

Dioxin and Fly Ash Free Incineration by Ash Pelletization and Reburning

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Dioxins (DXNs) in municipal waste incinerator fly ash were effectively reduced by pelletizing the mixture of ash, cement, and sodium phosphate and reburning the pellets in a laboratory scale bubbling fluidized bed (BFB) furnace. Three types of pellets—A, B and C, of various sizes and compositions were used in the experiments. The efficiency of DXN reduction in the pellet matrix was proportional to the incineration time, temperature, and degree of pellet incineration. At 700 °C and incineration time sufficient for a complete burnout, the efficiency of DXN reduction in the pellets of type A and C was found to be 99.9% and 99.7%, respectively. Correspondingly, the DXN concentration in the pellets decreased from 862 ng TEQ/kg to 0.9 ng TEQ/kg for pellets A and 2.2 ng TEQ/kg for pellets C. The residual concentration of coplanar polychlorinated biphenyls (coplanar PCBs) was below 0.2 ng TEQ/kg and 0.4 ng TEQ/kg, respectively. Assuming a tortuosity factor of $\tau = 3$ and the reaction rate constants of 0.013 m/s (at 700 °C) and 0.025 m/s (at 800 °C), the experimental pellet incineration times were reasonably predicted by using the shrinking core model. Possible DXN evaporation from the pellets was also studied. The amount of DXNs in the flue gas captured by an impinger trap was less than 3% when the reactor was operated at 700 and 800 °C. The described method of fly ash pelletization and reburning seems to be a relatively easy and inexpensive way to reduce both the emission of DXNs and the amount of fly ash.

Introduction

Among many waste treatment methods incineration seems to be a relatively inexpensive and reliable technology effective for further energy recovery, requiring no large space for waste storage and having no risk of groundwater pollution and CH₄ emission, as in the case of landfills. Incineration is also a more flexible technology compared to composting, which requires strict waste separation and may often be associated with limited demand from agriculture. In Japan, over 70% of waste mass is incinerated nowadays, because this technology offers the capability to reduce the large mass of waste produced from densely populated areas, and because there is a limited possibility of landfill due to costs (\$70–\$300 per ton) and space shortage (the remaining life of final disposal

sites in Japan is approximately 11.2 years for municipal solid waste and 3.1 years for industrial waste (1)).

However, flue gas and ashes from incinerators tend to contain unburnt carbon, heavy metals, and organic toxics, including DXNs. The DXNs can be created in the gas phase by reactions between aromatic rings containing chlorine (such as chlorobenzenes and chlorophenols) or by heterogeneous reactions between chlorinated organic precursors and fly ash based catalysts (2, 3) as well as by *de novo* synthesis (i.e. complex reactions between unburnt carbon and chlorine sources with metallic catalysts, such as Cu, Fe, Mn, etc. (4–14)).

In Japan, after a few serious DXN incidents in incineration facilities, with even a plant shut down (15), the government published a new anti-DXN act, effective since April 2000, where the emissions from all existing plants were to be regulated to meet the new standards by the end of the year 2001, i.e. 10 pg TEQ/dm³ for wastewater, 10 ng TEQ/Nm³ for flue gas from small-scale units (<2 t/h), 1 ng TEQ/Nm³ for flue gas from large plants (>4 t/h), and 3 ng TEQ/g for fly ash. The emission standards for newly built units are defined as 10 pg TEQ/dm³, 5 ng TEQ/Nm³, 0.1 ng TEQ/Nm³, and 3 ng TEQ/g, respectively. According to the anti-DXN act, the above values of TEQ include also the contribution from coplanar PCBs. However, in the present paper, the term “DXNs” refers only to polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs); coplanar PCBs are treated separately.

The objective of the present paper is to provide scientific support to an economic reduction of DXNs in the fly ash. By pelletizing the ash and then conducting its thermal treatment in a BFB reactor we may successfully reduce the emission of DXNs. As shown in Table 1, the feasibility studies conducted by Hirayama et al. (16) as well as by Kurita Ltd. indicate that ash reburning can be done in a less expensive manner than by melting in the electric arc furnace or by the Hagenmaier process.

For the pelletization process, a cement binder and a special sorbent are proposed to make the matrix into a glassy phase, so that emissions of lead and other heavy metals such as cadmium, copper, and zinc as well as chlorine by leaching can be sufficiently prevented (17, 18). Various candidate materials were tested in order to obtain maximum efficiency of lead retention, and as a result Na₃PO₄ was chosen for the present experiments (18).

Experimental Section

The municipal waste incinerator fly ash used in this study was sampled during 4 days of incinerator operation. The filter vessel was discharged twice a day, and about 12.5 kg of fly ash was obtained after each run. From each 12.5 kg of ash, 1 kg was taken out according to JIS M 8811 and put into a sampling bag. As a result, 8 kg of fly ash was obtained. The sampling bag was then shaken manually for about 1 min to obtain a homogeneous sample for analysis. As shown in Table 2, the fly ash consisted mostly of silica, calcium, aluminum, and iron oxides; however, large amounts of chlorine and MgO were also found. The ash was mixed with cement and sorbent and formed into three types of pellets (A, B, and C), the composition and physical properties of which are shown in Table 3. Pellets of types A and B were prepared with alumina (type A), or portland cement (type B), to test the effect of cement type on attrition resistance during incineration in the bed. To make pellets A or B, 50 g of fly ash as well as the required amount of cement and sorbent was put into a 500 mL plastic beaker and stirred for 10 min. Afterward, 20 g of

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TABLE 1. Comparison of Relative Costs of Three Ash Treatment Methods

system	fly ash melting (1 θ)	hagenmaier (1 θ)	reburning ^a
components	melting furnace flue gas treatment unit fly ash treatment	heating unit fly ash treatment	fly ash treatment
construction costs ^b	1	1/6	1/10
electricity charges ^b	1	1/3	unknown
unit area ^b	1	1/4	1/5
running costs ^b	1	2/3	1/3
DXN reduction	99.97%	99–99.7%	
maintenance freq	3 times/year	1 time/year	1 time/year

^a Feasibility study done by Kurita Ltd. ^b Values relative to those for fly ash melting technology.

TABLE 2. Composition of Fly Ash

element	wt %	element	wt %
SiO ₂	32.3	MgO	2.7
CaO	24.8	K ₂ O	1.7
Al ₂ O ₃	16.7	SO ₃	0.8
CO ₂	6.7	CuO	0.5
Cl	5.8	ZnO	0.5
Fe ₂ O ₃	4.2	MnO	<0.5
lg-loss (600 °C)	3.4	Pb	1.150 mg/kg

TABLE 3. Composition and Physical Properties of Pellets

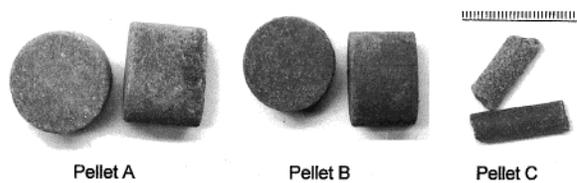
pellet type	A	B	C
diameter, mm	26	26	8
length, mm	20	20	20
ash, wt %	82	82	82
Portland cement, wt %	0	10	0
alumina cement, wt %	10	0	10
Na ₃ PO ₄ , wt %	8	8	8
DXN concentration, ng/kg		119 800	
TEQ, ng/kg	862	862	862
compressive strength, kgf/cm ²	55	36.2	
porosity ^a ϵ_v , –	0.36	0.39	0.35
density ^a ρ_{pel} , kg/m ³	1740	1860	1680

^a Determined by mercury porosimetry.

that mixture was put into a mold (diameter 26 mm, height 50 mm) to form a pellet by using a hydraulic hand press (Shimadzu: SSP-10A, compression pressure 1000 kgf/cm²). Pellets of type C were prepared with alumina cement but had different geometry, to study the effect of pellet size on DXN reduction. To make pellets C, 2 kg of fly ash, the required amount of cement and sorbent were put into a mortar mixer and stirred for 10 min. The mixture was then put into a disk pelletizer having 8 mm forming holes (Fuji Paudal F-5-S/11-175).

Some examples of raw pellets A, B, and C after formation are shown in Figure 1. Since the aspect ratio of pellets A and B was 1.3, they should be treated as a sphere of equivalent diameter of 22.6 mm. However, because the aspect ratio of pellets C was 2.5, they should be approximated by an infinitely long rod in the following analysis.

All the incineration tests were performed in a BFB reactor, a detailed schematic of which is presented in Figure 2. A ceramic tube (42 mm i.d.), adopted as a fluidized bed column, was placed in an electric furnace. A fixed bed of 2 mm o.d. alumina balls was used as a gas distributor (total bed height:



A scale [mm] is placed above pellet C

FIGURE 1. Raw pellets before incineration.

0.38 m). Silica sand of median diameter 385 μ m was used as a bed material. The bed pressure drop was measured by a differential pressure sensor. Fluidizing gas was supplied by a compressor via a computer-assisted mass flow controlling system, to keep the excess superficial gas velocity, $u_0 - u_{mf}$, constant regardless of bed temperature, so that the bubbling condition could be kept similar (u_0 is the superficial gas velocity, and u_{mf} is the minimum gas velocity needed to fluidize the bed, i.e. the gas velocity when the drag force of upward flowing gas equals the weight of particles in the bed). The excess superficial gas velocity in the bed, $u_0 - u_{mf}$, was kept at 0.1 m/s for all the experiments. To keep the $u_0 - u_{mf}$ constant, the reactor temperature, measured by a 2 mm o.d. K-type thermocouple immersed in the bed, was read by a computer to calculate gas viscosity and density and then the u_{mf} from Wen-Yu correlation (19). The value of the u_0 was obtained by adding 0.1 m/s to the u_{mf} . The signal was then sent to the mass flow controlling system.

Each test was a batch experiment. Before the test, 0.1 kg of silica sand was put into the reactor at ambient temperature. The bed temperature was then increased with a heating rate of 10 K/min. When the temperature in the reactor reached a predetermined level, a certain number of pellets of known mass were put into a basket (made of steel wire) and fed into the bed. The basket had large open holes, so that the bed material could flow in and out. After a certain predetermined incineration time, the pellets and basket were taken out of the bed and cooled. Afterward, the pellets were weighed and half-cut to study their burnout rate and then sent out for DXN analysis to Oekometric GmbH in Bayeruth, Germany (the samples were extracted in Soxhlet according to JIS K 0312, and the concentrations of DXNs were determined by a GC/MS: gas chromatograph column J&W DB-Dioxin, mass spectrometer: Finnigan MAT 95 VG Autospec).

To examine possible DXN evaporation from the pellets, the exhaust gases were introduced to a DXN trap (shown in Figure 2) immediately after feeding the pellets into a hot BFB (no basket was used in the evaporation tests). The trap consisted of four empty impingers (1000 mL each), followed by four 500 mL impingers, each one filled with 250 mL of dichloromethane. The impingers were connected into two trains (four impingers each) and put into a dry ice bath. The outlet of the DXN trap was connected to an activated carbon filter. After 1800 s of incineration (i.e. the time sufficient for a complete burnout of the pellets), the trap was disconnected, internal surfaces of all tubes and impingers were cleaned with dichloromethane, and all the liquids were condensed to prepare a sample sent for GC/MS analysis.

Theoretical Analysis of Pellet Burnout Time

Pellet incineration can be treated as a process of combustion at the surface of the unreacted core with diffusion of reactants through the porous solid product layer and mass transfer through the boundary gas layer surrounding the pellet. The process can be described by using the classical shrinking core model (20). The surface reaction rate of incineration, R^* [kmol/s], can be expressed by

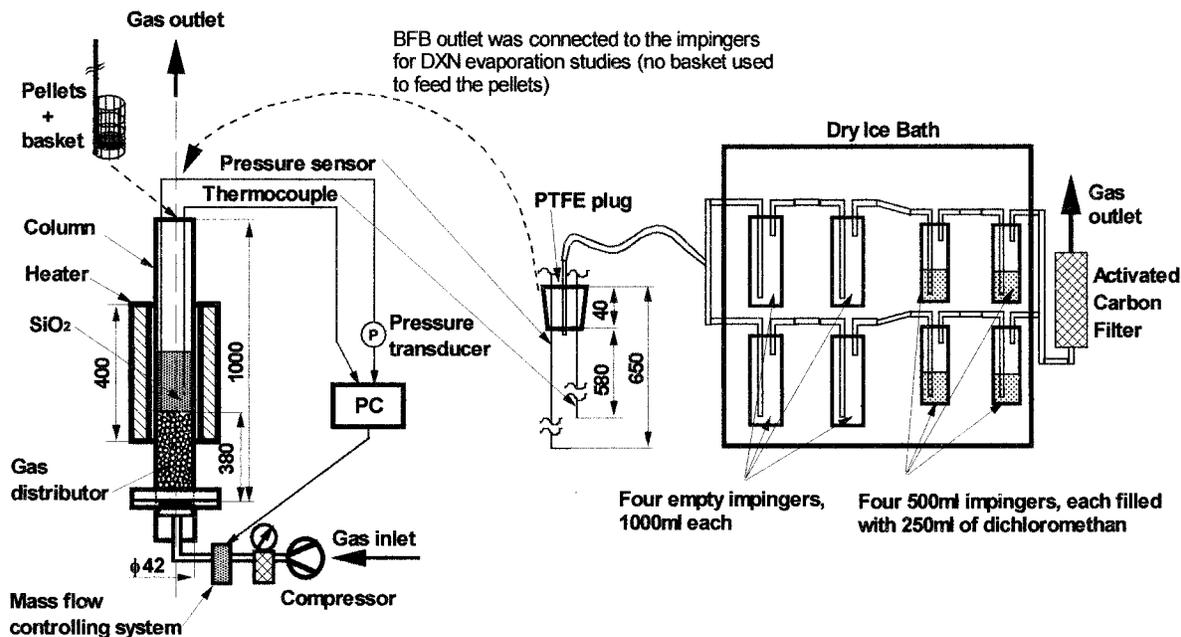


FIGURE 2. Schematic of the experimental setup.

$$R^* = A_s k_s X_{C,v} C_s \quad (1)$$

where A_s [m^2] is the surface area of the shrinking unreacted core containing carbonaceous material, C_s [$\text{kmolO}_2/\text{m}^3$] is the oxygen concentration in the pores at the core surface, k_s [m/s] is the surface reaction rate constant for a pure carbonaceous pellet, $X_{C,v}$ [m^3 carbonaceous material/ m^3 pellet] is the volume fraction of the carbonaceous material in the pellet, and, accordingly, $k_s X_{C,v}$ is the apparent reaction rate constant.

By expressing the surface reaction rate of incineration as in eq 1, we obtain the following expression for the incineration time, t_c (i.e. the time for a complete burnout of a pellet) as a function of pellet diameter, d_{pel} [m], pellet density, ρ_{pel} [kg/m^3], oxygen concentration at a place sufficiently apart from the pellet, C_∞ [$\text{kmolO}_2/\text{m}^3$], and initial mass fraction of carbonaceous materials in the pellet, X_C [kg/kg]:

$$t_c = \frac{\rho_{pel} X_C d_{pel}}{2 M_C C_\infty} \left(\frac{1}{k_s X_{C,v}} + \frac{d_{pel}}{\alpha D_e} + \frac{1}{\beta k_f} \right) \quad (2)$$

In eq 2, M_C is the molecular weight of carbon [kgC/kmol], D_e is the effective intraparticle mutual diffusivity [m^2/s], k_f is the mass transfer coefficient to the pellet in the bed [m/s], and α and β are coefficients ($\alpha = 8$ and $\beta = 2$ for a rod and $\alpha = 12$ and $\beta = 3$ for a sphere).

The effective diffusivity in the pellet, D_e , is calculated from the following equation of Satterfield and Sherwood (21)

$$D_e = D_{O_2/N_2} \frac{\epsilon}{\tau} \quad (3)$$

where D_{O_2/N_2} [m^2/s] is the mutual diffusivity of oxygen in oxygen–nitrogen mixture, ϵ [–] is pellet porosity, and τ [–] is the tortuosity factor of the pores.

D_{O_2/N_2} was calculated based on the equation of Fuller-Schettler-Giddings (22). To calculate the mass transfer coefficient, k_f [m/s], the following Avedesian and Davidson's (23) expression was adopted

$$k_f = \frac{2 D_{O_2/N_2} \epsilon_{mf}}{d_{pel}} \quad (4)$$

where ϵ_{mf} [–] is the voidage of the emulsion phase in the bed, assumed to be 0.5 in the present study.

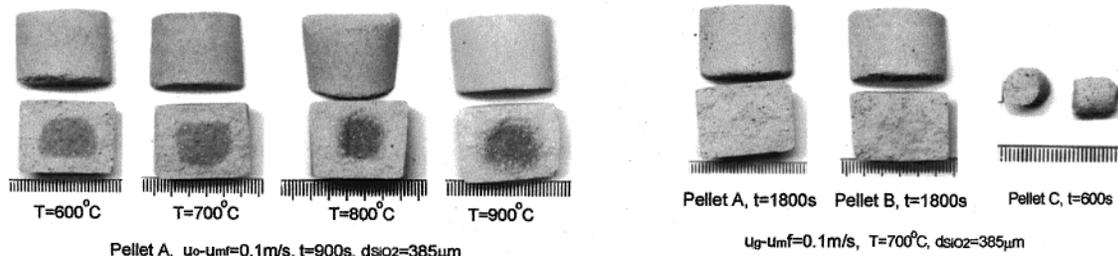
Results and Discussion

Figure 3a,b shows some photos of pellet cross-sections after incineration. A dark unreacted core, observed inside each sample in Figure 3a, indicated that the pellet incineration time was not sufficient for a complete burnout. The pellets with no dark core (shown in Figure 3b), corresponded to those incinerated long enough to be completely reburned. The size of the unreacted core was determined from a black-and-white digital image of pellet cross-section by using graphic software for image analysis, *WinROOF 3.5*. Each pixel in the picture was classified into one of 256 gray scale classes, and as a result a bimodal PDF histogram was obtained (PDF = probability density function). The minimum value of PDF between these two maximums was assumed to correspond to the boundary area between the incinerated zone and the unreacted core. By converting the picture and setting up all the gray scale classes below this minimum as white, and these above it as black, a clear separation surface between the unreacted core and the incinerated area was obtained. Length and width of the core was measured three times with an accuracy of 0.5 mm, and the average value was obtained. The approximately rectangular cross-sections of length a and width b of pellets A or B were expressed by the equivalent diameter d , which was calculated by the following formula:

$$d = \frac{2ab}{a+b} \quad (5)$$

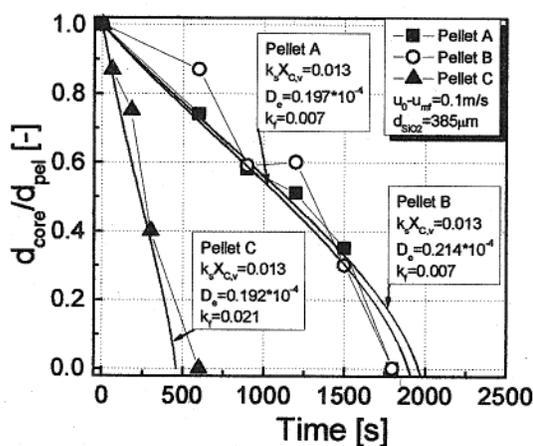
In Figure 3c,d the incineration rate of pellets (determined from the size of unreacted core) is presented as a function of time for $T = 700^\circ$ and 800°C . The incineration rate increased with temperature. For pellets A and B the unreacted core disappeared after 1800 s of incineration at 700°C and after 1500 s at 800°C . For pellets of type C these values were 600 and 300 s, respectively.

To determine the reaction rate constants, the experimental data of Figure 3c,d were compared with numerical calculations, substituting pellet porosity and density from Table 3. For each pellet the values of D_e and k_f were calculated from eqs 3 and 4, assuming the tortuosity factor $\tau =$

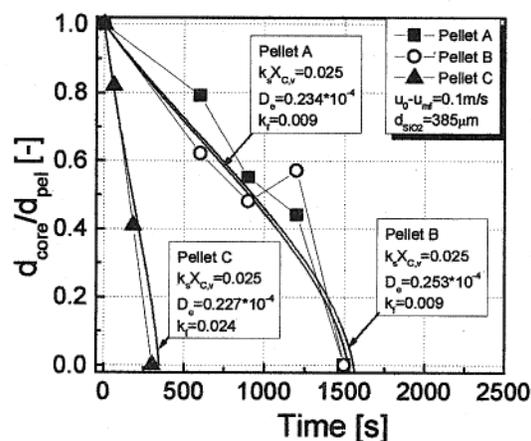


a) Incomplete incineration

(b) Complete incineration



c) T=700°C



d) T=800°C

FIGURE 3. Cross-sections of pellets (a and b) and pellet incineration rates versus time (c and d).

TABLE 4. Comparison of Calculated and Experimental Incineration Times^a

pellet type	temp [°C]	contribution of each rate step to the overall incineration time						time required for complete incineration	
		A: chemical reaction $\gamma/k_sX_{C,v}$		B: ash diffusion $\gamma d_p/\alpha D_e$		C: gas diffusion $\gamma/\beta k_f$		t_{calc} A+B+C [s]	t_{exp} [s]
A	700	669	35	830	44	397	21	1896	1500–1800
B	700	715	36	819	42	425	22	1959	1500–1800
C	700	230	50	156	34	73	16	459	300–600
A	800	384	25	773	50	373	25	1530	1200–1500
B	800	408	26	758	48	395	26	1561	1200–1500
C	800	132	38	145	42	68	20	345	180–300

^a $1/k_sX_{C,v}$: fitted for experimental data, $\gamma = \rho_{pel}X_{C,d_{pel}}/(2M_{C,w})$.

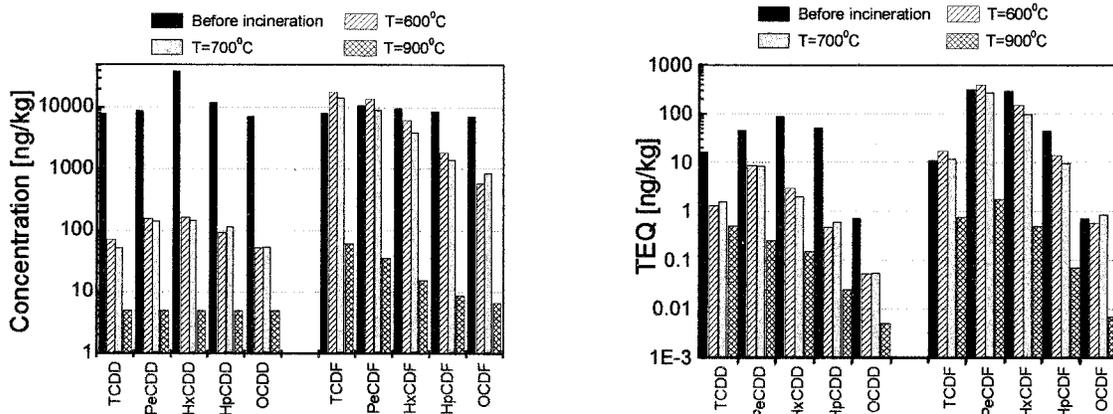
3 in eq 3. $k_sX_{C,v}$ was determined by fitting the model prediction with experimental data. For all three types of pellets, the calculations with the apparent reaction rate constant $k_sX_{C,v} = 0.013$ at 700 °C and 0.025 m/s at 800 °C agreed quite well with the experimentally determined core shrinking process.

To show the contribution of chemical reaction, ash diffusion and gas diffusion to the total pellet incineration time, the values of t_c , calculated from eq 2 with the experimentally fitted reaction rate constants, are presented in Table 4. For pellets A and B, regardless of the temperature, the main component, responsible for roughly 40–50% of the total incineration time, is the diffusion through the porous product layer surrounding the unreacted core. Contribution of chemical reaction is about 25–35% and external gas diffusion about 20–25%. For rodlike pellets C, the contribution of diffusion through the pellet was roughly 40%, the

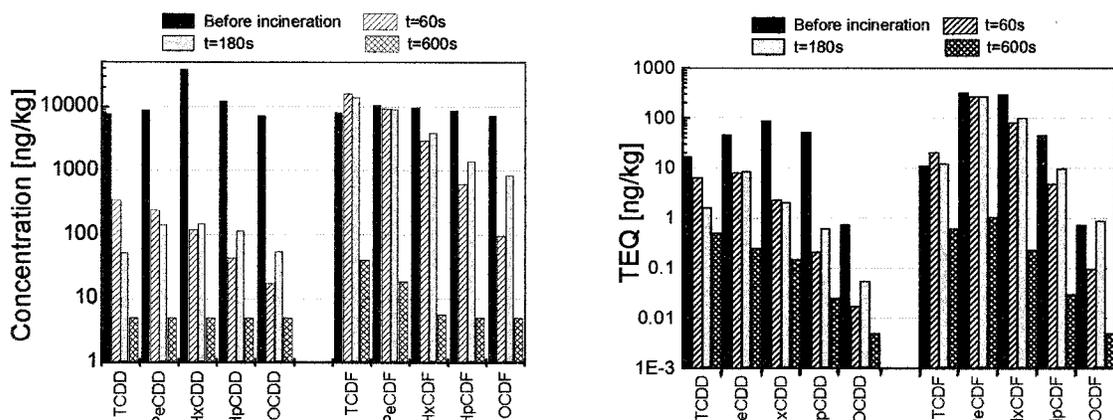
contribution of chemical reaction roughly 35–40%, and that of gas diffusion only about 15–25% of the total incineration time at both 700° and 800 °C.

Based on preliminary experiments on pellet attrition in the bed, the studies on DXN reduction were only carried out for pellets A and C containing alumina cement and having higher attrition resistance than pellets B, made with portland cement.

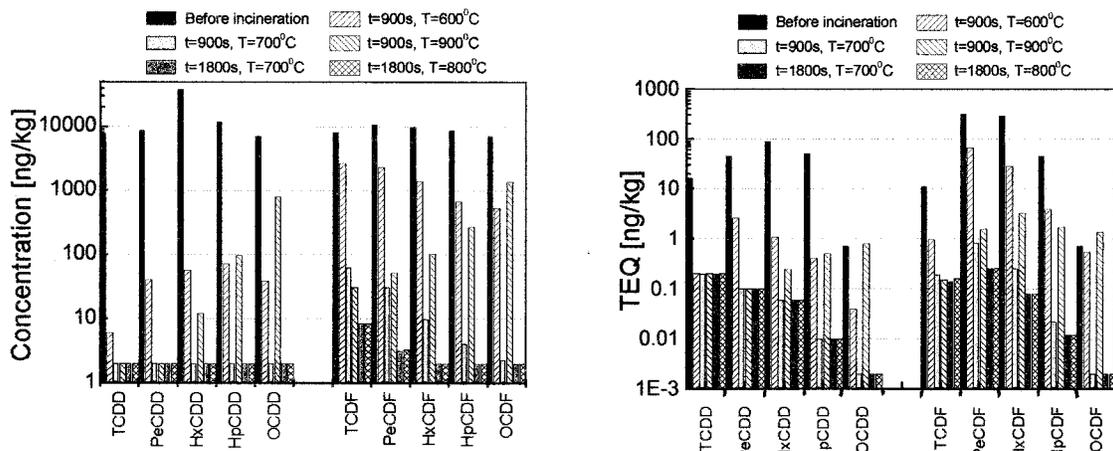
The effect of temperature and incineration time on DXN homologue pattern is shown in Figure 4. Higher incineration temperature of pellets C (Figure 4a) brought about higher DXN decomposition rate. After 180 s of incineration at 900 °C, the total concentration of each DXN homologue decreased from about 10 000 ng/kg to well below 100 ng/kg. As for the toxicity equivalent, TEQ, the total DXN concentration was 583.1 ng/kg, 405.6 ng/kg, and 4 ng/kg, for the temperatures 600, 700, and 900 °C, respectively. However, high reaction temperature is not recommended for incineration, because the efficiency of heavy metals and chlorine capture by Na_3PO_4 will decrease, and agglomeration of the bed may take place. As can be seen in Figure 4b the lowest DXN concentration in pellets C was obtained after complete burnout (i.e. after 600 s of incineration at 700 °C). The concentration of each homologue fell well below 10 ng/kg for PCDD and below 70 ng/kg for PCDF. The residual concentration of DXNs in the sample was 104.2 ng/kg or 2.2 ngTEQ/kg. The effect of incineration time and temperature on the efficiency of DXN reduction in pellets A is shown in Figure 4c. Similar to the results obtained for pellets C, a longer residence time brought about higher reduction efficiency. At 700 °C and complete incineration of pellets A (i.e. after 1800 s), the concentration of each homologue was below 10 ng/kg, what corresponded to the total TEQ of 0.9 ng/kg. Additionally, for reaction time sufficiently long for a complete



a) Effect of temperature – pellets C, $t=180s$



b) Effect of incineration time – pellets C, $T=700^{\circ}C$



c) Effect of temperature and incineration time – pellets A

FIGURE 4. Effect of temperature and incineration time on DXN homologue pattern (all tests were done with a bed of silica sand of $d_{S102} = 385 \mu m$ and at $u_0 - u_{mf} = 0.1 m/s$).

incineration, there was almost no change of DXN concentration, regardless if the bed was operated at 700 or 800 °C.

Figure 5 shows the effect of temperature and incineration time on the reduction of DXNs (parts a and b) and coplanar PCBs (parts c and d). As shown in Figure 5a, at 600 °C but with the residence time shorter than required for a complete burnout (i.e. 900 s for pellets A and 180 s for pellets C), the

total TEQ of DXNs decreased from 862 ng/kg to 105.2 ng/kg (pellets A) and 583.1 ng/kg (pellets C). At 700 °C, the residual TEQ was 1.7 ng/kg and 405.6 ng/kg, respectively. However, the incineration of pellets A for 1800 s brought about the decrease of TEQ below 1 ng/kg at both 700 and 800 °C. As can be seen in Figure 5b, the lowest concentration of DXNs was obtained at incineration time sufficient for a complete

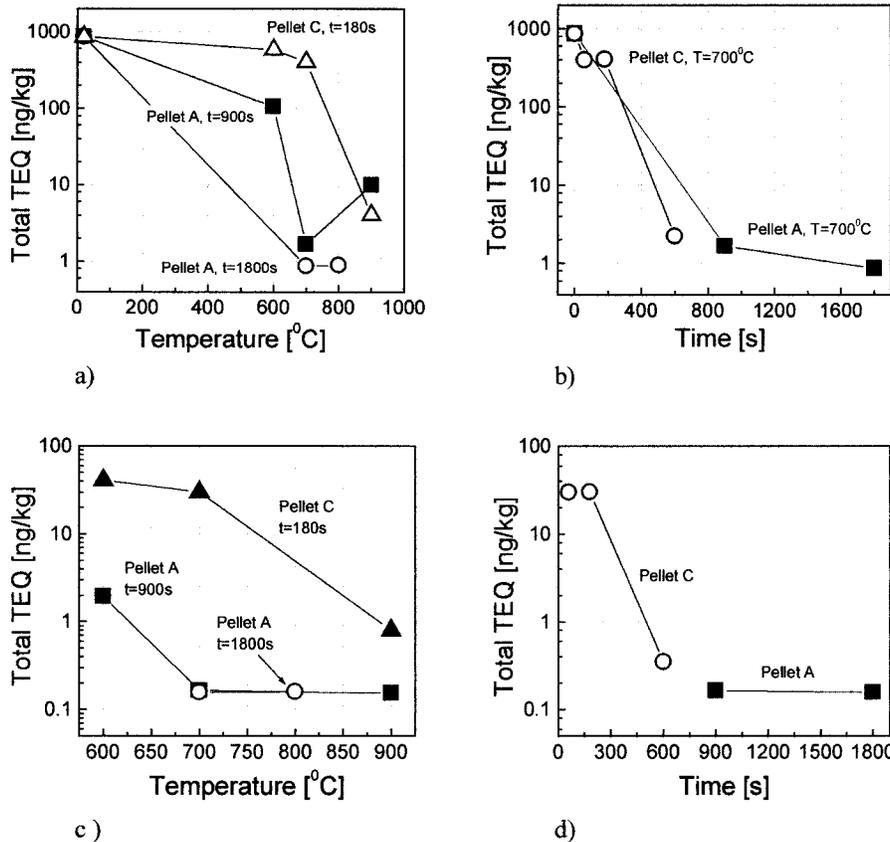


FIGURE 5. Residual concentrations of DXNs (a and b) and coplanar PCBs (c and d) versus incineration temperature and time ($d_{SiO_2} = 385 \mu m$, $u_0 - u_{mf} = 0.1 \text{ m/s}$).

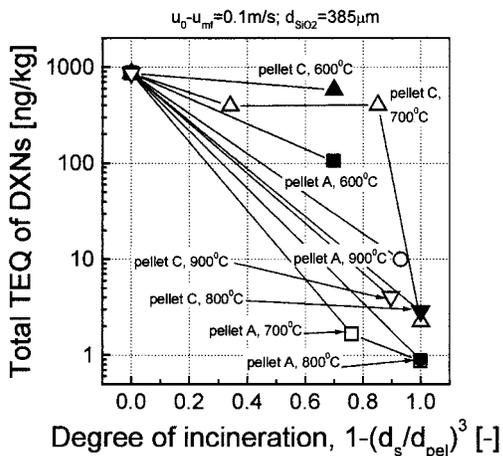


FIGURE 6. Concentration of DXNs versus degree of pellet incineration at various temperatures ($d_{SiO_2} = 385 \mu m$, $u_0 - u_{mf} = 0.1 \text{ m/s}$).

burnout (i.e. 1800 s for pellets A and 600 s for pellets C). In such case the TEQ of pellets decreased to 0.9 ng/kg and 2.2 ng/kg. This corresponded to the efficiency of DXN reduction in the pellet of 99.9% and 99.7%, respectively.

As shown in Figure 5c, the incineration of pellets C at 600, 700, and 900 °C for 180 s brought about the decrease of TEQ of coplanar PCBs to about 40 ng/kg, 30 ng/kg, and below 1 ng/kg, respectively. The TEQ of coplanar PCBs was calculated according to WHO/IPCS 1997. For pellets A, the concentration of coplanar PCBs was 2 ng TEQ/kg after 900 s of incineration at 600 °C, but it fell below 0.2 ng TEQ/kg if the pellets were incinerated at 700 or 900 °C. Similar concentrations of coplanar PCBs were measured after 1800 s of incineration at 700 and 800 °C. As shown in Figure 5d, the lowest

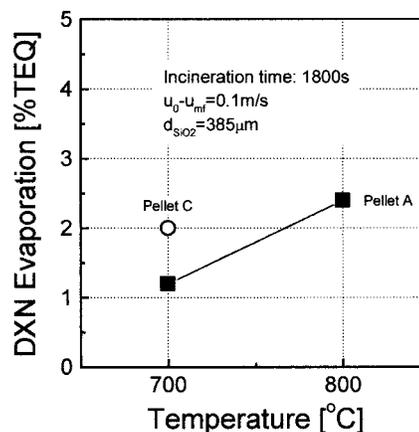


FIGURE 7. DXN evaporation versus temperature.

concentrations of coplanar PCBs (i.e. below 0.4 ng TEQ/kg for pellets C and below 0.2 ng TEQ/kg for pellets A) were obtained for completely incinerated pellets. This is similar to the results obtained for DXNs (cf. Figure 5b).

As shown in Figure 6 the efficiency of DXN reduction was proportional to the degree of incineration, reaching maximum for a completely incinerated pellet, regardless of pellet type and bed temperature.

Figure 7 shows the results of DXN evaporation studies. After 1800 s of incineration of both pellets A and C, the amount of evaporated DXNs did not exceed 3%, regardless of pellet type and incineration temperature. In an industrial system most of these evaporated and nondecomposed DXNs should probably be adsorbed by fly ash particles, which are then captured by the bag filter and again pelletized and returned in the bed.

The incinerated pellets can be disposed to landfill as an ordinary municipal waste incineration bottom ash.

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