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## The use of thermochemical recuperation in an industrial plant

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### ABSTRACT

Thermochemical recuperation of exhaust gases heat can be a promising way to save energy when it comes to high-temperature plants heated by natural gas, for example glass melting furnaces. This technology can help to save up to 10-25% of energy without affecting other parameters of the process.

Schemes of glass melting furnaces with various options for thermochemical recuperation were simulated using Mathcad code and Aspen Plus code. It is shown that the dependence of specific primary energy consumption on the volume fraction of recirculated exhaust gases used as an oxidant for the reforming process is non-monotonic and has a minimum at the range of values 0.19–0.22 of mentioned volume fraction.

Determination of the oxidant flow coefficient for the reforming process for all considered options of thermochemical recuperation were suggested, as well as method of that calculation. In the industrial plant with reforming based on the recirculated exhaust gases transition to the optimal value of this coefficient (close to 1.00) is considered to be energy-saving, which reduces fuel consumption by 16% and reduces environmental pollution.

Use of economic evaluation of environmental pollution will allow to choose the scheme of thermochemical recuperation more efficiently.

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#### 1. Introduction

Industrial furnaces heated by natural gas are considered to be high-temperature plants (HTP). In general the exhaust gases of HTP are a mixture of combustion products (waste gases of combustion processes) and batch gases (waste gases of technological processes). It is well known that in HTP energy of hot exhaust gases can reach a considerable part of furnace input heat flows. For example, even today's most efficient regenerative or oxygen-fired glass melting furnaces generally show waste gas heat losses of about 25–30% of the total furnace energy input [1]. Utilizing a part of these heat losses can lead to a significant reducing of fuel consumption in HTP.

Several ways of exhaust gases heat recuperation in HTP considered nowadays are: fuel oxidant preheating, raw material preheating [2,3]. The hot exhaust gases can also be used for hydrocarbon fuel reforming. This method is often called as thermochemical recuperation (TCR). Technology of TCR reduces

hydrocarbon fuel consumption compared with conventional recuperation/regeneration when usually only combustion air is preheated and also significantly reduces air pollutant emissions [4].

TCR is based upon endothermic process of hydrocarbon fuel reforming. The primary fuel is recycled into the secondary fuel (syngas), which is used (partially or completely) in HTP.

Research is being conducted in the field of using of methanol steam reforming to increase energy efficiency of an internal combustion engine [5].

Positive impact of TCR on characteristics of steam-and-gas power plants has been thoroughly investigated [6]. Recuperation of waste heat in the methane reformer increases the exergy content of syngas, i.e. methane reformer operates like a "chemical exergy pump" [7].

Maruoka N. et al. [8] suggested using TCR-steam system for recuperation of various hot wastes generated by the steelmaking industry. This and other studies on the use TCR for utilization of the metallurgical slag thermal energy have been detailed in Ref. [9].

TCR technology may be effectively used for recovering exhaust gases heat of biomass combustor. In this way, biomass energy is converted into chemical energy contained in the produced syngas. It is shown that biomass can replace up to 5% of the energy in the





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natural gas feed [10].

Lab-scale test unit for research of TCR based on reforming natural gas using both steam and recirculated exhaust gases was constructed [4,11]. Experiments regarding TCR chemical efficiencies (increase in heating value not including increase in thermal energy) for varying reforming temperatures and exhaust gas/ natural gas ratios were done. At a reforming temperature of 620 °C and an exhaust gas/natural gas ratio of 2.5, the reformed fuel heating value is about 10% higher than that of natural gas [4].

A study of TCR-steam technology and construction of thermochemical recuperator for glass furnaces have been represented in Refs. [1,12,13]. The optimum process conditions for syngas production have been investigated. It was shown that complete conversion and degradation of the catalyst can be avoided for temperatures above 800 °C at a steam/natural gas ratio of about 3.0 [1]. Designed thermochemical recuperator refers to heat exchanger type with bayonet tubes.

Numerical study for the design of a steam reforming reactor with bayonet tubes has been conducted with respect to the various operating and design parameters [14].

Experimental investigation of exhaust gas thermochemical recuperation in tubular reformer has been performed [15]. The unit was designed for a maximum inlet exhaust gas temperature of 1040 °C. Reforming temperatures were as high as 760 °C. Exhaust gas recirculation was varied from 0 to 50%. At the highest temperature of 685 °C carbon monoxide and hydrogen were 10.5 and 25.0% of syngas mixture. A H<sub>2</sub> to CO ratio of approximately 2.5 was observed at all reforming temperatures of 520 °C to 685 °C. Syngas composition was essentially different from equilibrium one.

Using of regenerative exchanger for TCR on the base of natural gas conversion in combustion products was suggested [16] and implemented in industry. TCR study based on the conversion by recirculating exhaust gases has recently moved up to the level of industrial experiments at regenerative glass furnace with capacity of 50 tons/day [17–19].

Thermochemical recuperation of exhaust gases heat can be a promising way to save energy when it comes to high-temperature plants heated by natural gas. The exhaust gases heat, consumed by the reforming process, not only raises the temperature of the natural gas, but transforms into chemically bound energy of syngas. Due to this, thermochemical recuperation in industrial plants can provide additional fuel savings, as against traditional thermal recuperation, and reduce harmful effects on environment.

The TCR technology can help to save up to about 10-15% [2] or 20-25% [1] of energy in glass melting furnaces. Substitution of natural gas with syngas has no significant effect on batch melting kinetics and gives no significant changes in the flow patterns [1].

Most part of researches about TCR used the exhaust gases heat can be divided into two categories on the basis of the oxidant for reforming:

- 1) TCR based on reforming natural gas using steam [1,2,7,10–13,15];
- 2) TCR based on reforming natural gas by the recirculated exhaust gases. The components of exhaust gases are considered to be the oxidant for fuel reforming: carbon dioxide  $CO_2$  and water vapor  $H_2O$  [4,6,15–20].

Along with these trends comes another interesting tendency, when the oxidizing agent for the conversion is a mixture of recirculated exhaust gases and steam [4,16]. Syngas composition can be controlled by changing the ratio between the flows of steam and recirculated exhaust gases.

This article focuses at results of research on energy consumption in glass melting furnaces with three options of TCR using Mathcad code and Aspen Plus code. As an oxidizer are used steam, recirculated exhaust gases and their mixture. Analysis of research shown, that if recirculated exhaust gases are used as an oxidant for the fuel reforming, then dependence of the fuel consumption and the costs of primary energy on the volume fraction of recirculated exhaust gases used for the reforming is non-monotonic and has a minimum. In this paper propose determination of the oxidant flow coefficient for the reforming process for all considered options of TCR and a method of its calculation. In the industrial furnace with reforming, based on the recirculated exhaust gases, minimum of the specific energy consumption is observed at value of oxidant flow coefficient for the reforming process close to 1.00. Transition to volume fraction of recirculated exhaust gases, which corresponds to the optimum value of oxidant flow coefficient for the reforming process reduces fuel consumption by 16% and lowers environmental pollution. Proposed determination and method of calculation of the oxidant flow coefficient for the reforming process give more possible ways for energy saving in industrial furnaces.

# 2. Parametric analysis of industrial plants with thermochemical recuperation

In this paper the options of glass melting units with exhaust gas TCR in the recuperating reactor were researched. The schemes shown in Fig. 1 are different in oxidant composition for the reforming process: either water vapor (scheme I), or recirculated exhaust gases (scheme II), or a mixture of steam and recirculated exhaust gases (scheme III).

In scheme III the possibility to control the ratio of  $CO/H_2$  is provided in the composition of the syngas and the generation of the syngas in quantities exceeding the needs of the industrial furnace. In such a case, the excess syngas can be used in related technologies as energy source or raw material.

In the presented schemes the streams of exhaust gases (stream 5 and 6) of heating and technological processes in HTP are characterized by  $V_{eg}^2$ , m<sup>3</sup>/(m<sup>3</sup> of natural gas). Here and below, the volumes of natural gas and other gases are measured with conventional standard conditions 273.15 K and 101325 Pa.

In schemes II and III part of the exhaust gases (stream 13) is characterized by a volume fraction of recirculated exhaust gases –  $\varphi = V_{eg}^{reform}/V_{eg}^{\geq}$  which are used as an oxidant for the reforming process. This stream mixes with natural gas and gets into the reforming reactor. There  $V_{eg}^{reform}$  – specific consumption of exhaust gases which are used as an oxidant in the process of reforming, m<sup>3</sup>/ (m<sup>3</sup> of natural gas).

In scheme III the part of the exhaust gases (stream 13) in an amount of  $V_{e,g}^{reform}$  and water vapor (stream 11), characterized by a specific steam consumption  $V_{steam}$  (m<sup>3</sup> of steam/(m<sup>3</sup> of natural gas)), constitute the mixture that is the oxidant for the reforming process. Controlling the composition of the syngas occurs by changing  $\varphi$  and  $V_{steam}$ .

Simulation was performed using Mathcad code and Aspen Plus code for recuperative glass melting furnace with a capacity of 250 tons per day at an exhaust gas temperature of 1500 °C. Primary fuel – natural gas (% vol.): CH<sub>4</sub> = 89.7; C<sub>2</sub>H<sub>6</sub> = 5.2; C<sub>3</sub>H<sub>8</sub> = 1.7; C<sub>4</sub>H<sub>10</sub> = 0.5; C<sub>5</sub>H<sub>12</sub> = 0.1; CO<sub>2</sub> = 0.1; N<sub>2</sub> = 2.7. The oxidant flow coefficient in the combustion of the fuel is  $\alpha$  = 1.1. Specific consumption of raw material is 1.206 kg/(kg of molten glass). The fraction of cullet is 30% of the raw material. The humidity of the raw material is 4%. In this simulation was assumed the fuel is burned completely and the syngas is in the state of thermodynamic equilibrium.

When using only thermal recuperation (air preheating by the exhaust gases up to 800 °C), the specific fuel consumption is 0.189 m<sup>3</sup>/(kg of molten glass). In this case the specific primary

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