MODELLING PCDD/PCDF FORMATION IN THE EFFLUENT GAS OF A SINTER PLANT

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Introduction

PCDD/F as well as other chloroaromatics are an unwanted by-product of most metallurgical processes, both in primary metal production and in recycling. Iron ore sinter plants are a notorious source of PCDD/PCDF and most plants in the E.U. have taken both primary and secondary measures, to reduce formation and eliminate PCDD/F from the effluent gas. In sinter plants a mixture of iron ore, recycled ferriferous products and various additives (e.g. lime), together with coke breeze as a fuel, are converted into a sintered agglomerate with a chemical composition and a size distribution, optimal for blast furnace operation. In the framework of the MINIDIP-project (**Mini**mization of **Di**oxins in Thermal Industrial **P**rocesses: Mechanisms, Monitoring, Abatement) the formation of PCDD/F were studied as a function of temperature, time, oxygen, and inhibitor addition. The resulting kinetic data are introduced into a computer fluid dynamics (CFD) model, featuring a geometric representation of the dividers and flue gas duct of a sinter plant, and the temperature and gas flow field is modelled, as well as the trajectories and fate of entrained particulate of a sizes of 10, 30, 100, and 300 μm. This CFD model is used to estimate the de-novo formation of PCDD/F from the moment the gas emerges from the sintering layer until its cleaning, in order to define the role of this part of the plant in PCDD/F-formation, as well as the identification and extent of possibilities for minimizing PCDD/F emissions.

Method

For modelling the flue gas duct the commercial program FLUENT® is applied[1]. The CFD solution process is subdivided into three main steps. Pre-processing, the first step, consists of building and analysing the flow model. After pre-processing, the CFD solver carries out the calculation of the temperature and flow field including particle transport and chemical reaction. Post processing, involving visualization and interpretation of the data, is the final step in the CFD process.

Results and Discussion

The sinter plant consists of a long continuously moving horizontal belt, on which a layer of moist iron ore and additives is ignited on top, to initiate sintering. Air is aspired through the layer, so that the sintering zone is slowly moving downwards, while the belt moves from the feed side to the discharge side. The off-gas is collected in a series of plenum chambers, situated underneath the belt and linked by gas dividers with two gas collectors (cf. figure 1), which lead the gas to an electrostatic precipitator. Coarse and fine dust drop through the moving belt and are either collected in hoppers at the bottom of the collector or, if they are fine enough, entrained. After kinetic study of the 'de novo' formation the latter is modelled on the basis of the operating conditions of a typical sintering plant. Since both collectors are symmetrical to each other, only

one of them is modelled. Based on a three-dimensional CAD geometry an unstructured grid with approx. 250,000 cells is modelled. The cell resolving is optimised to the effect, that the cells in areas of high-expected gradients have a smaller size. The gas temperature and flow fields are calculated as stationary fields respecting the individual gas flow rates and temperatures at the dividers (cf. figure 2). The temperature losses are calculated using a heat transfer coefficient and turbulence is considered by a k-ε-model.
Particles of different sizes enter the computational domain together with the process gas and at the same temperature. The particles are seen as a discrete phase carrying out a momentum and heat transfer with the gaseous phase. Contact between particles and walls results in elastic strokes (reflection of the particle) and collision of particles with funnel walls leads to their leaving the computational domain. Additionally, the particles leave the computational domain through the gas outlet on their way to the filter.

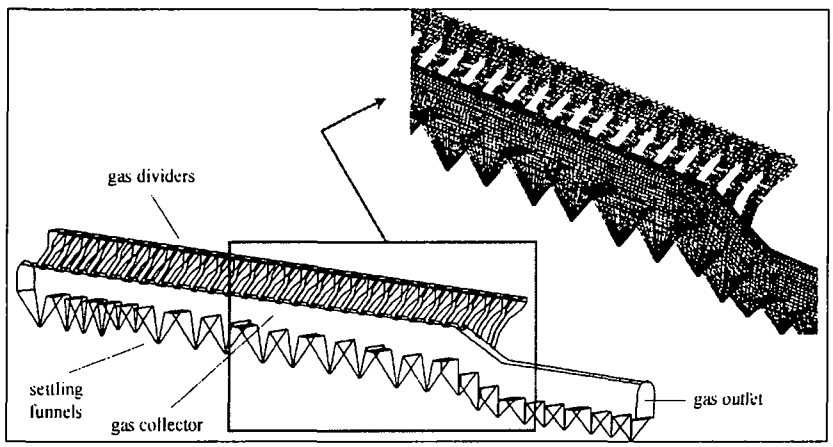

Figure 1
The computational domain of the flue gas duct with a length of some 115 m, a width of 8 m and a maximum height of 15.5 m (representation of the outside walls)

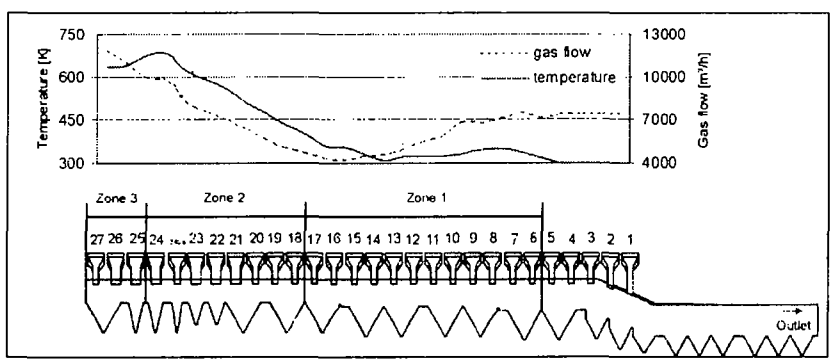

Figure 2
Designation of the zones and the dividers as well as temperature and gas flow distribution on the dividers

As expected, the calculated flow and temperature fields (cf. figure 3 and 4) show a main flow towards the gas outlet as well as the effect of the entrance of hot gas (zone 3 and 2) or cold gas (zone 1) on both the longitudinal as the axial temperature distribution; moreover, at the end of the collector and in the series of funnels various recirculation zones with low velocity are formed. In the middle of the collector a "flow roll" develops resulting from the feeding of process gas stepwise becoming cooler from zone 3 to zone 1.

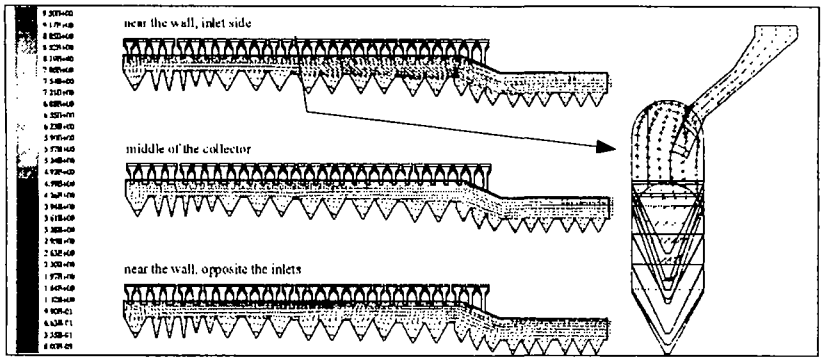

Figure 3
The flow field [m/s] in the flue gas duct (left figures: vertical cuts, right figure cut in cross section)

Particle calculations have been performed separately for particles of 4 sizes between 10 µm and 300 µm, whereby the particles enter the collector equally distributed over all dividers. The results show clearly the dependency of particle inertia on its diameter. Larger particles have a straight-line flight path and generally leave the computational domain directly through the settling funnels. On the other hand, small particles show trajectories very similar to the flow field and up to 80% of them leave the flue gas duct via the gas outlet.

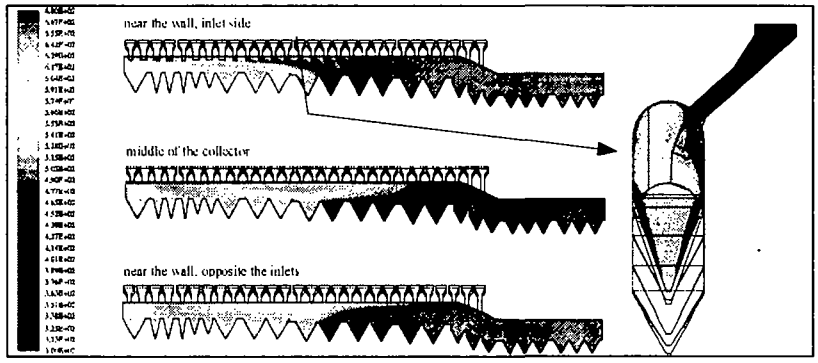

Figure 4:
The temperature field [K] in the flue gas duct (left figures: vertical cuts, right figure: cut in cross section)

For simulation of PCDD/F formation proceeding from the particles flight path it was necessary to program an User Defined Function (UDF) which can be dynamically linked with the FLUENT® solver. The PCDD/F formation kinetics are based on measurements of FZK[2]. The UDF is only active for moving particles and depends on the particle temperature upon leaving a cell as well as on its residence time and concentration in a cell. On the assumption that the turn over is small and without detailed information on the influence of other flue gas components PCDD/F-formation is calculated on a pseudo zero order kinetics basis (the rates depend on the oxygen content, but the latter is assumed to be essentially constant in the duct, since the temperature is already too low for a fast oxidation of TOC or residual carbon).

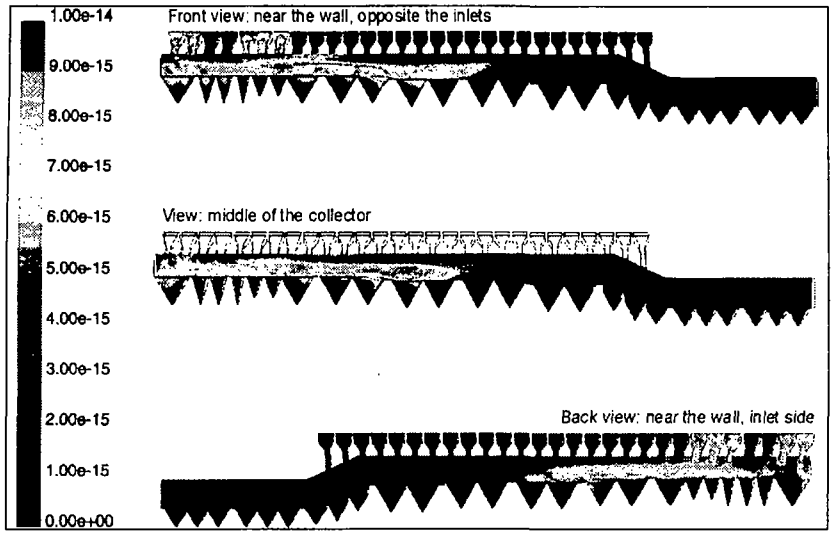

Figure 5: Contours of PCDD/F formation rates in kg/s at vertical cuts, calculated for particles with a size of 10 µm

Exemplary for all calculations considering PCDD/F formation a short description of the results of the calculation with the 10-µm particles is now presented. PCDD/F formation takes place at high rates in a number of high temperature inlets because of optimal temperatures, as well as in the collector below the inlets 16 until 22 above the funnel entries (cf. coloured zones in figure 5). In this region a high residence probability of particles from all zones is found as a result of the flow field. The particles of zone 1 contribute very little to PCDD/F formation. No PCDD/F formation is found in the region between the flow roll and the gas outlet as well as in the dividers of zone 1 and in a great part of the dividers of zone 2 due to the particle temperatures being below 200 °C or no particles are moved through this cells. Furthermore only small amounts of PCDD/F come from the funnels of zone 2 because the particles are eliminated when touching the wall. Figure 6 represents the spatial distribution of PCDD/F concentration in the flue gas duct.

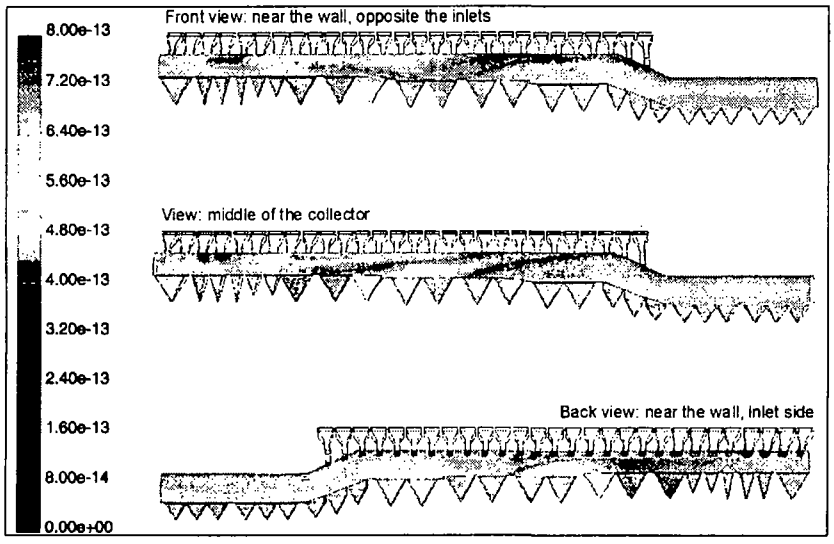

Figure 6: Contours of PCDD/F concentrations in kg TE /m³ (distribution of the formed PCDD/F, transported through the flow field in the computational domain) at vertical cuts, calculated for particles with a size of 10 µm

PCDD/F formation is modelled for the different particle sizes separately. The results (cf. table 1) show, that 10 µm particles contribute more to the PCDD/F formation in the flue gas duct than 300 µm particles.

Table 1: Maximum and average values for calculated PCDD/F rates and PCDD/F concentrations

Particle Size	PCDD/F formation rates [ng I-TEQ/s]		PCDD/F concentration [ng I-TEQ/m³]		
	Highest value	Average value	Highest value	Average value in the whole flue gas duct	Average value at the gas outlet
10 µm	0.035	$8.5\ 10^{-4}$	0.85	0.25	0.27
300 µm	0.021	$2.3\ 10^{-4}$	0.91	0.11	0.10

The mean calculated PCDD/F-concentration by de-novo formation from reactive dust in the gas collector is approximately 0.3 ng I-TEQ/m³. No additional formation in a large part of the collector or the electrostatic precipitator is to be expected, due to the low operating temperature (approx. 120°C). Typical emission concentrations, however, are 3 to 5 ng I-TEQ/m³. This means that the main PCDD/F formation takes place outsides of the computational domain, i.e. in the sinter bed. This is in agreement with earlier estimations showing that a low reactivity of the raw materials and sinter on the belt is more than offset by a combination of a much larger mass and residence time, when compared with mass and residence time of the much more reactive entrained dust. The de-novo formation of PCDD/F in the gas collector is less than 10% of the total PCDD/F formed in the sinter plant. This means that any primary measures to minimize PCDD/F formation have to aim at preventing the dioxin formation in the sinter bed.

This study is the first ever to compute PCDD/F formation on a basis of laboratory scale kinetic data in combination with CFD-simulation of particle temperature and residence time.

Acknowledgement

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1. Fluent User's Guide (1999): Version 5.5, FLUENT® Deutschland GmbH
2. Stieglitz L., Polzer J., Hell K., Weber R., Buekens A., Prakhar P. and Rivet F. (1999): Dioxins from the Sintering Process. (II) Samples and their propensity to form Dioxins, as derived by a 'de novo' laboratory test. Organohalogen Compounds, Vol. 41, 113-116