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**IMPROOF: INTEGRATED MODEL GUIDED PROCESS OPTIMIZATION OF  
STEAM CRACKING FURNACES**

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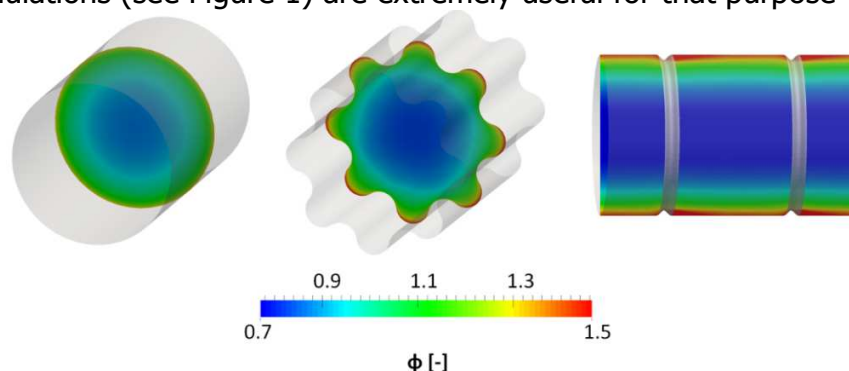
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Abstract: In IMPROOF the steam cracking furnace of the 21st century will be developed and demonstrated. The ambition is to drastically improve the energy efficiency compared with the current state-of-the-art, and this in a cost effective way. Simultaneously the greenhouse gases and NOX emissions per ton ethylene produced will be reduced by 25%. This project will implement and combine several of the latest technological innovations in the field of fouling minimization and energy efficiency at pilot and industrial scale. These include the use of renewable fuels, oxy-fuel combustion, and high emissivity coatings which emit in the non-absorbent flue gas spectrum. Also, new advanced high temperature alloys that lower the coking rate will be implemented in combination with novel 3D reactor technologies leading to reduced coking and enhanced heat transfer between flue gas and the process. In 2019, the furnace will be deployed at the demonstrator at commercial scale using propane as feedstock based on the experimental and modeling data provided by the industrial partners, knowledge institutions and research organizations.

## **Introduction**

Although steam cracking is considered a mature technology for olefin production, the complexity of the process and the harsh operating conditions allow the implementation of technology developments towards substantial heat transfer enhancement. One of the most important ways to reduce the energy input in steam cracking furnaces per ton ethylene produced is to reduce coke formation on the reactor wall of the long tubular reactors that are mounted in the furnaces<sup>1</sup>. Steam cracking is the most energy-consuming process in the chemical industry and globally uses approximately 8% of the sector's total primary energy<sup>2</sup>. Additionally, it is responsible for massive amounts of CO<sub>2</sub> emissions<sup>3</sup>. Improving the energy efficiency has an immediate payout because energy cost counts for a substantial part of the production costs in typical ethane or naphtha based olefin plants<sup>3</sup>. Typically, ethylene furnaces have to be decoked after 30-60 days to remove the coke that builds up in the coil. When the furnaces are decoked, production of the desired products is stopped for approximately 48 hours<sup>4</sup>. During the course of one run, deposited coke can reduce the heat-transfer efficiency of the firebox by 1-2 %, resulting in a 5 % increase in fuel consumption<sup>5</sup>. The use of advanced coil materials, combined with 3D reactor designs, improved process control, and more uniform heat transfer could

increase run lengths <sup>6</sup>, reducing simultaneously CO<sub>2</sub> emissions and increasing the lifetime of the furnaces. It has been proven that improved metallurgy of radiant coils can reduce catalytic coking <sup>1</sup>, and that advanced 3D coil design such as the swirl flow reactor design <sup>7</sup> and the SCOPE design (by Schmidt + Clemens GMBH +CO. KG) can mitigate coke deposition leading to heat transfer improvement. Improved geometries can result in smaller radial temperature gradients inside the coil, and thus lower wall temperatures where the coke formation occurs. Advanced 3D CFD reactor simulations (see Figure 1) are extremely useful for that purpose <sup>8</sup>.



**Figure 1:** Three-dimensional representation of the studied reactor designs with contour plots of the corresponding local fluid age correction factors, from left to right: a bare tubular reactor, a longitudinally finned reactor and a transversally ribbed reactor <sup>8</sup>.

An important challenge for the petrochemical industry is the upcoming stronger environmental regulations, in particular related to NO<sub>x</sub> and CO<sub>2</sub>. The total NO<sub>x</sub> emissions of all the furnaces currently operating in the EU are approximately  $16.3 \times 10^3$  tpa. To reduce NO<sub>x</sub> emissions the use of advanced oxy-fuel combustion is demonstrated on pilot plant scale. The advantage is clear: because no nitrogen is added (apart from leakages), almost no NO<sub>x</sub> is produced <sup>9</sup>. An additional advantage is that the produced flue gas is a concentrated CO<sub>2</sub> stream, that can be more easily captured, stored or used, for example, in chemical looping <sup>10</sup>. Bio-gas and bio-oil are renewable fuels, and hence, decrease net CO<sub>2</sub> production <sup>11</sup>. It is expected that these fuels will become available in substantial amounts in the near future in Europe and therefore can be used as fuels for steam cracking furnaces.

A final point for improvement is related to the radiant section of a steam cracking furnace, where the major part of heat transfer occurs by radiation. The radiation is emitted by the refractory walls towards the process radiant coils. Application of high emissivity coatings on the external surface of the radiant coils could improve the energy consumption <sup>12</sup>. Applying the improved emissivity coatings on the furnace walls <sup>13</sup> will decrease the required firing to reach the same process temperatures in the radiant coils. This will reduce fuel gas consumption and CO<sub>2</sub> emissions by an anticipated 10 to 15%. In addition to higher heat absorption, coating the external surface of radiant coils can improve the surface homogeneity and eliminate hot spots on the tube walls. An additional benefit will be extended run lengths resulting in less energy spent annually for decoking the radiant coils.

The IMPROOF project will demonstrate the advantage of combining all these technological innovations, with an anticipated reduction of emissions, and increase of the time on stream and energy efficiency.

## Objectives

To answer the market need and implement novel technology for olefin production the project has set the following technical objectives:

1. Demonstrate the individual impact of novel emissive, reactor and refractory materials on pilot scale (TRL5),
2. Demonstrate the power of advanced process simulation (high performance computing and CFD) for furnace design and optimization,
3. Demonstrate the technical economic and environmental sustainability of the IMPROOF furnace at TRL6,
4. Novel combustion technology using alternative fuels and oxy-fuel combustion, and
5. Coke formation reduction and real time optimization.

In the following section the first results obtained on the first two objectives will be presented and discussed.

## Results

### *High emissivity coatings*

Emissivity is a radiometric relative property of a material that quantifies the ability of the real surface to emit radiation. It is defined as the ratio of radiances emitted by the real surface and an ideal black body at the same temperature, spectral and geometrical conditions. In the case of coatings intended for high-temperature applications in furnaces, their emissivity is among the most crucial characteristics that considerably influence thermal efficiency of the whole furnace <sup>14</sup>.

Two novel high-emissivity coatings designed for radiant walls/floor of furnace and process coils/tubes were investigated in the experimental setup for the normal spectral emissivity at the New Technologies Research Centre within University of West Bohemia. Next to coatings provided by Emisshield company, also reference materials, namely regular stainless steel 304 and Alamo<sup>®</sup> refractory bricks (produced by HarbinsonWalker International) were inspected.

The measuring apparatus of the normal spectral emissivity measurement method has been recently described <sup>14,15</sup>. The experimental setup (see Figure 2) consists of a laboratory blackbody as a reference source of radiation and a FTIR spectrometer that detects emitted radiation both by the blackbody and real materials. An infrared camera is used to measure the surface temperature. The blackbody and the sample are positioned against each other. A rotary parabolic mirror is positioned in the middle of the blackbody and the sample, and is used to switch the optical paths. The radiation collected by the mirror enters the spectrometer through an entrance port. Optical and optomechanical components (translation stages, mirrors, aperture,

shutter, alignment laser) define the analyzed area size and accomplish equal optical paths from both radiation sources. The optical path is enclosed inside a box; however, no evacuating or purging is used <sup>14,15</sup>.

Steel, ceramic and coated disc substrates of 25 mm in diameter and 5 mm thickness are used (see Figure 3). The measured sample is clamped to a ceramic fiber insulation case and it is attached to the optical bench by translation stages <sup>14</sup>.

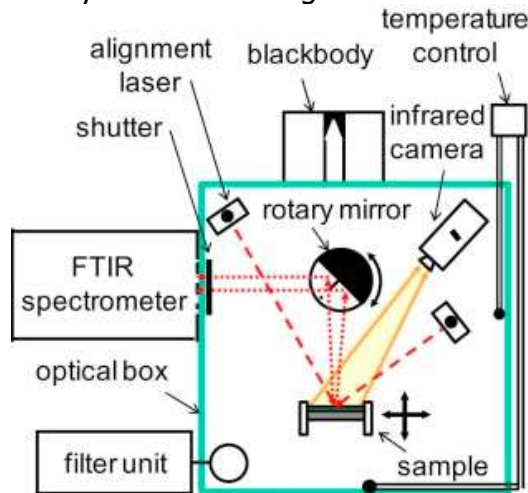


Figure 2: Schematic view of the emissivity measuring apparatus with the optical path for the radiation coming from the sample to the detector <sup>14,15</sup>.



Figure 3: (Un)coated 304 stainless steel and (un)coated Alamo® refractory samples prepared for normal spectral emissivity measurements.

Normal spectral emissivity measurements of a coated metal sample were performed at different temperatures in order to check the temperature dependency. As can be seen from the left side of Figure 4, all spectral emissivity experiments appear to be temperature independent in the observed temperature range (700 – 850 °C). The right side of Figure 4 gives a comparison of normal spectral emissivities of coated and uncoated ceramic material obtained at 800 °C. As

can be seen from the right side of Figure 4, a big difference exists in the sub 5  $\mu\text{m}$  region, which is the most important for radiation in steam cracking furnaces. Thus, coating the refractory wall can significantly improve the thermal efficiency of the cracking furnaces. Nevertheless, a decrease of spectral emissivity is observed for wavelengths shorter than 5  $\mu\text{m}$ . As this is not aligned with current literature data <sup>16</sup>, the spectral emissivity measurement method will be further improved in order to evaluate this in more detail.

Figure 5 shows the comparison of normal spectral emissivity of uncoated and coated 304 stainless steel. As can be seen from Figure 5, a clear difference exists between the samples throughout the entire range of wavelengths. Emissivity of 304 stainless steel appears to be slightly higher than data found in literature <sup>17,18</sup>. However, the surface is rather rough compared to the smooth surfaces typically used in emissivity experiments, which as a consequence has a positive effect on emissivity values <sup>19</sup>. Important to note is that once the 304 stainless steel material is oxidized, spectral emissivity will increase <sup>19</sup> and will result in a smaller gap with coated 304 stainless steel. Figure 6 gives a comparison of the two different coatings designed for ceramic and metallic substrates. It is clear that although the composition of the two materials is different, the emissivity behavior is very similar.

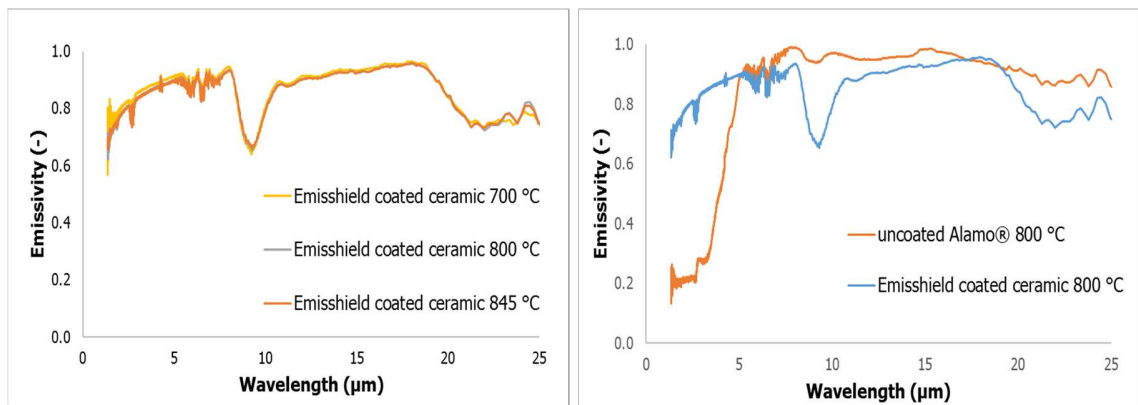


Figure 4: Left: Temperature dependency of normal spectral emissivities of coated ceramic samples and Right: Comparison of normal spectral emissivities of coated and uncoated ceramic materials at 800 °C.

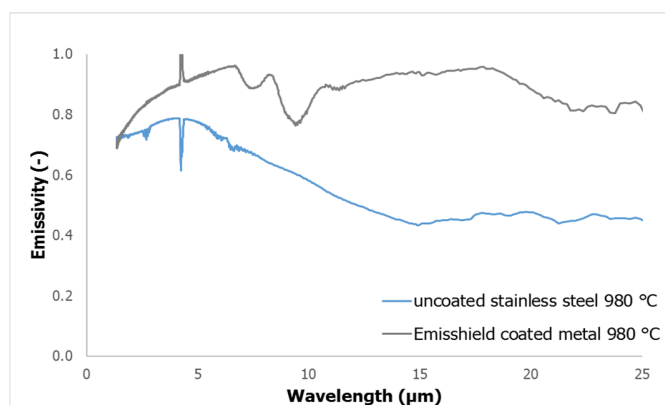


Figure 5: Comparison of normal spectral emissivity of uncoated and coated 304 stainless steel.



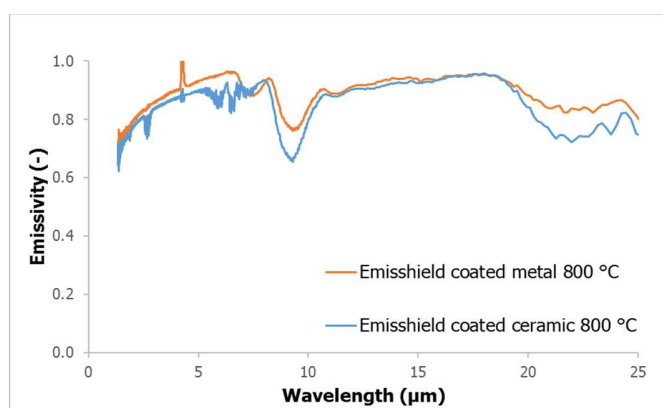


Figure 6: Normal spectral emissivities of coated metal and coated ceramic at 800 °C.

### ***3D Reactor Technology***

A lot of research focusses on the enhancement of heat transfer between the reactor coil and the process gas, as this positively affects the process in multiple ways <sup>20</sup>. Firstly, heat transfer enhancement at this point implies a lower temperature at the metal surface, which results in a decreased rate of coke formation and, consequently, longer run lengths. Next to this, an enhanced heat transfer allows to decrease the residence time of the process gas, resulting in a more favorable product distribution, i.e. a higher selectivity towards light olefins.

To achieve this heat transfer enhancement, different reactor designs are proposed that either increase the reactor surface, e.g. longitudinally finned reactors <sup>21</sup>, or promote turbulence, and radial mixing, e.g. MERT <sup>22</sup> or SFT <sup>7</sup>. The increased radial mixing implies a more uniform temperature distribution of the process gas, which has additional beneficial effects on the product distribution.

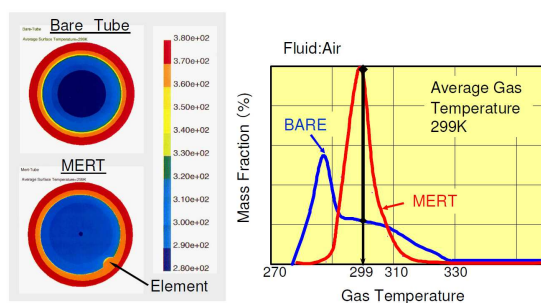


Figure 7: Gas temperature distribution in bare tube and MERT tube <sup>22</sup>.

Next to the beneficial increase in heat transfer, the strong turbulence induced by 3D elements increases the shear stresses in the flow, resulting in an increase in pressure drop. As this is detrimental to the product selectivity, the design of 3D elements can be viewed as balancing these two effects against each other. A detailed study of the flow behavior and the overall performance of these 3D reactor designs is thus necessary.

It should be noted that small modifications to the reactor design can introduce secondary radial or swirling flow patterns, thereby completely altering the reactor performance both in terms of heat transfer and pressure drop. As these additional flow patterns are highly three dimensional in nature, simple one-dimensional simulations, which are today's industrial standard, are unable to accurately capture all flow phenomena and consequently cannot assess the impact on heat transfer enhancement. Consequently, it is necessary to assess the performance of 3D reactor technologies by means of three-dimensional CFD <sup>23</sup>.

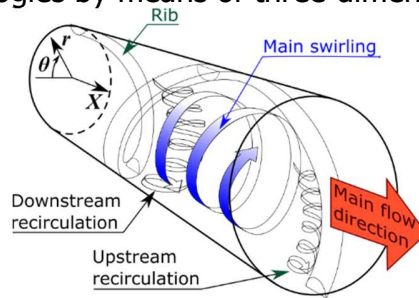


Figure 8: Three dimensional secondary swirling flow patterns and smaller recirculation zones in ribbed geometry <sup>24</sup>.

For this performance assessment, validation of the applied CFD methodology is crucial, but only few adequately validated cases are available in the open literature. In order to have confidence in the simulation results, a cooperation with the Von Karman Institute (VKI) has been established. Their expertise in flow field measurement allows the extraction of highly detailed information of both the velocity field, by means of Stereo-PIV, as well as of the heat transfer at the wall, by means of liquid crystal thermography (LCT) for swirling flows. This experimental campaign showed that for this type of reactor design RANS (Reynolds Averaged Navier-Stokes) simulations, in which all scales of turbulence are modelled, are incapable of capturing important flow phenomena like recirculation zone size and reattachment lengths. Fully resolving all turbulence, even to the smallest scales, with Direct Numerical Simulation (DNS) is currently still impossible at industrially applied flow regimes and therefore swirling flows in 3D reactors should typically be resolved by means of Large Eddy Simulation (LES) <sup>25</sup>. This technique resolves the largest eddies, which contain the most turbulent kinetic energy, and models the smaller scales, thereby decreasing the computational demand.

The application of stream wise periodic boundary conditions (SPBC) <sup>8</sup> in the open source CFD package OpenFOAM<sup>®</sup> allows to significantly decrease the computational domain, thereby further reducing the computational demand up to a level that allows a highly accurate and detailed study of the influence of geometric parameters of the 3D design.

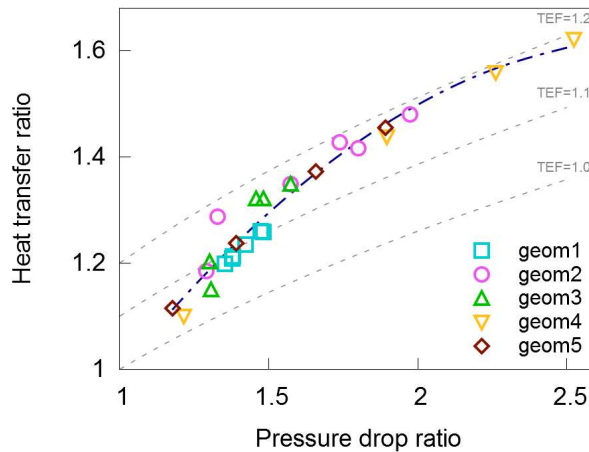


Figure 9: Parametric study of 3D reactor design showing the trade-off between heat transfer and pressure drop.

Based on this information, the performance of different designs can be compared against each other allowing to further optimize these designs to improve heat transfer with little additional pressure drop. Next to this, information from this kind of simulations can also be used to redesign the reactor in a more fundamental way, e.g. decreasing recirculation zones or enhancing mixing effects, thereby leading to novel steam cracking coil designs.

As metal temperature is less uniform due to the application of 3D elements, the coke layer will generally grow non-uniformly. This non-uniform growth will influence the reactor geometry over time which will in turn influence the flow pattern and consecutive coking behavior. In order to assess the long term performance of these 3D coils, one should not only evaluate the performance at start-of-run conditions but monitor the coke formation and overall performance throughout the run. It has been shown that there is a pressure drop penalty for the enhanced heat transfer. However, this effect is leveled out throughout the run due to the decreased rate of coke formation (see Figure 10).

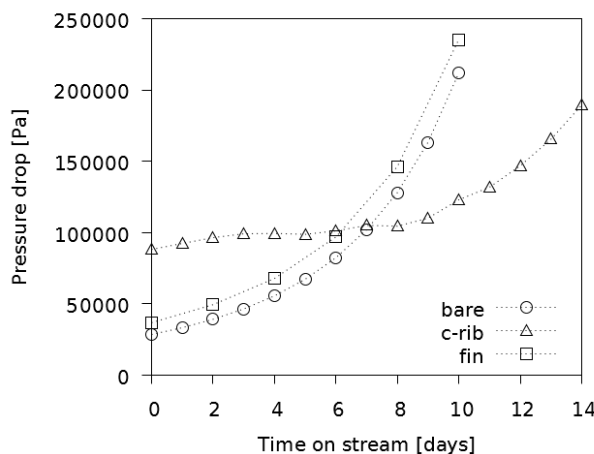


Figure 10: Pressure vs. time on stream for bare, c-rib and finned tubes for propane cracking.

Furthermore, run length simulations have shown that the on-stream time can be drastically improved by the implementation of these 3D designs.

## **Conclusions**

The first results of the IMPROOF project show that applying high emissivity coatings on the refractory wall can substantially improve the thermal efficiency of the radiant section of a steam cracking furnace. Formulations can be optimized to fine-tune the performance. Moreover, applying the high emissivity coatings on the external surface of the reactor tubes can additionally improve the surface homogeneity and eliminate hot spots. Combining the advanced coil materials and novel 3D reactor designs can lead to more uniform heat transfer, which increases run lengths, product selectivities as well as lifetime of the furnace, while simultaneously reducing CO<sub>2</sub> emissions. In addition, it is believed that the current design of the 3D reactors is far from optimal, and that CFD can lead to the design of radiant coils with intensified performances. By implementing the 3D designs the on-stream time of the cracking furnaces can be drastically improved. It is key to focus not only on start of run conditions, but also on evaluating the performance over a complete run length because initial larger pressure drops can be misleading for the overall performance over the time on stream.

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