MODELLING COMBUSTION AND THERMAL NO_X FORMATION IN ELECTRIC ARC FURNACES FOR THE PRODUCTION OF FERRO-SILICON AND SILICON-METAL

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ABSTRACT

Combustion and thermal NO_x formation have been modelled using the Computational Fluid Dynamics (CFD) commercial code Fluent 6.1. The aim was to predict and give a better understanding of the combustion in electric arc furnaces for the production of ferro-silicon and silicon metal. Another important objective was to assess the impact of a number of operating parameters on the NO_x formation. The CFD tools used are described with a special focus on models for species and turbulence transport. Sensitivity studies have been carried out to study the influence of several parameters: sub-models, process gas composition and distribution at the charge surface and influence of the presence of SiO.

The main conclusions are:

- *Using the combination of a realistic design, boundary conditions and models, the simulated NO_x level is in quantitative agreement with measurements. The presence of water vapor is particularly important because of its influence on the specific heat of the mixture and resulting temperature.*
- *Release of significant amounts of SiO from the charge surface results in an important increase of NO_x, even if we applied a simplified combustion model for SiO gas. However, because of lack of thermal and compositional data for blowing situations (when SiO flows directly from the arc cavity through open channels to the charge surface), no quantitative validation is possible. The way SiO is distributed (uniformly or as a jet) has a limited impact on the result in our computations.*

1. INTRODUCTION

Norway has decided to fulfill the Gothenburg protocol that focuses on limiting the emission of four gasses, among which we find nitrogen oxides or NO_x. NO_x is by far the gas for which the largest reduction is needed. A reduction of approximately 30% in NO_x in the period 2005-2010 is necessary. NO_x are detrimental to the environment, the main reason being that high NO_x level in the atmosphere result in acid rains.

The Norwegian ferroalloy industry has been working on reducing its NO_x emissions over ten years through several research programs. These programs have been very successful. Specific low cost process adaptations have lead to very significant reduction of NO_x levels, typically 50%. Theory, modeling, advanced measuring techniques and industrial trials have been very important in understanding the main phenomena.

In the present paper, some of the modeling work using Computational Fluid Dynamics (CFD) is presented. These simulations show good agreement with observations in ferro-silicon and silicon metal plants.

2. THEORETICAL BACKGROUND

2.1 Overview Over NO_x Formation Mechanisms

There are four main NO_x formation mechanisms: thermal NO_x, fuel NO_x, prompt NO_x and nitrous oxide mechanism. The thermal NO_x mechanism is thought to be dominant in electric arc furnaces producing ferro-silicon and silicon-metal because it is dominant at high temperature when air is present. As a matter of fact, under the hood of these furnaces, in the combustion region over the charge, there are hot zones (typically above 1600°C) in which both nitrogen and oxygen are available, allowing the Zeldovich mechanism (direct oxidation of the atmospheric nitrogen leading to thermal NO_x). Figure 1 gives an overview of the relative importance of the different NO_x formation mechanisms, as a function of temperature, for coal combustion [1]. This is relevant to the ferroalloy industries where it is used biological materials, coal and coke as reduction materials. Fuel NO_x, coming from the oxidation of the nitrogen component in the reduction materials, is thought to be the other significant source of NO_x. In the present paper the main focus has been on modeling thermal NO_x.

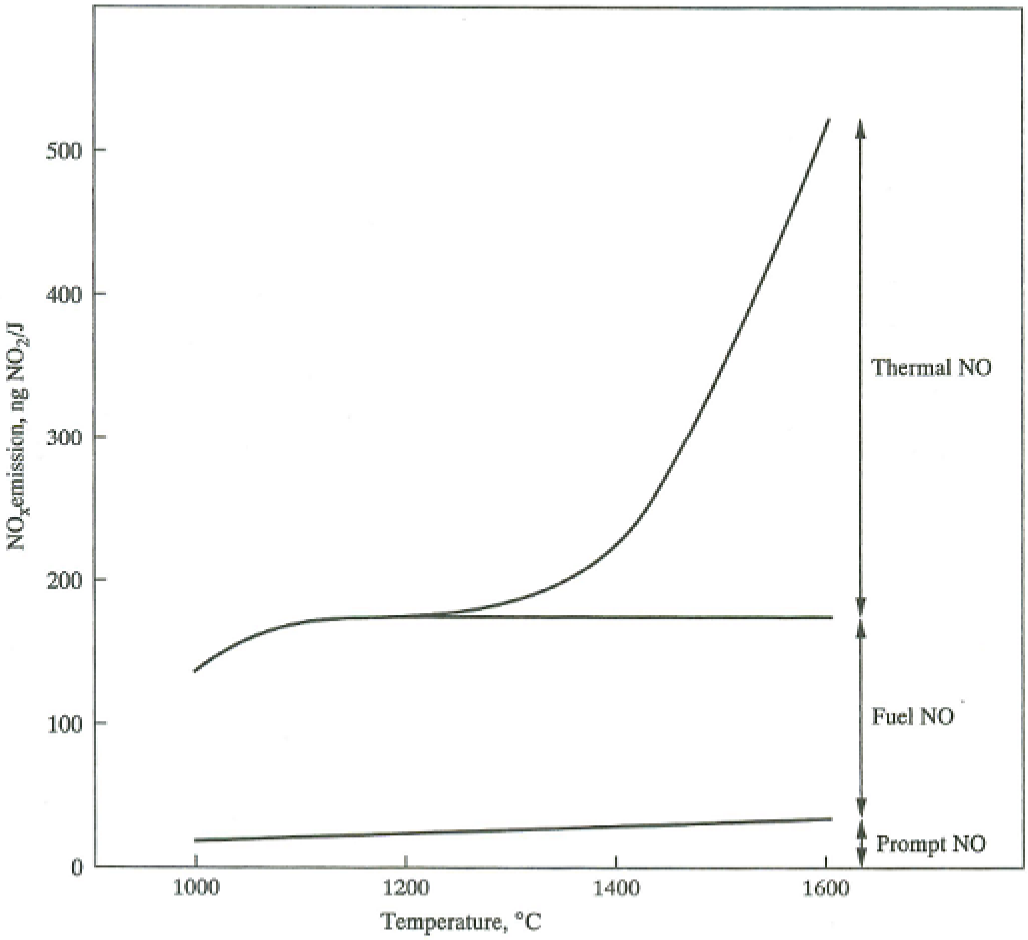

Figure 1: Typical contributions of three NO_x mechanisms in coal combustion as a function of temperature [1]

2.2 Methodology

Several combinations of physical models (e.g. turbulence and chemistry) and boundary conditions have first been tested on simplified 2D and 3D geometries. Based on this preliminary study, a set of models was chosen and used on a realistic 3D design for a typical furnace producing silicon metal. In the present paper, only this last part and the corresponding models are presented.

2.3 Computational Models

By modeling combustion, the temperature and species distribution can be obtained. In this context, turbulence, chemical reactions and their interactions need to be thoroughly modeled to get reliable results. The modeling work was performed using the commercial CFD code Fluent 6.1.22 with its standard models. Details of the equations for the models are not presented here. They can be found in the documentation of the code [2].

Turbulence model

For most industrial applications, there is a need to transform the Navier-Stokes equations in such way that the small-scale turbulent fluctuations do not have to be directly simulated. The Reynolds averaging approach represents transport equations for the mean flow quantities only, with all the scales of turbulence being modeled. It is generally adopted for practical engineering calculations. Many engineering models, such as the k-ε models, use Reynolds averaging. Here, the ReNormalization Group k-? model was used for computations because it gave more satisfactory results in our previous studies [3].

Combustion model

Two models have been tested: the Eddy Dissipation model [4] and the Non-premixed Combustion model, which is based on a mixture fraction. The Eddy Dissipation model assumes that as long as fuel and oxidant are mixed, they will react until one of the reactants disappears (mixed-is-burnt). This model will overestimate the reaction rate in cold zones where the chemical kinetics is the actual limiting factor [3]. However, this model was used when SiO combustion was included, despite being less advanced that the Non-premixed Combustion model. The reason was that we were not able to introduce this actual reaction in the thermochemistry tables used by the latter model.

SiO combustion model

Combustion of SiO is special because the kinetics appears to be much faster than other components, in particular CO and hydrocarbons. The adiabatic flame temperature is very high [3]. In addition, the final product is a solid product, silica dust. Developing a good model for a solid product requires important work that should be done in the future. In the present paper, SiO_2, the product of the combustion is assumed to be gaseous but the heat released during the solidification process to obtain silica dust is included in the heat of combustion. An important aspect is the distribution of SiO gas from the charge surface. Under a “quiet” operation of the furnace, the charge surface mass flux distribution of SiO is rather uniform and follows the flux distribution of other furnace gases. Under blowing however, large quantities of SiO gas flow directly from the crater into the atmosphere above the charge through one or several channels. This is known to result in high NO_x formation [5].

Radiation model

The Discrete Ordinate model was used. With this model, wall-to-wall radiation and variable absorption coefficient based on gas composition are included.

NO_x formation model

Here, only thermal NO_x, which is thought to be the main mechanisms for silicon furnaces, have been modeled. The NO_x transport equations are solved based on a given flow field and combustion solution. In other words, NO_x is postprocessed from a combustion simulation. It is thus evident that an accurate combustion solution becomes a prerequisite for an accurate NO_x prediction. For example, thermal NO_x production doubles for every 90 K temperature increase when the flame temperature is about 2200 K. Great care must be exercised to provide accurate thermophysical data and boundary condition inputs for the combustion model. Appropriate turbulence, chemistry, radiation and other sub-models must be employed. To be realistic, one can only expect results to be as accurate as the input data and the selected physical models. Under most circumstances, NO_x variation trends can be accurately predicted but the NO_x quantity itself cannot be pinpointed. The focus in the present work has been to work out appropriate sub-models and boundary conditions for combustion to give realistic temperature and species concentration fields, and not to assess different NO_x formation models. Consequently, the only model we used for all our computations for NO_x formation is the thermal NO_x model, with assumed equilibrium concentration of the radical O.

3. COMPUTATIONAL SET UP

3.1 Geometry And Grid

The geometry corresponds to typical dimensions of a silicon metal of ferro-silicon furnace. The modeled geometry is shown in Figure 2. The diameter of the furnace is approximately 12 m and the electrodes have a diameter of 1.7 m. In addition, there is an air volume around the furnace. The meshing grid for the computational domain has approximately 280 000 cells. Most of the cells are hexahedral except for in some zones at the interface between the off-gas channels and the hood where tetrahedral cells were used. The grid has been refined near the charge surface so that right above the charge surface, cells have a typical dimension of 10 mm.

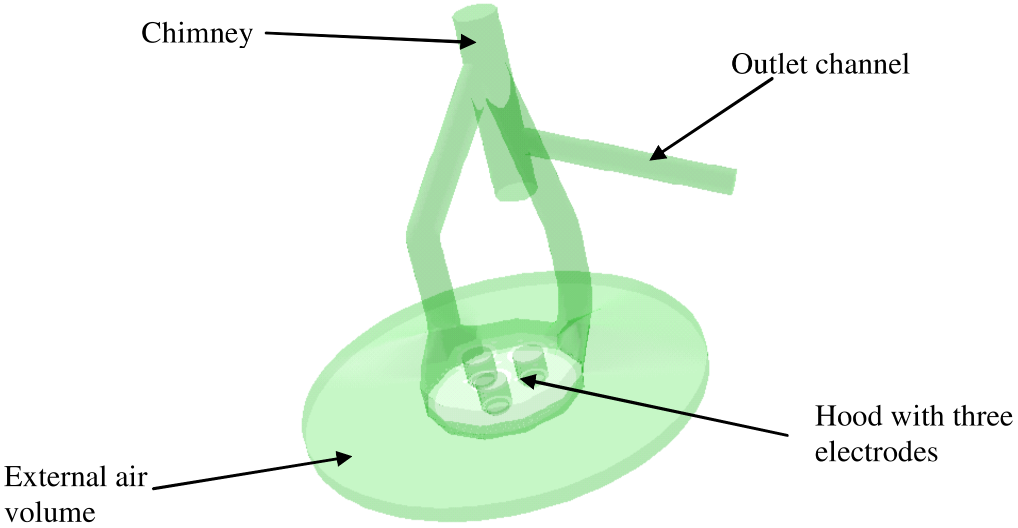

Figure 2: Modelled geometry

3.2 Boundary Conditions

For the furnace top, the off-gas system, the charging pipes and the electrodes, the heat transfer is characterized by giving the wall materials and heat transfer coefficient to the surroundings. For the side walls of the furnace and the ports, the heat transfer to the external volume is calculated based on the materials conductivity and the flow field.

The process gas coming from the charge surface is distributed between separate inlet zones in order to approximate the actual distribution at the charge surface. The total mass flow rate of process gas is approximately 5 kg/s. The major gas flow entering the hood is air. Air is sucked into the furnace chamber through ventilation openings in the side walls and under the ports. Some air is also injected from the charging pipes and around the electrodes. The total air flow rate is about 50 kg/s so that the ratio q_{air}/q_{gas} is approximately 0.1. The gas temperature at the inlets is: 293 K for the air and 1623 K for the process gas.

Two cases have been studied. The first set is named *No jet cases* and the second is named *SiO jet cases*:

"*No jet cases"*: The process gas has the same composition all over the charge surface. When pure CO is injected and the Eddy Dissipation model is used, the case will be designated as *Base case*.

"*SiO jet cases*": Here the aim was to model the furnace when the blowing phenomenon occurs. A small zone in the centre of the electrodes approximates a 0.2 m diameter disk. Pure SiO is introduced from this zone, so called *Jet zone*.

3.3 Results from "No jet cases"

In these cases, the flow coming from the charge has a uniform composition. From one case to another, only the composition of the process gas or the model for combustion have been changed. Either the Eddy Dissipation model or the Non-premixed Combustion model with the equilibrium submodel have been used. When SiO was introduced only the Eddy Dissipation model could be used.

In the *Base case*, only CO comes from the charge surface and the Eddy Dissipation model is used. The other cases are named *No jet N*, where *N* varies between 1 and 4. The composition and mass flow rates from the process gas are provided in Table 1.

Table 1: Overview of the inlet composition, total mass flow rate of process gas (q_{gas}) and model for combustion (ED and NC stand respectively for Eddy Dissipation and Non premixed Combustion). q: mass flow rate (kg/s), Y: mass fraction in the process gas. The flow rate of air is constant q_{air} = 50 kg/s

Case	*CO*		*SiO*		CH_4		H_2O		H_2		q_{gas}	*Model*
	q	Y	q	Y	q	Y	q	Y	q	Y		
Base case	4.98	1	0	0	0	0	0	0	0	0	4.98	ED
No Jet 1	4.98	1	0	0	0	0	0	0	0	0	4.98	NC
No Jet 2	3.57	0.80	0	0	0.086	0.02	0.62	0.14	0.21	0.05	4.49	NC
No Jet 3	4.47	0.85	0.79	0.15	0	0	0	0	0	0	5.25	ED
No Jet 4	4.10	0.84	0	0	0	0	0.78	0.16	0	0	4.88	NC

The main results of the simulations are presented in Figure 3.

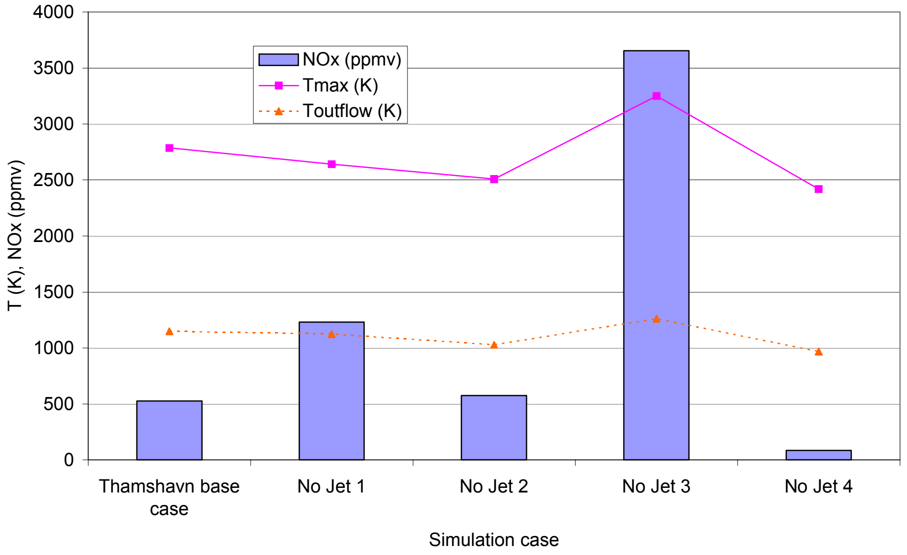

Figure 3: Comparison of NO_x, maximum temperature and average temperature at the outlet for different cases without SiO jets

The base case does not include water vapour, so when compared to measurements both temperature and NO_x are high. There is complete combustion for all the models so that the change of combustion model does not result in significant change in off-gas temperature, as can be observed by comparing the results from the *Base case* (Eddy Dissipation) and *No Jet 1* case (Non-premixed Combustion). For the pure CO cases, the Non-premixed Combustion model results in a lower maximum temperature but NO_x are higher. This indicates that NO_x producing hot zones must be larger with the Non-premixed Combustion model. When more realistic process gas composition are used, in particular with the inclusion of water vapour which increases significantly the specific heat of the gas, as in *No Jet 2* and *No Jet 4* cases, temperatures and NO_x drop and are in a range in agreement with measurements. Lastly, for No Jet 3, with 15w% SiO and CO, temperatures and NO_x increase dramatically as seen in a previous study [3].

For the *No Jet 1* case and even more for the *No Jet 3* case, the level of NO_x is very high, above what has been measured on real furnaces, indicating that the composition chosen for the process gas (resulting in total absence of water vapour in the product mixture) does not correspond to a realistic industrial composition. It is well know that significant amounts of water is always available from sources such as leaking water cooling pipes and wet raw materials.

The *No Jet 4* case corresponds to a realistic heat content and flow rate of process gas (the composition was adjusted to match the measured temperature at the outlet). In this case, 85 ppmv NO_x is obtained, which is in good agreement with measurements under a "quiet" operation. The average measured NO_x lies in a typical range 100 to 250 ppmv. In our simulation, only thermal NO_x has been modelled. By adding the contribution from fuel-NO_x (that was not modelled here), the NO_x level should come right into the measured range.

3.4 Results from "SiO jet cases"

In these cases, a zone with a shape close to a 0.2 m diameter disk has been used as inlet for pure SiO to model the blowing phenomenon that is when SiO flows directly from the arc cavity through open channels to the charge surface. The flow rate of air was kept constant and equal to 50 kg/s. Only the Eddy Dissipation model was used since SiO is present in the simulations. The overview over the main inlet data is provided in Table 2. The velocity in the inner zone of the charge surface is approximately 0.9 m/s. The velocity of the SiO jet has been varied from this velocity to 30 m/s. The *No Jet 5* case has been computed to provide a case without the jet, but having the same composition as the *SiO-Jet 3 case* and thereby allow an assessment of the influence of the spatial SiO flux distribution through the charge surface.

Table 2: Overview of the mass flow rates of CO and SiO in the different zone with SiO jets and relevant cases without SiO jets

Case	*Inlet rest of charge*			*Inlet jet*			q_{gas} *(kg/s)*
	CO (kg/s)	SiO (kg/s)	V_{inner} (m/s)	CO (kg/s)	SiO (kg/s)	V_{jet} (m/s)	
Base case	4.98	0	0.88	0.003	0	0.88	4.98
No Jet 3	4.47	0.79	0.88	0.003	0.001	0.88	5.25
SiO-Jet 1	4.98	0	0.88	0	0.005	0.88	4.98
SiO-Jet 2	4.98	0	0.88	0	0.05	8.8	5.03
SiO-Jet 3	4.98	0	0.88	0	0.18	30.0	5.16
No-Jet 5	4.98	0.18	0.91	0.003	0.001	0.91	5.16

The main results are presented in Figure 4.

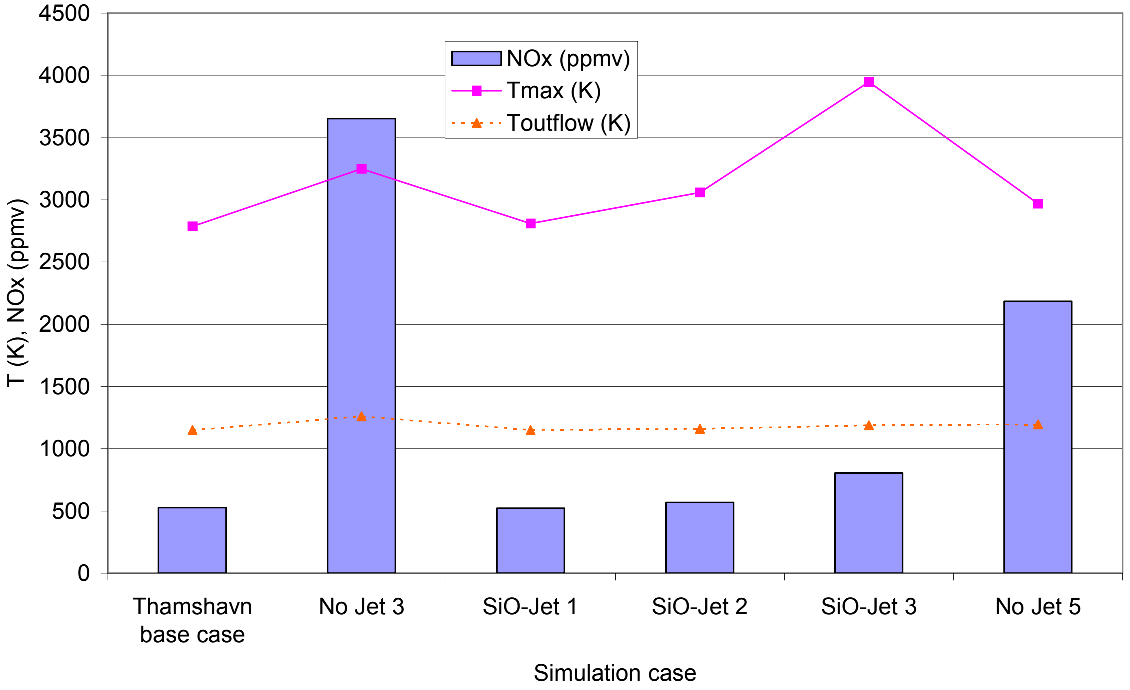

Figure 4: NO_x concentration, maximum temperature and average outlet temperature for SiO-jet cases, together with relevant cases without jets

Increasing both the SiO jet velocity and the SiO mass flow rate leads to an increase in maximum temperature and NO_x. However, it does not modify significantly the average outlet temperature because of the relatively small quantities involved. This is in good agreement with the observation that NO_x and silica dust are well correlated but this is not the case for NO_x and off-gas temperature [5].

When the same amount of SiO as for the *SiO-Jet 3* case is distributed over the charge surface (*No Jet 5* case), the maximum temperature decreases but the total NO_x increases significantly. This may be due to the fact that the SiO jet was put at the centre of the furnace, in a relatively oxygen poor region (zone where most process gas is introduced). Therefore, a more evenly distribution allows SiO to be in oxygen rich region and result in larger high temperature zones where thermal NO_x can be produced. In this particular case, a jet-like distribution for SiO does not promote NO_x compared to a uniform distribution. A test of the position of the SiO jet should be done in the future to assess the sensitivity to this parameter.

4. CONCLUSION

Combustion processes typical for a furnace producing ferro-silicon or silicon metal have been modelled using CFD. SiO combustion was modelled using the simplifying assumption that SiO_2 was gaseous (no change in volume coming from the solidification but the energy released by the solidification process is included in the total combustion enthalpy). Only thermal NO_x (which is believed to be the major contribution) has been modelled.

When adapted to realistic heat content for the process gas, together with realistic boundary conditions and geometry for a full furnace and off-gass systems, the NO_x level obtained are about 90 ppmv while measurements have shown average values over day periods in the range 100 to 250 ppmv (16v% oxygen). Therefore, the correct combination of models and realistic conditions result in a good quantitative agreement with measurements. Introducing methane and water vapour in the process gas results in significantly lower temperature, partly because of the reduction of fuel in the process gas as far as water vapour is concerned, and partly because water vapour in the product (also product of methane combustion) increases significantly the specific heat of the gas.

The Non-premixed Combustion model gives results that seem in better quantitative agreement than the Eddy Dissipation model, in most cases. However, unexpected limitations of this model made it impossible to use it with SiO gas.

In the simulations, increasing the quantities of SiO gas released from the charge result in large increases of maximum temperatures and NO_x without resulting in any sharp or large increase of the off-gas temperature. This is what is observed in operation under the blowing phenomenon. The qualitative trend for SiO injection is therefore correct in the computations. The correct qualitative trend can be observed even without kinetic effects: the higher heat released by SiO is sufficient to provoke the NO_x increase. In our simulations, the NO_x formation did not depend on how the SiO mass flux through the charge surface was spatially distributed (even distribution or jet).

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