Kinetic study of abatement of low concentration of dibenzofuran by oxidation – Effects of co-reactants

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HIGHLIGHTS
- Our work consists in studying dibenzofuran destruction by oxidation.
- The oxidation mechanism of dibenzofuran radical consists of two competing channels.
- The dibenzofuran reactivity strongly decreases when the reactant is much diluted.
- The presence of methane or VOCs leads to better total oxidation of dibenzofuran.

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ABSTRACT
A detailed kinetic mechanism based on elementary steps is used to model the oxidation of low concentration of dibenzofuran in a post-combustion area of municipal waste incinerator. In a first approach, some simplifications are used, polychlorodibenzofuranes are represented by the dibenzofuran molecule and the post-combustion area is modeled by a continuous stirred reactor. The mechanism consists in 213 species involved in 1200 elementary steps and describes the dibenzofuran oxidation. This model has been validated on experimental data of dibenzofuran oxidation realized in a perfectly stirred reactor.

The simulations performed show that the reactivity of dibenzofuran strongly decreases when this species is very diluted. The oxidation reaction starts when the radical concentration increases with the temperature or with the reactant concentration through the initiation rate and branching steps. An addition of methane or VOCs, such as toluene, induces an important increasing of radical concentration and promotes the dibenzofuran oxidation.

This oversimplified approach brings a new contribution to understand the complex reaction of polychlorodibenzofurans oxidation.

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1. Introduction
Polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are persistent organic pollutants. Because of their lipophilic properties they can be bio-accumulated by humans but also by fauna and flora. PCDD/Fs are well known for their toxicity, mutagen, teratogen and potential carcinogen properties. Dioxins are produced unwittingly in trace amounts in thermal process, such as municipal waste incineration, medical waste incineration, cement and metal industries but also in what is commonly called “backyard incineration” of domestic wastes.

Most of studies proposed on dioxins concern their formation [1,2], they are based on quantum chemical calculations (ab initio or DFT calculations) [3], or on experimental data [4–7]. Few theoretical studies have also been proposed on oxidation of non-chlorinated dioxins [8–13]; they discuss mainly the first steps describing the oxidation of dibenzofuran.

Since dioxins are produced invariably when a chlorine source is present in thermal process, another approach, proposed in this paper, consists in studying their destruction and that of the intermediate products generated, by total oxidation in order to avoid the reformation of dioxins by catalytic or de Novo ways. Indeed, the optimization of the combustion in thermal process such as municipal waste incinerator (MWI) might reduce the level of emissions of PCDD/Fs but also of other incomplete combustion products harmful for the environment.

In this work, the influence of parameters of post-combustion (temperature, co-reactant) of MWI on the abatement of small amount (0.1–500 ppm) of dibenzofuran (DBF) is studied by...