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Transition towards Carbon Footprint Reduction and Energy Intensification in Natural Gas Reforming

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Abstract

One of the possible options for carbon footprint reduction in ammonia production is energy intensification of the natural gas reformer furnace. This paper addresses how process modelling and simulation concepts can be combined with proven methods such as minimum energy consumption analysis and heat exchanger networks to identify and optimise the main bottlenecks in natural gas reformer furnaces. The main outcome is finding the best possible reconstruction options for natural gas savings and related CO_2 reduction. The applied method delivered an overall reduction of natural gas of 834 $m^3\,h^{-1}$ or 0.51 GJ with accompanying carbon footprint reduction of 0.026 t CO_2 *per* ton of NH_3 produced. A large part of EU's ammonia producers will be able to reach net zero emissions by 2050 by following the policy initiatives of the "Green Deal" thanks to this case study. In addition, it provides the basis for the further execution of other possible non-conventional retrofit measures.

Keywords

Ammonia production, carbon footprint reduction, design, natural gas reformer furnace

1 Introduction

According to the International Energy Association (IEA),[1] approximately 450 million tons of carbon-dioxide emissions and around 8.6 EJ of final energy consumption in 2020 were attributed to the ammonia production process. Of this, over 95 % of the energy came from fossil fuels. This accounts for about 20 % of the entire chemical industry's energy use, and around 35 % of the global CO_2 emissions. Ammonia production, as an intensive chemical process, accounts for around 2 % of final energy consumption and 1.3 % of energy sector emissions when compared to the energy industry as a whole. As stated in the IEA report,[1] if the ammonia industry were a nation, it would be the 16th largest emitter in the world, listed between Australia and South Africa.

Ammonia producers face the huge challenge of eliminating CO_2 emissions by 2050, turning near-zero CO_2 emission production technologies from a theoretical possibility to a practical reality. Different water electrolysis technologies, methane pyrolysis, and fossil-based routes with carbon capture storage and/or utilisation (CCS/CCSU) are some of the emerging technologies for producing hydrogen (as the raw material for ammonia production) with almost no CO_2 emissions. However, these technologies are, in most cases, 10–100 % more expensive *per* ton of ammonia produced in comparison with mature technologies, depending on hydrocarbon prices and other regional and policy factors. Near-zero-emission technologies are not yet available at commercial scale in the market because of constraints related mostly to the commercial and technical maturity. Although CO_2 sequestration is a necessary component of the ammonia industry today, it is obvious that permanent CO_2 storage is not yet commonly used due to the lack of facilities in the vicinity of most of the ammonia production sites in the European Union (EU). Proton exchange membrane (PEM) and alkaline water electrolysis-based ammonia production is already present at commercial scale using high load factor electricity, but challenges remain regarding the intermittent supply of renewable energy sources.[1]

The average net energy use for the 35 ammonia plants operated by 15 companies located in the EU was 34.8 GJ based on lower heating value (LHV) with an average generation of 1.9 t of CO_2 *per* ton of NH_3 produced.[2] On average, 1.34 t CO_2/ton NH_3 (69.5 %) was process-generated CO_2, and the remaining 0.59 t CO_2/ton NH_3 (30.5 %) was from fuel burning. The main part of the fuel burning is related to the largest piece of equipment in an ammonia production facility, namely the natural gas reformer furnace.

The natural gas reformer furnace, consisting of reformer tubes and several high intensity heat exchangers, is one the most energy intensive sections of an ammonia plant.[3] It is the greatest energy user, since essentially all of the hydrocarbon feed and fuel are consumed in the reforming section. Because of the combustion processes taking place in the natural reformer furnace, it is one of the main contributors defining plant energy efficiency and a chief source of CO_2 emissions. Possible limitations on energy efficiency can have their origin either in convection coils or in an air preheater.

To overcome this problem and to improve the overall natural gas reformer furnace efficiency by recovering excess stack waste heat, ammonia producers can also consider the replacement of old designed combustion air preheaters with a new improved design in order to reduce energy and minimise CO_2 emissions. This was done in Petrokemija Kutina ammonia plant in Croatia in combination with

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installing a new medium pressure (MP) steam heat exchange coil. This paper reflects the operational experience of these two heat exchangers commissioned in early 2021 to provide a transitive technical solution until additional near-zero technologies will be deployed.

The new air preheater is based on modern counter-flow heat-exchange technology rather than the traditional cross flow heat exchanger used in the old Kellogg, Braun and Root (KBR) plant. This kind of heat transfer results in a much narrower pinch point within a very small space. The performance of this new air preheater has been precisely monitored and the fuel saving with subsequent CO_2 reduction has been carefully measured.

2 Description and design of process modifications

2.1 Natural gas reformer furnace

The natural gas reformer is the largest energy consumer in the production of syngas, consuming more than 65 % of all energy supplied to the ammonia production process. Three aspects are important during operation of natural gas reformer: 1) providing enough heat for endothermic reaction between hydrocarbons and steam inside of the reformer tubes filled with the nickel based catalyst, 2) heat input by radiation to the external wall of the reformer tubes, and 3) heat transfer inside the convection section to the series of heat exchangers. The ultimate goal is to properly balance all of them with respect to the maximum hydrocarbon conversion while keeping the minimum possible steam-to-natural gas molar ratio with a reasonable reformer tube wall temperature, thus avoiding carbon deposition at the catalyst surface, and securing the ability to maximise heat recovery from the flue gas.[3] There are several standard arrangements of burners inside of reformer furnace, which can be classified into four main categories depending on their position: top and bottom fired, side and terraced wall fired.[3] In this case study, the top-fired reformer has the advantage of using high temperature flue gas for heating the reformer tubes. After transferring heat to the reformer tubes by radiation, the flue gas exits the radiation section at the bottom by passing through refractory lined tunnels at a temperature between 1,000 to 1,050 °C. Various process streams guided through convection section use this waste heat for further energy recovery. The major objective of the natural gas reformer furnace is to maximise decomposition of hydrocarbons with steam in the reformer tubes filled with nickel catalyst, and at the same time utilise maximum amount of waste energy from the flue gas. The heat remaining in the flue gas must be optimally used to preheat different process streams (mixture of natural gas and steam, boiler feed water (BFW), and air for secondary reformer), as well as to produce steam at various pressure levels before the flue gas exits into the atmosphere. In case of natural gas firing, a relatively clean source of energy that does not contain sulphur compounds, the flue gas outlet temperature can be reduced to approximately 120 °C without having to worry about sulphuric acid corrosion attack (sulphuric acid dew point in this case is approximately 93 °C). However, it can be a problem for oil-fired furnaces because of higher amount of sulphur compounds in oil-based fuels.[3]

Petrokemija operates a KBR ammonia plant in Kutina, Croatia. The ammonia plant was originally designed by KBR with a nameplate capacity of 1,360 t/day and was commissioned in 1983/1984. The natural gas reformer furnace in original design operates with 198 down-firing burners between 10 rows of 520 catalyst tubes, 11 tunnel burners for preheating the flue gas at the outlet of radiation section, and 21 superheater burners for preheating the high-pressure steam. In parallel with the natural gas reformer furnace, there is an auxiliary boiler fired with five burners to allow for steam production, which is necessary to keep the ammonia plant in self-sustaining mode of operation. The natural gas reformer furnace is designed to attain maximum thermal efficiency by utilising waste heat of the flue gases in the convection section. The waste heat is utilised to preheat the air supplied to the secondary reformer, preheat the natural gas feed and fuel, superheat steam, preheat and generate high pressure steam and, finally, to increase the temperature of the combustion air. The configuration of the natural gas reformer furnace with cold and hot process streams is shown in Fig. 1.

2.2 Analysis, synthesis, and process design

The specific motivation for the optimisation of natural gas reformer furnace was to improve utilisation of waste heat contained in the flue gases. Process analysis was performed, which indicated that significant amounts of the flue gas waste heat could be recovered to lower the flue gas exhaust temperature to the design value of 189 °C (or below).

A process model of the natural gas reformer furnace was created for performing simulation runs and calibrated with actual operating data. This model was the starting point for assessment of the existing equipment, especially the two parallel cast iron air preheaters (APH).

The distributed control system (DCS) data readings before the retrofit showed that flue gases at the stack of the natural gas reformer furnace still had a temperature of 223 °C that could be further reduced to maximise performance, improve energy efficiency, and reduce CO_2 emissions.

The process simulation included a minimum energy consumption analysis and a heat exchange network (HEN) design of the natural gas reformer furnace. Two main goals were in scope of simulation: 1) verification of the long-term energy conservation measures, 2) reduction of the natural gas, and thus the overall CO_2 footprint of the ammonia plant.[4–8]

The minimum energy consumption analysis and HEN synthesis was used to define the minimum energy requirement and maximum energy recovery (MER), respectively. It indicated that the hot flue gas streams with 605 °C (after cold superheater coil) and 405 °C (after the BFW coil) were ideal sources for energy utilisation. According to the results of the analysis and synthesis procedure, these

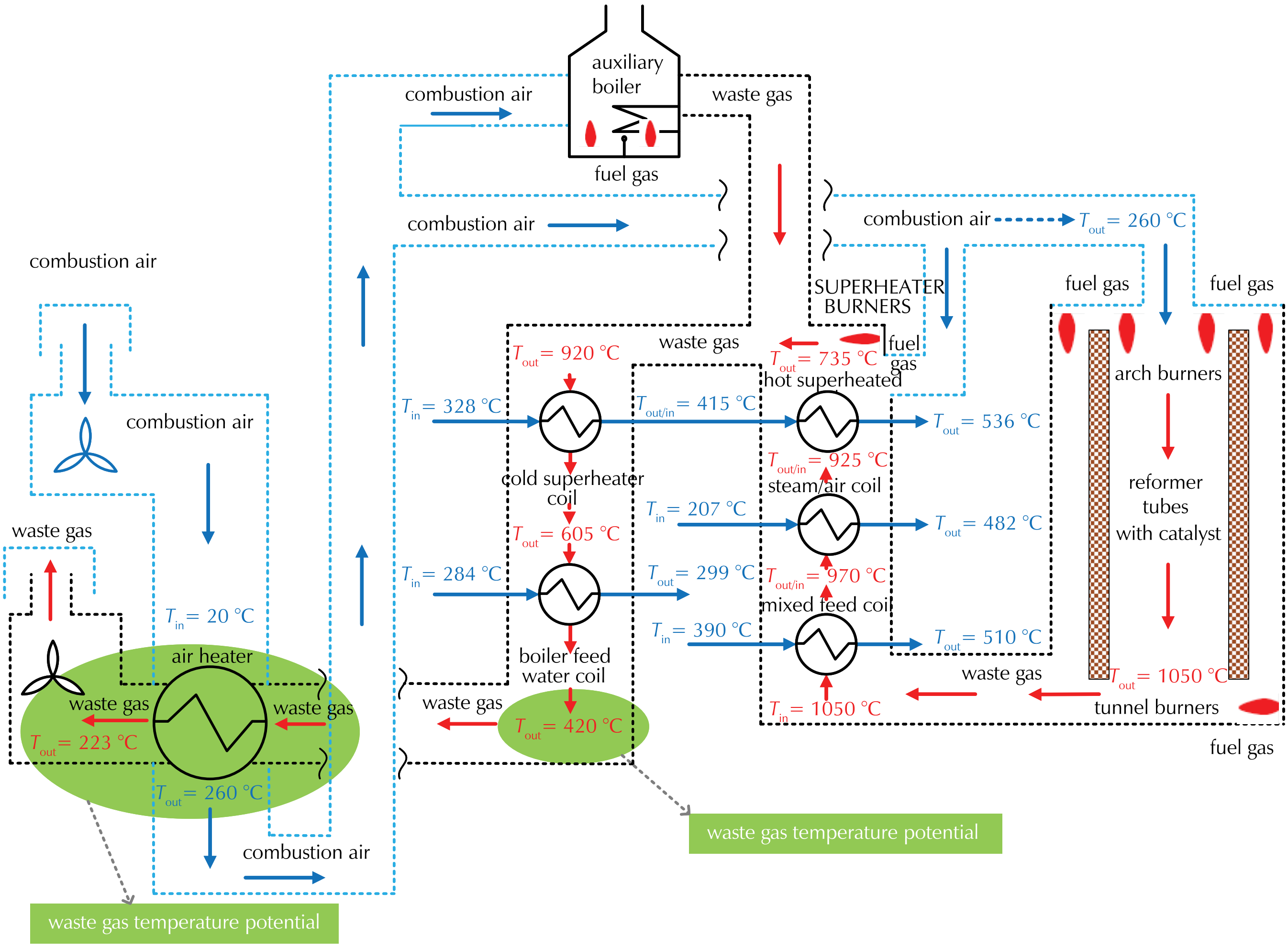

Fig. 1 – Configuration of top-fired natural gas reformer furnace with temperature profiles of cold and hot process streams (hourly average results obtained from DCS)

Slika 1 – Prikaz hladnih i toplih procesnih tokova u peći reformera prirodnog plina loženog svodnim plamenicima (prosječne satne vrijednosti temperatura očitanih sa sustava za upravljanje procesom)

two streams with indicated temperatures were identified as suitable heat sources for long-term energy conservation measures.

Some of the results have already been published;[9] according to the results, a detailed engineering design (DED) for a new preheating coil for medium pressure (MP) steam (40 bar) and air preheater has been developed. The results showed that it was feasible to achieve long-term measures in energy and CO_2 emission reduction with minimum capital expenditure. These results have been again validated through MER recovery and are presented in Fig. 2.[9] They show that the best locus for the new MP steam heating coil is above the existing BFW coil.[8,9]

Validated analysis showed that above the existing BFW coil, a new MP steam coil could appropriately utilise the remaining waste heat from the hot process stream with the starting temperature of 605 °C. This heat exchange coil preheats the MP steam from the main condensate header before mixing with most of the process steam from the main MP steam header. This new heat exchanger operates with 21.5 $t\,h^{-1}$ of MP steam (40 bar) entering at 249 °C. After preheating, the final MP steam temperature (before combining with the natural gas) of approximately 360 °C is reached.

The existing air preheater was able to deal with the waste gas from natural gas combustion, as well as with the waste gas from heavy fuel oil firing. Therefore, the last set of heat exchanger tubes was made of acid resistant glass. The future natural gas reformer will use natural gas only, which will improve the performance of the new combustion air preheater. DED verified that the new design of air preheater with high-temperature elements would be the most feasible technical solution for maximum energy recovery. The new air preheater was constructed to ensure combustion air temperature of 380 °C. In addition, the volume flow of air stays at the level to secure designed ammonia plant production capacity. Figs. 3 and 4 show the location of the two new heat exchangers in the retrofit design.[8,9]

After commissioning of the MP steam heat exchanger, the hot stream has a temperature of 420 °C, which perfectly

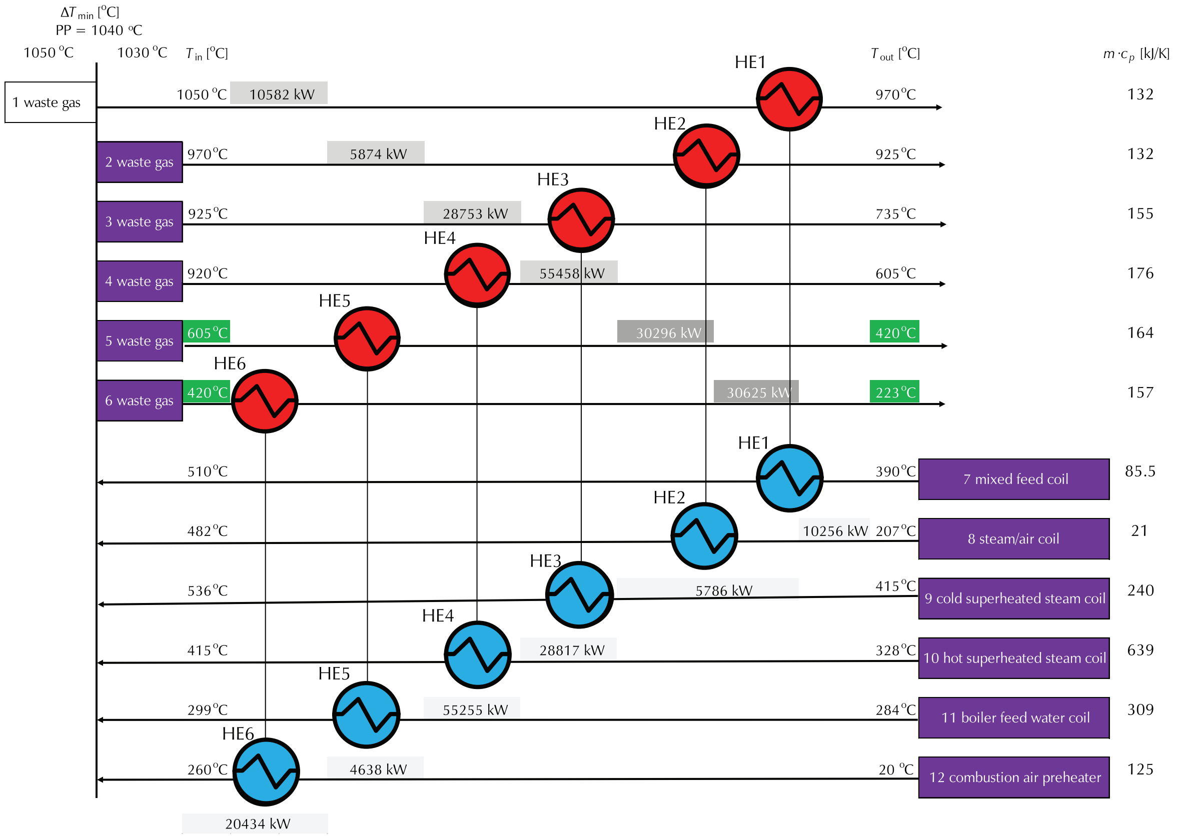

Fig. 2 – Validated MER heat exchange network design
Slika 2 – Validirani model mreže izmjenjivača topline

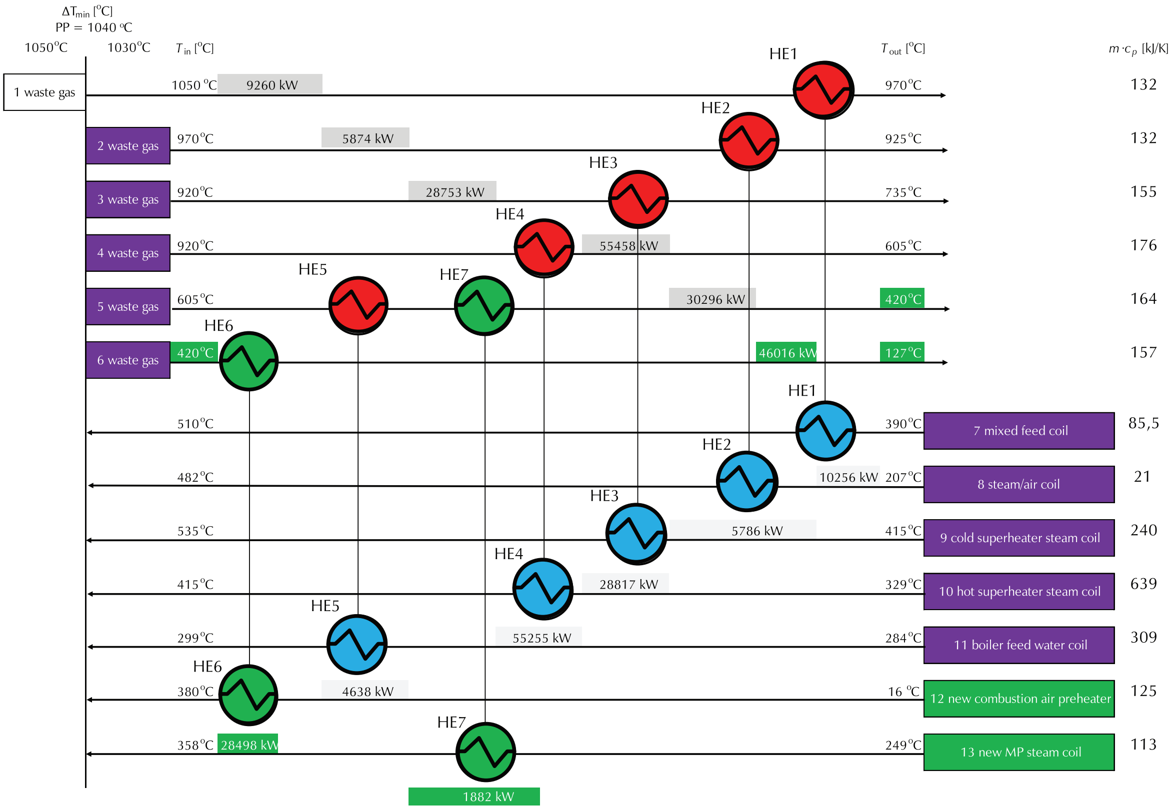

Fig. 3 – Retrofit MER heat exchanger network design
Slika 3 – Model mreže izmjenjivača topline nakon provedene rekonstrukcije

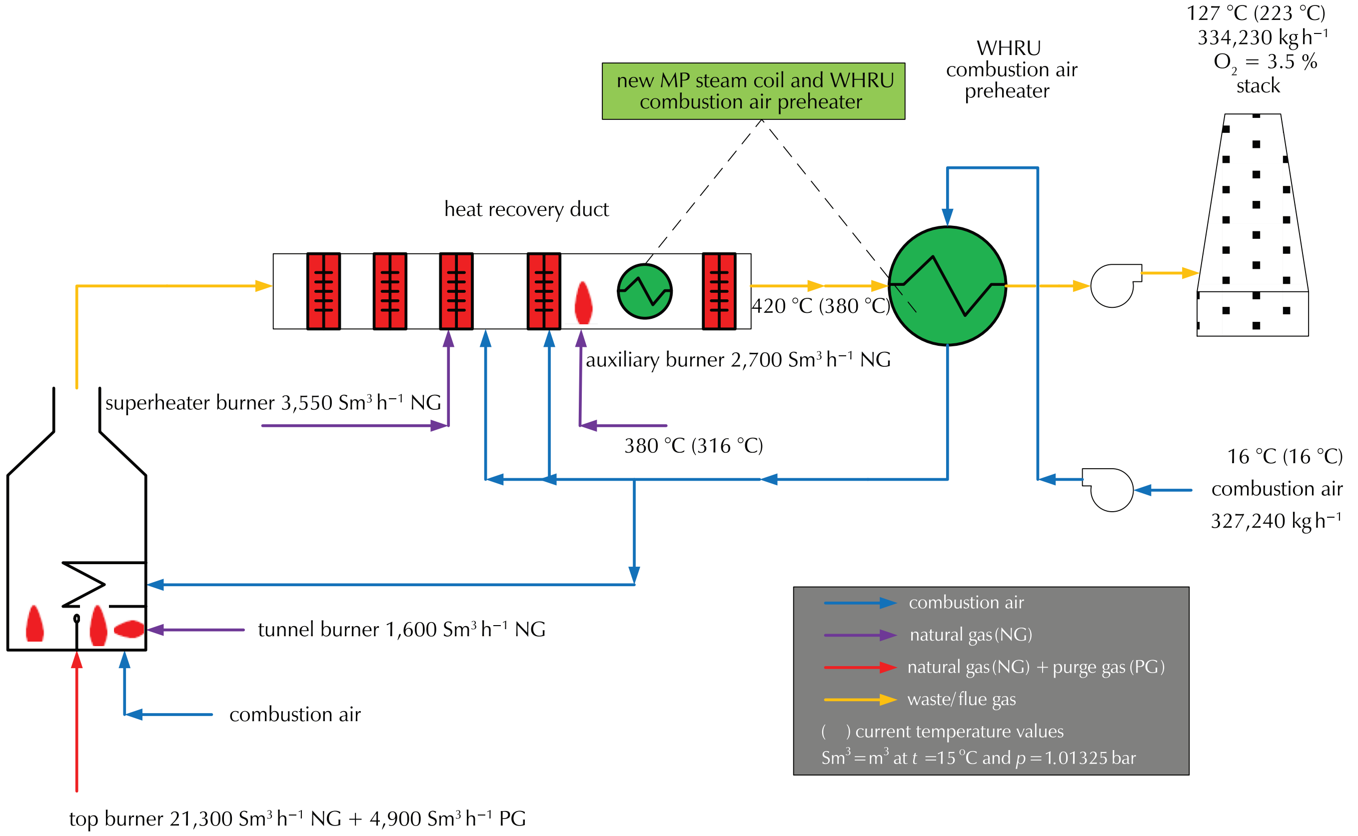

Fig. 4 – Simplified mass and process flow diagram of new heat exchangers within natural gas reformer furnace
Slika 4 – Lokacija novih izmjenjivača topline u reformeru prirodnog plina

fits the DED value of 420 °C. The hot stream, after the installation of the new air preheater, has a temperature of approximately 130 °C, which is an even better temperature in relation with the DED value of 120 °C, thus increasing safety margins related to acid corrosion attack. With the installation of the new heat exchanger, predicted reduction in natural gas is expected to be as high as 865 $m^3\,h^{-1}$ with directly related reductions of CO_2 emissions by 0.029 t CO_2 *per* ton of NH_3 produced. This accounts for approximately 3 % savings in comparison with the performance of the reference natural gas reformer furnace.

In order to minimise the amount of site work while still reaching the target performance, a new APH design was constructed based on a counterflow heat exchanger, a design where the flue gas and air streams are flowing vertically in opposite directions, as shown in Fig. 5.

Counterflow heat exchangers offer the most efficient way of exchanging heat between two gaseous media. The flue gas and air streams flow over the plates in opposite directions in a single stage, thus avoiding the need of a return duct and minimising the pressure drop. The heating surface consists of plates welded together and assembled into plate packs, which are then assembled into a casing to form heat exchanger modules (HEM).

The hot flue gas enters the unit *via* distribution hoods above the exchangers, and flows vertically downwards passing by the heating plates before being routed away from the unit *via* lower distribution hoods. The combustion air takes the opposite path, entering *via* the lower hoods, flowing vertically upwards, before being routed away *via* the top hoods above the unit. Similar to the old device, the new APH was built as two twin units, each of them consisting of two large heat exchanger modules.

Fig. 5 – Counterflow heat exchanger
Slika 5 – Protustrujni izmjenjivač topline

In order to fully use the available installation space, the width of the plates was also adapted to fit the support structure of the old APH. The material of construction for the heat exchanger plates was SS304 of 0.9 mm thickness.

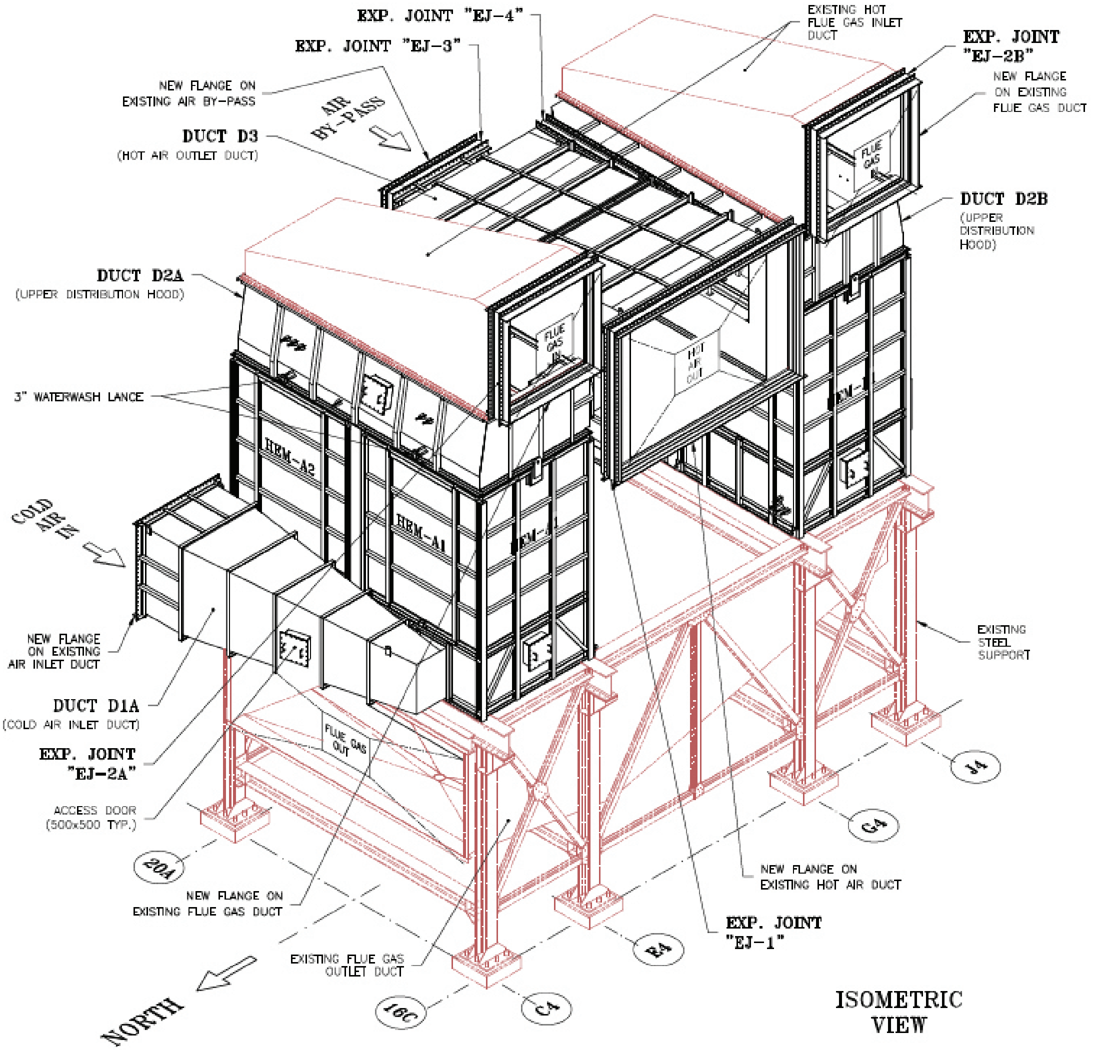

Fig. 6 – New combustion air preheater arrangement
Slika 6 – 3D prikaz novog izmjenjivača topline za pregrijavanje zraka za gorenje

The full system was designed to re-use as much as possible any existing equipment such as flue gas inlet ducts or air inlet ducts. In addition, the size of the delivered equipment was maximised to reduce the amount of site work and handling. Each delivered heat exchanger module had a weight of 43 t. Fig. 6 shows the arrangement of the new combustion air preheater which comprises four heat exchange modules, retains the same interfaces, maximises the use of existing duct, and keeps the same steel support structure.

Fig. 7 shows the 3D position of the new MP steam coil within the natural gas reformer furnace convection section arrangement.

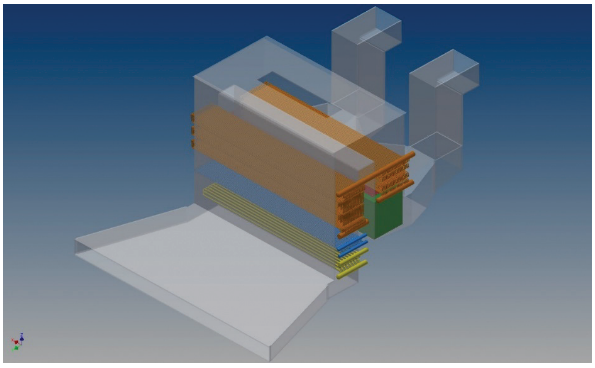

Fig. 7 – Location of the new MP steam coil in the convection section of the reformer furnace – blue coil
Slika 7 – 3D prikaz novog izmjenjivača topline za pregrijavanje pare tlaka 40 bar u konvekcijskoj zoni peći primarnog reformera – plavi izmjenjivač topline

3 Results of retrofit measures

3.1 Project execution

After analysis, synthesis, and design activities, Petrokemija and BD Heat Recovery agreed on contractual obligations in July 2020 regarding the delivery of engineering, procurement, fabrication, and installation of the new APH. Headquartered in Seminole, FL, BD Heat Recovery Division, Inc., is a member of BD Group Industries, LLC, and has the flexibility and ingenuity to handle the most challenging demand for heat recovery and NO_x reduction applications.

In parallel, Petrokemija initiated, independently of BD Heat Recovery, a procedure for fabrication, delivery, and installation of the new MP steam coil according to Petrokemija's "know-how" engineering services, which was based on the KBR basic engineering design (BED).[10,11]

During the engineering phase of the project, special precaution has been taken to select suitable construction materials that would maximise the anticipated lifespan of the equipment. Based on the operating experience from both contractual partners, it was estimated that the equipment would maintain adequate performance for a period of 15 to 20 years.

Both heat exchangers were installed during the planned turnaround in January 2021. Including all necessary pre-assembly activities, the installation took 40 calendar days. The delivery schedule of the APH heat exchanger was less than six months from signing the letter of intent (LOI) to the final delivery on site.

3.2 Start-up procedure and analysis of process performance

The start-up procedure of the ammonia plant commenced on January 30, 2021. Steady-state production was achieved on February 6, 2021.

After stabilisation of the entire ammonia plant production process, a preliminary analysis of the process parameters related to the installation and performance of the new APH and MP steam coil was conducted. The comparison was done with respect to the old APH design and without an additional MP steam coil. All relevant process parameters were compared, allowing for a precise evaluation of the achieved benefits.

With respect to estimation of the natural gas savings and subsequently reduction of CO_2 emissions, 1 h averages for five consecutive days were used for comparison of the fuel consumption between an old and the new APH. The 5 consecutive days were taken in the operating period of the new APH from 2 March to 6 April 2021. The operating period with the old APH is from 25 to 30 November 2020. This was the most recent period with similar outside atmospheric temperatures and production rate. All fuel flow rates were then converted into the energy rates based on LHV values calculated by the HRN EN ISO 6976:2016 standard.[12] Fuel gas equivalents are calculated with a mean LHV for both observed periods (34.975 GJ m^{-3}). Besides that, the performance of the new MP steam coil was also evaluated in the same period. The results of achieved energy savings and CO_2 emission reductions are presented in Table 1.

Table 1 – Comparison of fuel gas flow rate and energy rates based on natural gas LHV of 34.975 GJ m^{-3} before and after retrofit

Tablica 1 – Usporedba volumnog protoka prirodnog plina (donja kalorička vrijednost = 34,975 GJ m^{-3}) i potrošnje energije prije i nakon rekonstrukcije

Fuel gas and energy flow rates	Period		Difference
	November 25–30, 2020	March 2 – April 6, 2021	
$m^3\ h^{-1}$	24,192.99	23,359.26	833.73
$GJ\ h^{-1}$	846.15	816.99	29.16
$GJ\ t_{NH3}{}^{-1}$	14.93	14.42	0.51
t_{CO2} / t_{NH3}	0.826	0.800	0.026

The results showed that the installation of the new APH unit and additional MP steam coil reduced the fuel consumption by 834 $m^3\ h^{-1}$, representing energy savings of 0.51 GJ and CO_2 emission reductions of 0.026 t CO_2 *per* ton of NH_3 produced.

Table 2 shows the difference between model and DED predicted savings and the operation based savings data. The total natural gas savings are around 3.6 %. From this data, it is obvious that design procedure based on performed heat analysis and HEN synthesis was in excellent alignment with practically obtained results.

Table 2 – Comparison between model and process values

Tablica 2 – Usporedba između modela i procesnih vrijednosti

Fuel gas and energy flow rates	Model/DED	Process	Difference
$m^3\ h^{-1}$	865.0	833.73	31.27
$GJ\ h^{-1}$	30.25	29.16	1.09
$GJ\ t_{NH3}{}^{-1}$	0.53	0.51	0.02
t_{CO2} / t_{NH3}	0.029	0.026	0.003

To additionally verify the performance of the APH and MP steam heat exchangers, Eq. (1) was used for the calculation of heat exchanger efficiency, and the results for both units are presented in Table 3.

$$\text{efficiency} = \frac{t_{\text{cold stream}}(\text{outlet}) - t_{\text{cold stream}}(\text{inlet})}{t_{\text{hot stream}}(\text{inlet}) - t_{\text{cold stream}}(\text{inlet})} \cdot 100 \qquad (1)$$

Table 3 – Heat exchanger efficiencies
Tablica 3 – Energetska učinkovitost izmjenjivača topline

Heat exchanger	$T_{\text{cold stream}}$(outlet) / °C	$T_{\text{cold stream}}$(inlet) / °C	$T_{\text{hot stream}}$(inlet) / °C	Efficiency / %
APH	370.96	22.92	397.43	92.93
MP steam coil	358.72	249.64	366.30	93.50

The heat exchanger efficiency achieved was about 93 %. The efficiencies for both heat exchangers were also consistent with the designed values.

Flue gas from the natural gas reformer is a major source of continuous atmospheric emissions from ammonia plants. The quantity of flue gas depends on the design and energy efficiency of the process; the main constituents are nitrogen, oxygen, carbon dioxide, water, carbon monoxide, oxides of nitrogen (NO_x), oxides of sulphur (SO_x), unburned hydrocarbons and particulates.[3] NO_x, are of greatest relevance to ammonia plants and they may be formed by the combustion of materials containing nitrogen or by reaction of atmospheric nitrogen and oxygen at high temperatures. In addition, due to higher temperature of the combustion air, the emission of NO_x will increase in these favourable process conditions. This observation was made after the installation of the new APH heat exchanger. According to predictions from the model, one drawback of the project was the resulting higher emissions of NO_x due to the higher temperature of combustion air.

After installation of the new APH heat exchanger, the NO_x emission increased by approximately 100 mg Nm^{-3}. Taking into account that even before installation of the new APH heat exchanger, the NO_x emissions were above the allowed limit of 230 mg Nm^{-3}, the next step will be the replacement of the originally installed arch burners by low-NO_x burners.

4 Conclusion

The paper describes an example of minimum energy consumption analysis and HEN integration as the mature techniques in designing retrofit options of a natural gas reformer furnace as one of the major contributors of energy consumption and CO_2 emission in ammonia plant.

The project for utilisation of the waste heat from the flue gases of the natural gas reformer furnace was based on simulation data. The simulation was successfully used to design transitional technology on the route to the implementation of near-zero CO_2 emissions from the ammonia plant.

The transition included the replacement of the existing APH heat exchanger with the new air preheater with enhanced design, and the construction of the additional MP steam coil. The total achieved savings were: 834 m^3 h^{-1} or 0.51 GJ *per* ton of NH_3 produced with accompanying carbon footprint reduction of 0.026 t CO_2 *per* ton of NH_3 produced. The total achieved overall efficiency of the heat exchanger network was approximately 93 %.

This case-study demonstrated that, before implementing of emerging near-zero technologies, the ammonia producers should consider the implementation of proven technologies such as pinch analysis and HEN integration for designing retrofit options to conserve energy and reduce CO_2 emissions.

List of symbols and abbreviations
Popis kratica i simbola

APH	– air preheater – pregrijač zraka
BED	– basic engineering design – glavni projekt
BFW	– boiler feed water – napojna kotlovska voda
CCS	– carbon capture and storage – hvatanje i skladištenje ugljika
DED	– detail engineering design – izvedbeni projekt
DSC	– distributed control system – sustav za upravljanje procesom
EJ	– exajoule (10^{18} Joule) – eksa džul (10^{18} džul)
EU	– European Union – Europska unija
GJ	– gigajoule (10^9 Joule) – giga džul (10^9 džul)
IEA	– International Energy Association – Međunarodno energetsko udruženje
HEM	– heat exchanger module – modul izmjenjivača topline
HEN	– heat exchanger network – mreža izmjenjivača topline
KBR	– Kellogg, Braun and Root – Kellog, Braun i Root (ime tvrtke)
LHV	– low heating value – donja kalorična vrijednost
LOI	– letter of intent – pismo namjere
MER	– minimum energy requirement or maximum energy recovery – minimalni utrošak energije/maksimalna oporaba energije

MP	– medium pressure – srednji tlak
N	– normal gas conditions (101,325 Pa and 0 °C) – normalni uvjeti stanja plina (101,325 Pa i 0 °C)
PEM	– proton exchange membrane – membrana za izmjenu protona
S	– standard gas conditions (101,325 Pa and 15 °C) – standardni uvjeti stanja plina (101,325 Pa i 15 °C)
SS	– stainless steel – nekorodirajući čelik

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SAŽETAK

Smanjenje utroška energije i emisije stakleničkih plinova u peći reformera prirodnog plina

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Jedna od mogućih opcija za smanjenje ugljičnog otiska u proizvodnji amonijaka je smanjenje utroška energije u peći reformera prirodnog plina. U radu je prikazana kombinacija suvremene tehnologije temeljene na konceptu procesnog modeliranja i simulacije s dokazanim metodama kao što su analiza minimalnog utroška energije i sinteza mreže izmjenjivača topline. Glavni cilj bio je prepoznavanje tzv. "uskih grla" u radu peći reformera prirodnog plina. Na toj osnovi definirane su i provedene mjere, što je rezultiralo uštedom energije i smanjenjem emisije CO_2. Nakon projekta ostvarena je ukupna ušteda u potrošnji prirodnog plina od 834 $m^3\,h^{-1}$ ili 0,51 GJ uz smanjenje ugljičnog otiska od 0,026 t CO_2 po toni proizvedenog NH_3. Provedeni projekt sukladan je cilju postizanja nultih emisija CO_2 do 2050., prema smjernicama Europskog zelenog plana ("Green Deal") za većinu proizvođača amonijaka u EU-u.

Ključne riječi
Proizvodnja amonijaka, smanjenje ugljičnog otiska, model, peć reformera prirodnog plina

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