of the process, the efficiency of the addition thermodynamically different ballasts converges. This can be explained by the assumption the dominance of kinetic aspects over thermodynamic ones.

Separate NO and NO₂ measurements showed that the dependence of the NO₂ fraction on the H₂ content in the solution, supplied to the laminar methane flare, is ambiguous and significantly depends on the sampling location at the flame front cut section of the.

In the anterior front border (Figure 4, graph 1) the NO₂ share reaches 78% of the maximum at a 9.85% hydrogen content in solution (corresponding to concentration of H₂O₂ solution 22%), reaches the limit values and remains unchanged with further hydrogen in solution reduction that is equal to increasing in the hydrogen peroxide solution concentration. In the average front cut section (fig. 4), graph 2 has its extremum, which corresponds to a hydrogen content of 0.63 in the ordinate. That is, a decreasing the hydrogen concentration in the solution (increasing the H₂O₂ concentration to 15%) for this cut flame front section is also leads to a significant increase in the NO₂ proportion. However, a further increasing the hydrogen peroxide percentage leads to a decreasing the ratio to 0.3, which roughly corresponds to the hydrogen content effect of 9.44% in the solution (3% H₂O₂ solution). Graph 3, corresponding to the point with coordinate 2.0, equidistantly repeats graph 2, but with less pronounced maximum at the point with the coordinate where the hydrogen content in the solution is about 9.4%.

Figure 4. The hydrogen content in the solution effect on the NO₂ formation in the laminar methane flare flame front, where:

- 1 – anterior front border, coordinate $\delta = 0,0$ mm;
- 2 – average front cut section, the coordinate $\delta = 1.0$ mm;
- 3 – rear front boundary, coordinate $\delta = 2,0$ mm.

Thus, temperatures above 800 K significantly affect the reaction rate and change the kinetics of processes. This occurs with the participation of hydrogen in a hydrogen peroxide solution. Moreover, in different front cuts, these changes are different by nature, which can be explained decreasing in the H_2O_2 existence time in the flame to 20 ms or less.

It is also important that the influence degree of hydrogen, which is present in the hydrogen peroxide solution, on the NO to NO_2 oxidation process for different temperatures have its extremum in the added substance concentration. Figure 4 shows that for this maximum NO_2 , the H_2 content in solution is about 9.4%.

Calculation and experimental research allow determining the influence degree of the moisture content in the initial components on the hydrocarbon fuels combustion. The initial moisture in the fuel-air mixture should be about 8-9% by combustion products weight. This fact underlies the idea that the moisture content in the initial products influence on the combustion process insignificantly.

Experimental data in this work were obtained for the air excess coefficient of $\alpha = 1.1$. These data illustrate that the nitrogen monoxide content increase at the beginning of the flame front and decreases behind the flame front when the combustion products temperature reducing because of the dilution and recombination reactions.

The nitrogen monoxide concentration decreases depending on the temperature and approximately repeats the changes of last one with some delay. These results have a good agreement with the results in (Sigal I.Ya. *et. al.*, 2020) by the form of temperatures and concentrations absolute values curves and also confirm the assumption, made as a result in analytical studies (Sigal A. *et. al.*, 2021), about the mismatch between the maximum temperatures and the maximum concentrations zones.

Mostly just such a dual role moisture explains the diversity and discrepancy in data on its effect on combustion process, resulted in various research (Klimenko V.M. *et. al.*, 2008; Sigal A. *et. al.*, 2021). There are even opposing views to this problem, from the combustion process intensification to an increase in the inefficient ballast volume. It can also be assumed that such common methods as combustion products recirculation to the flame root and flame turbulence in diffusion burners, which has the same function, provide flame root fertilization with the first type of moisture. The kinetic studies lead to a simplified kinetic scheme for NO_x formation and reduction under conditions of sludge combustion. (C. Philippek *et. al.*, 1997)

In (Sigal I.Ya. *et. al.*, 2020) for burning gas-oil emulsions at the gas-turbine units, the steam injection used as an additive. Calculations performed for the following values of relative between the fuel consumption and injected steam: $G_s / G_f = 0; 0.02; 0.053; 0.11; 0.33$ and 1.0, where G_s , G_f – weight consumption of steam and fuel respectively. With increasing the steam injection, the combustion zone is stretching in some measure and the maximum local flame temperature decrease by 80-100 K. After adding a small amount of H_2O , the CO content may decrease, and with a large amount, it may increase. The fuel NO concentration decreased by approximately 7%, which shows a weak effect of moisture additives on the fuel nitrogen oxides formation. An intensive increasing the nitrogen oxides concentration is observed in the initial cut flame front sections from the moment where the mixture start the heating to the maximum temperature. Incomplete combustion processes lead to the formation of many gaseous byproducts that can be challenging to monitor in flue gas released via chimneys. (Magnus Galfalk *et. al.*, 2018),

Moreover, the higher the H_2O_2 solution concentration is injected into the flame front (similarly, the smaller the hydrogen proportion in the solution), the more intense the increase in the nitrogen oxides content.

When a 30% H₂O₂ solution injected into the flame front, the NO concentration practically reaches the initial one, which obtained without the drip moisture injection (90-95 mg/m³). At the same time, the H₂O injection into the flame front leads to a decrease in NO concentration by 35–40%, up to 65 mg/m³. This can be explained by the fact that hydrogen peroxide forms the OH radical, which is actively involved in the hydrocarbons and CO oxidation, more intense than water. The spatial distribution of the flame generated radicals (OH and CH) (Mörtberg M. *et. al.*, 2006).

The OH radical forms hydroperoxide-radical HO₂ from H₂O₂, which dissociates with the superoxide ion O₂-formation and gives the H-radical from H₂O₂. The reaction rate is determined by the reaction:

$$W^1 = 4,4 \cdot 10^6 T^{1/5} \exp(3,1/(RT)) \quad (5)$$

It can be concluded that the most appropriate conditions for the NO in NO₂ oxidation is the hydrogen injecting with a hydrogen peroxide concentration in a solution 10.3% and the temperature range 800-1000 K. The flammable area, but not the flame front is the most suitable for these conditions. However, these assumptions need further study. Therefore when hydrogen is burned in the boiler furnace, the NO concentration increases accordingly, and the NO₂ share decrease accordingly.

Decreasing temperature with a change in the excess air coefficient leads to a decrease NO concentration in the flame front both without moisture addition and with it. H₂O drops addition reduces the maximum local temperature by at least 200 degrees for any excess air coefficient and the nitrogen monoxide content in this case is significantly decreases too. The combination helps in emission reductions, saving of fuel consumption and increased efficiency (Nugroho Agung Pambudia *et. al.*, 2017). Number of moisture that can be used in the combustion process chemically, obviously, must be different for different fuels and depend on the excess air coefficient. Any information about the experiments of other researchers with the moisture addition to the combustion process with a variable proportion of hydrogen is unknown to the authors.

Experiments showed that droplet moisture addition to the flame front provided a reduction in the maximum local temperature in the drop trace by 200 ± 10 degrees that led to decreasing the nitrogen monoxide content in the combustion products by 30-50%. After co-processing, the concentrations of NO_x, SO₂, CO, and all heavy metals in the flue gas emitted from the coal-fired drop-tube furnace did not change a lot. (Chen T. *et. al.*, 2020).

Thus the most effective reduction of NO formation due to the drip moisture addition into the flame front was achieved at lower α, or at higher flame temperatures. Local mean concentrations of O₂, CO, SO₂ and NO gas species and gas temperatures were determined in the region above the grate (Li Z. *et. al.*, 2009).

This effect can be used for reducing nitric oxide emissions by purifying the flue gases in the contact apparatus, for example the NO and NO₂ absorption by water. It is known that NO₂ is completely soluble in water, and the NO dissolution proportion does not exceed 7% and depends on the temperature and the catalysts presence. The hydrogen solutions injection into the combustion zone can be carried out by different ways and, in particular, in the liquid droplets form, as it is shown in this work.

Conclusions

1. The analysis of obtained results showed that with increasing hydrogen content in the solution the NO_x formation decreases, which can be explained by the oxygen consumption in fast reactions that require less energy. It is shown that with increasing hydrogen content in the solution, the NO₂ formation increases to maximum values at $\overline{H_2} = 10,32\%$, and then decreases again by 70% of the maximum concentration.
2. Research and analysis of the NO₂ concentration in the relevant investigated flame front areas with the hydrogen peroxide solution injection depends on: the H₂ concentration in the solution, maximum local temperature in the flame front, points of additional ingredients injection. The formation of up to 80% NO₂ in the composition of NO_x is observed near the anterior flame front border and in the first part of the flame front that is, NO is only 20% of the NO_x composition, if the hydrogen peroxide concentration is 15%.
3. The study of obtained results shown, the most appropriate conditions for the NO to NO₂ oxidation is the injection into the flame a solution, when the hydrogen content is 10.32% and the temperature range 800 – 1000 ° K. The flammable area rather than the flame front suitable for these conditions. Therefore, when burning hydrogen in the boiler furnace increase the NO concentration and the NO₂ share decrease.
4. It was established that with decreasing of the H₂ concentration in the hydrogen peroxide solution of (respectively, an increase hydrogen peroxide to 30% the concentration) H₂O ballast cooling effect in the solution decreases to zero by increasing the thermal CO oxidation effect by OH radicals, H₂O disintegration and increase the flame temperature by approximately 150°.

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