

Electrification of methane conversion process through induction heating in graphite felt catalyst

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Abstract

The conventional production of ethylene is performed in steam cracking furnaces, which require heating a stream of light alkanes above 700°C through a natural gas combustion. As a result, the carbon footprint of a ton of ethylene is important, and the fossil fuel mass required for its production is larger than the mass of the ethylene produced. Non-oxidative coupling of methane to ethylene could serve as an alternative pathway for ethylene production; on top, if such process is electrified, the process could have almost zero emissions.

Electrification at scale of such a process is challenging as it must work in the range of 10kt – 1 Mt per reactor per year. TotalEnergies and SIMTEC developed a COMSOL Multiphysics® model to investigate this new process and evaluate the induction heating of a graphite felt catalyst.

Coil height, current frequency and tube radius are varied to identify important design variables, discard poor configurations, and shortlist a relevant starting point for further experimental investigations.

Keywords: Induction, electrification, graphite felt, catalyst.

Introduction

The global chemical industry faces increasing pressure to decarbonize its operations, particularly in energy-intensive processes such as ethylene production. Traditional pathway consists in heating a stream of light alkanes above 650°C through a methane combustion inside a “firebox”. The later results in CO₂-lean flue gas, which is expensive to capture and is a significant contributor to global warming. Along with electrification efforts for such furnaces or with replacement of used fuels to non-fossil alternatives, important volumes of methane would become available as feedstocks. To address this environmental and industrial challenge, non-oxidative coupling of methane (NOCM) to ethylene presents a promising alternative pathway. However, its industrial viability hinges on the ability to electrify the process at scale, targeting production capacities between 10 kt and 1 Mt per reactor per year. This study, conducted by TotalEnergies and SIMTEC, explores the electrification of NOCM through induction heating of a graphite felt catalyst. Using a COMSOL Multiphysics® model, we investigate key design variables, such as coil height, current frequency, and tube radius to efficiently understand the influence of each parameter on electrical yield and the heating homogeneity.

1. Description of the numerical model

Induction heating consists in generating electrical

currents without geometrical contact between the coil inductor and the components which require heating. Instead, the power is transferred through an AC electromagnetic field. The current generated in the components dissipates heat through Joule effect. By carefully designing the coil, the heated components and power supply, high yield can be achieved.

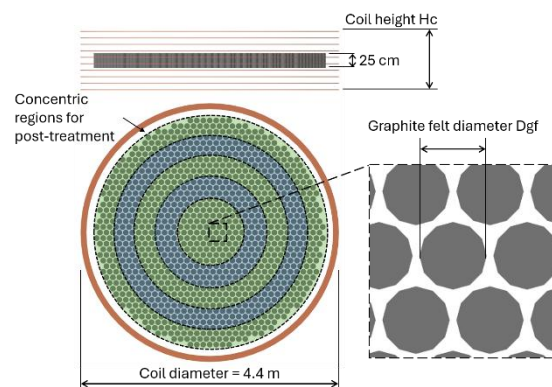


Figure 1. Coil geometry and graphite felt heated cylinders

1a. Geometry

To meet the high production capacity target, a 4.4m diameter coil is selected, while its height H_c is a parameter of the study (see Figure 1). Within the coil, many graphite cylinders are arranged according to a hexagonal pattern to form the reactor.

The cylinder height is 25 cm, and they are individually placed within high resistivity tubes (not represented) to force the stream of gas through

the graphite felt. The cylinder diameter D_{gf} is another parameter of the study. Figure 1 also displays 5 concentric groups of graphite felt cylinders for post-treatment purposes. Top-down symmetry is used and only a 30° portion is simulated to reduce the degrees of freedom without loss of precision. A spherical volume of air surrounds the coil and the cylinders, with an external layer to apply an infinite element domain.

1b. Governing equations and mesh

Ampère's law is solved everywhere in the frequency domain, using the magnetic potential vector $\mathbf{A}$ and is here described using bold to identify vectors:

$$\begin{aligned}\nabla \times \mathbf{H} &= \mathbf{J} \\ \mathbf{B} &= \nabla \times \mathbf{A} \\ \mathbf{J} &= \sigma \mathbf{E} + j\omega \mathbf{D} \\ \mathbf{E} &= -\nabla V - j\omega \mathbf{A}\end{aligned}$$

With $\mathbf{H}$ the magnetic field, $\mathbf{J}$ the electrical current, $\mathbf{B} = \mu_0 \mu_r \mathbf{H}$ the magnetic induction field, μ_0 the permeability of the vacuum, μ_r the relative permeability, $\sigma = 1/\rho$ the electrical conductivity, ρ the electrical resistivity, $\mathbf{E}$ the electric field, j the imaginary unit number, $\omega = 2\pi f$ the pulsation, f the frequency, $\mathbf{D} = \epsilon_0 \epsilon_r \mathbf{E}$ the electric induction field, ϵ_0 the permittivity of the vacuum, ϵ_r the relative permittivity and V the electric potential. To correctly account for the very low frequency cases, the current conservation is solved in the air:

$$\nabla \cdot \mathbf{J} = 0$$

As $\sigma = 0 \text{ S/m}$ in the air, this equation reads:

$$\nabla \cdot (-j\omega \epsilon_0 \epsilon_r \nabla V + \omega^2 \epsilon_0 \epsilon_r \mathbf{A}) = 0$$

At the surface of the coil, an impedance boundary condition is applied:

$$\sqrt{\frac{\mu_0 \mu_r}{\epsilon_0 \epsilon_r - j\frac{\sigma}{\omega}}} \mathbf{n} \times \mathbf{H} + \mathbf{E} - (\mathbf{n} \cdot \mathbf{E}) \mathbf{n} = 0$$

Where μ_r , ϵ_r and σ are the properties of the copper. To apply a current in the coil, a ground is applied on one end of each coil turn, while a different electric potential is applied on each turn end. This potential is determined thanks to one ODE for each turn, in order to obtain a coil current of 3000 A.

At the spherical outside surface of the infinite element domain, a magnetic insulation is applied:

$$\mathbf{n} \times \mathbf{A} = 0$$

with $\mathbf{n}$ the unit vector normal to the surface.

On the 30° portion lateral boundary conditions, in the air and in the graphite felt cylinders, $\mathbf{n} \times \mathbf{A} = 0$ is also applied.

The top-down symmetry is accounted for through a perfect magnetic conductor boundary condition:

$$\begin{aligned}\mathbf{n} \times \mathbf{H} &= 0 \\ \mathbf{n} \cdot \mathbf{J} &= 0\end{aligned}$$

Finally, the unicity of the solution is obtained by adding a gauge fixing on the potential magnetic vector:

$$\nabla \cdot \mathbf{A} = 0$$

Since all equations are linear, the final result is scaled at the stage of post-treatment to obtain 2 MW in the graphite felt cylinders.

The equation system defined is solved on an adapted mesh for the different parameter combinations.

2. Main results

The physics being linear, all results are scaled to obtain 2MW power deposition in the graphite felt.

2a. Influence of coil height H_c

Different coil heights are investigated, knowing that it would lead to different magnetic induction field homogeneity. Figure 2 shows homogeneity results across the reactor radius for 10cm diameter graphite felt cylinders. Induction field squared is rather homogeneous across the reactor for 4 and 7m high coils but is more concentrated at the outside of the reactor for 1m high coil. The theoretical infinitely long coil creates a constant internal magnetic field. In the present configuration, infinity almost starts at 4m, and the result is a homogeneous power density for the 4 and 7m cases.

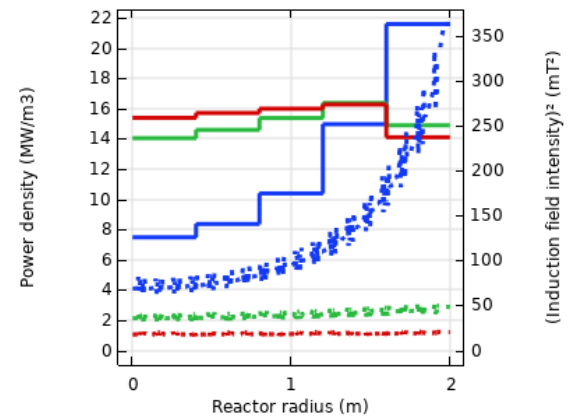


Figure 2. Influence of coil height H_c on reactor power homogeneity for $D_{gf} = 10 \text{ cm}$, $f = 20 \text{ kHz}$ and $\rho = 0.02 \Omega \cdot \text{cm}$ in the graphite felt; solid line: power density averaged in the 5 concentric post-treatment groups, dotted line: induction field intensity squared, blue colour: $H_c = 1 \text{ m}$, green colour: $H_c = 4 \text{ m}$, red colour: $H_c = 7 \text{ m}$

This means that it is possible to induce similar currents and generate similar power losses in all the graphite cylinders. And this, despite some of them being close to the coil and some other close to the centre.

There is however a power yield drawback to longer coils. Figure 3 displays the power yield for the same coil heights: 1, 4 and 7m. The coil frequency

is varied, and the higher the frequency, the higher the power yield: more of the desirable Joule power losses are concentrated in the graphite felt cylinders, and less undesirable losses are in the coil. However, at identical power yield, the longer coils require the higher frequencies. This may be an issue for the power line, and a careful balance between pros and cons must take place.

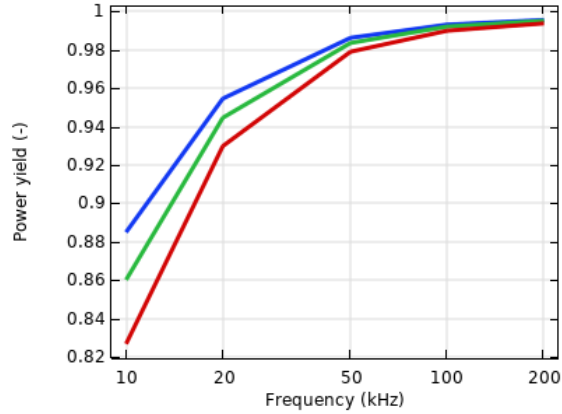


Figure 3. Influence of coil height H_c on power yield for $D_{gf} = 10$ cm; blue colour: $H_c=1$ m, green colour: $H_c=4$ m, red colour: $H_c=7$ m

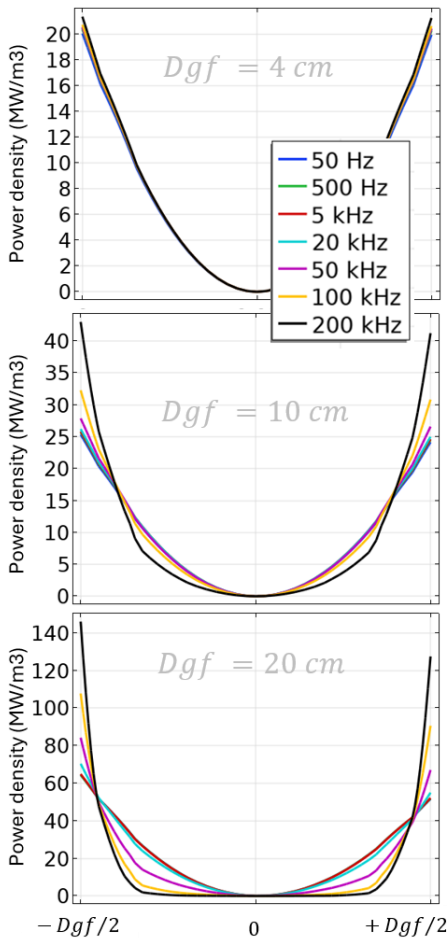


Figure 4 - Influence of graphite felt diameter D_{gf} on the power distribution within the graphite cylinder for a 1m coil.

2b. Influence of graphite felt diameter D_{gf}

Induced currents flow in a circular loop in the felt cylinders. This loop axis is aligned with the cylinder axis. Thus, the centre of the cylinders has no current flow and no local power losses. This can be observed in Figure 4. The $D_{gf} = 4$ cm case shows this pattern for all frequencies.

This means that although all cylinders can receive similar power from the coil, there are inherent inhomogeneities within each cylinder: the outer layer receives more power than the core. However, for larger graphite felt diameters, an additional effect takes place: skin effect. For a semi-infinite flat body, the thickness of this skin is:

$$\delta = \sqrt{\frac{2\rho}{\omega\mu_0\mu_r} \left(\sqrt{(1 + (\rho\omega\epsilon_0\epsilon_r)^2)} + \rho\omega\epsilon_0\epsilon_r \right)}$$

In the present case, $\rho\omega\epsilon_0\epsilon_r \ll 1$ and $\delta \approx \sqrt{2\rho/\omega\mu_0\mu_r}$. The higher the frequency, the smaller the depth at which the currents can be induced. If the practical skin depth is smaller than the cylinder radius, then the inhomogeneity within the cylinder is further reinforced: the outer layer receives more power, and the core even less. The skin effect power concentration slightly appears at 200kHz for the $D_{gf}=4$ cm cylinders, 50kHz for the $D_{gf}=10$ cm cylinders and 20kHz for the $D_{gf}=20$ cm cylinders.

In addition to power concentration, the cylinder diameter affects the power yield similarly to the coil height. Figure 5 shows the trend, and a minimum 90% power yield is attained for frequencies beyond 6kHz for $D_{gf}=20$ cm cylinders, 13kHz for $D_{gf}=10$ cm cylinders and 45kHz for $D_{gf}=4$ cm cylinders.

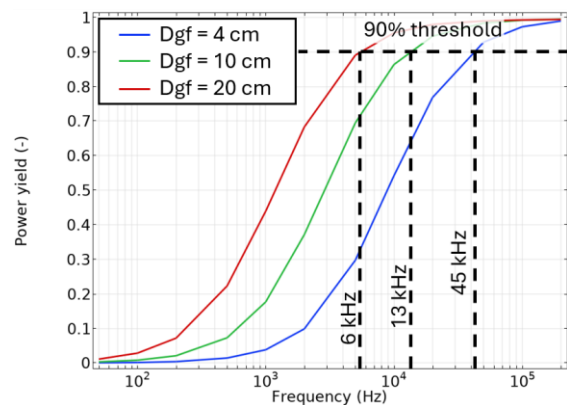


Figure 5. Influence of the frequency on the yield, for different graphite felt diameters and a 1m coil.

The effect of frequency on power yield and power homogeneity needs to be accounted for in the design of a new reactor, but this effect depends on the graphite felt cylinder diameter, the coil height and the graphite felt resistivity which is not investigated in this document.

2c. Discussion

To summarize the different effects, Figure 6 is proposed. It gathers the upper frequency limit, to minimize the power density inhomogeneity in the cylinders, and the lower frequency limit, to make sure the power yield is above a 90% threshold. Therefore, the reactor design parameters for a 1m coil must be selected within the green area defined in the figure. The skin depth expression is plotted for reference, and it roughly corresponds to the upper frequency limit.

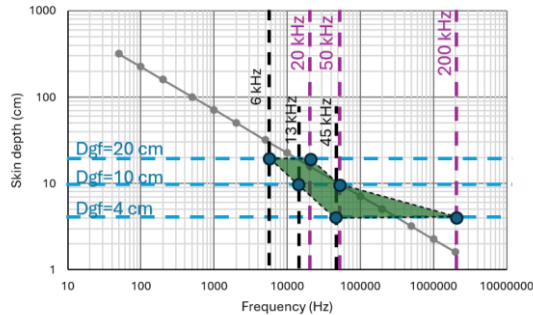


Figure 6. Identification of the different physics constraints on a frequency - dimension graph for a 1m coil.

Such figure must be generated for each cylinder height and each graphite felt resistivity considered.

Conclusions

Thanks to a numerical approach, it is possible to show that induction heating is possible within a large number of elements arranged in a pattern. Such element can be graphite felt cylinder that let methane gas pass through, thanks to their porosity. Being both a catalyst and a heating element thanks to the induced currents, this reactor design can harness electrical power to activate chemical reactions at high-power yield.

However, induction heating systems need to be carefully designed, and its sweet spot must be identified to avoid time-consuming and expensive prototyping. A lower and upper operating frequency can be determined for each geometrical and material configuration investigated. Therefore, the design of equipment such as the coil, graphite felt cylinders, the vessel and the power supply can be informed by physics.

A patent is filed to describe the new process [1]

References

[1] WO2023072996A1

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