Thermochimica Acta, 85 (1985) 419–433
Elsevier Science Publishers B.V., Amsterdam — Printed in The Netherlands

CONTAINMENT OF DIOXIN EMISSIONS FROM REFUSE FIRED THERMAL PROCESSING UNITS - PROSPECTS AND TECHNICAL ISSUES

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ABSTRACT

There are reports (ref.1-5) that dioxins ("dioxins" means any of the chlorinated dibenzo-p-dioxins) and related compounds, e.g., furans ("furans" means any of the chlorinated dibenzofurans) are emitted from some refuse-fired thermal processing units ("thermal processing units" means incinerators and incinerators equipped for energy recovery, i.e., boilers). The extent to which such emissions occur and the conditions that lead to such emissions have not been clearly defined at the present time. If it is determined that dioxin or furan emissions from refuse-fire thermal processing units should be contained or minimized, then it will be necessary to examine strategies for achieving goals of containment or minimization of these emissions. This report addresses the prospects for and technical issues concerned with the utilization of chemisorption as a technique for containment of dioxin or furan emissions from refuse-fired thermal processing units.

BACKGROUND

There is evidence (ref.6) that desorption energies of chemisorbed dioxins on fly ash may be as large as 40-50+ kcal/mol (ref.6). Furans, being in some respects chemically similar to dioxins (see Fig. 1) may have comparable desorption energies. If desorption energies are substantial for these compounds, then their successful chemisorption on some inexpensive adsorbent, e.g., fly ash, may provide a basis for containment of these materials. If fly ash containing chemisorbed dioxins or furans is disposed of into the environment at an ambient temperature of 25°C and if the rate constant, k_d, for thermal desorption of dioxins or furans is estimated to be (ref.6)

$$k_D < 10^{13} e^{-40000/(RT)} \sec^{-1} \qquad (1)$$

then the time required for one percent of the dioxins or furans to leach from the fly ash due to thermal desorption is about 6.9

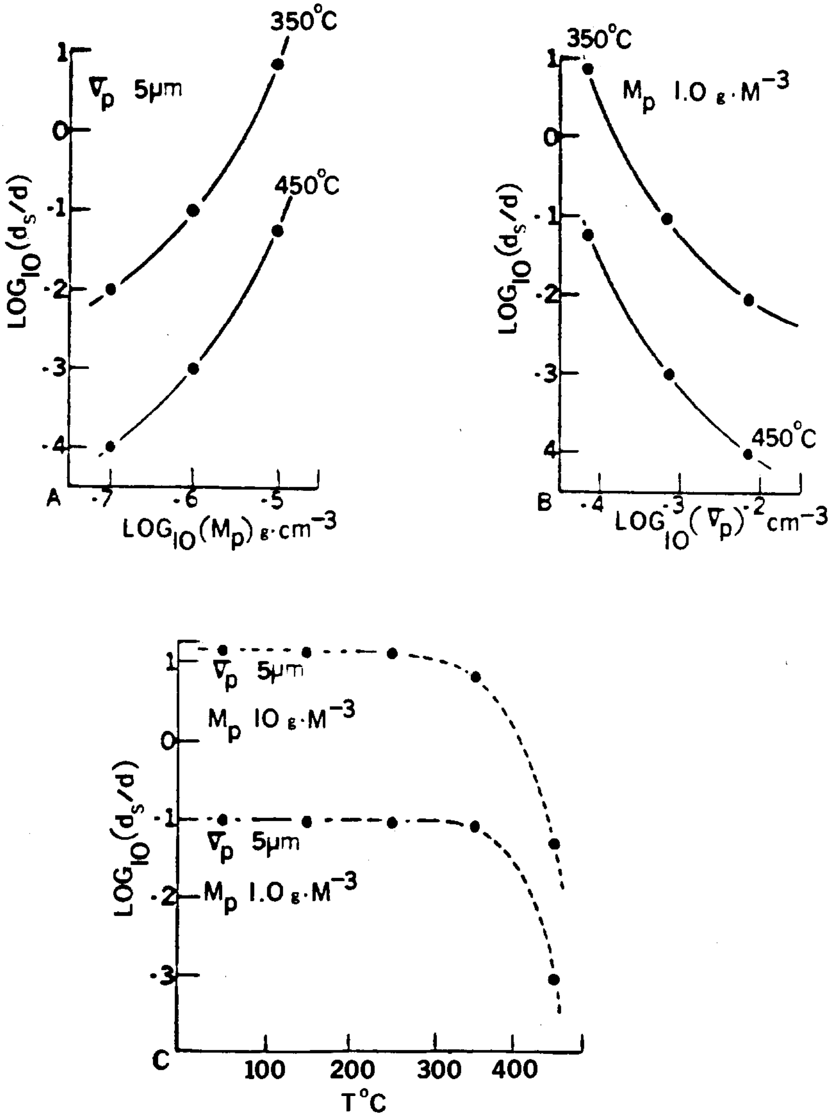

Fig. 1. Parametric dependence of dioxin chemisorption on input parameters.

million years. Thus, unless chemisorbed dioxins can be leached in the environment by other than ambient thermal stresses, they may remain chemically bound for extremely long periods of time. If this is validated experimentally, then chemisorption may offer a means for safe containment of dioxins or furans.

This report provides a feasibility analysis for dioxin or furan containment by chemisorption. Chemisorption of dioxins on fly ash is modeled under conditions which may be achieved in thermal processing units. The possibilities for utilizing other adsorbents are also discussed. It has been suggested that chemisorption of furans may be considered similar to dioxins. Therefore, an analysis of furan chemisorption is not presented (ref.10).

Tests of concepts outlined in this report are recommended. Suggested research, development and testing may optimize understanding of chemisorption of dioxins and furans as a containment technology.

References are provided to suggest a basis for the chemisorption model that will be developed and for understanding physical-chemical aspects of dioxin emissions from thermal processing units.

Numerous assumptions and approximations are utilized. These are described in this report and in (ref.10). Since the intent of this report is only to suggest concept feasibility, the assumptions and approximations made should be acceptable. Substantial refinements are probably not warranted without further research, development and testing.

MODEL DEVELOPMENT

A model is constructed to describe chemisorption in thermal processing units. Assume that chemisorption of competitively adsorbed species is described by a Langmuir isotherm (ref.7). Assume that chemisorption of dioxins and other species onto entrained fly ash in a hot flowing gas stream may be modeled by treating all species other than dioxins as having equivalent chemisorption properties. This assumption is made to simplify calculations and demonstrate the effect of a difference in the adsorption energy, E_A, between dioxins and other species, upon the chemisorption properties of the gas/fly ash mixture. These assumptions permit the reader to use the model to test input parameter variations with a pocket calculator.

Review the material presented in Table 1 prior to reading the following discussion. Table 1 provides the terminology, assumptions and parameter assignments which are incorporated into the discussion and model development that follows. The basis for the parameters and assigned values reported in Table 1 is discussed in (ref.8-10).

Dioxins are denoted by, d. All other competitively chemisorbed species are denoted by, a. If dioxins and other species are in the gas phase, their respective symbols are not subscripted. If dioxins and other species are chemisorbed, they are subscripted as d_s and a_s. Adsorption sites for chemisorption are denoted by, S. For simplicity all adsorption sites are considered to be identical.

Competitive chemisorption can be described by the following representation:

$$d + S \underset{k_{-1}}{\overset{k_1}{\rightleftarrows}} d_s \tag{2}$$

$$a + S \underset{k_{-2}}{\overset{k_2}{\rightleftarrows}} a_s \tag{3}$$

Time dependent rates of chemisorption of d and a can be respectively represented by the equations (ref.7-10):

$$\dot{d}_s \equiv \frac{d}{dt}(d_s) = k_1\, d\, (1 - \theta_a - \theta_d) - k_{-1}\, d_s \tag{4}$$

$$\dot{a}_s \equiv \frac{d}{dt}(a_s) = k_2\, a\, (1 - \theta_a - \theta_d) - k_{-2}\, a_s \tag{5}$$

The following definitions and assumptions are made (ref.9,10):

$$d = d_o - d_s \tag{6}$$

$$a = a_o - a_s \tag{7}$$

$$\theta_a \equiv \frac{1}{4N_p}\left(\frac{\sigma_a}{\sigma_p}\right)^2 a_s \tag{8}$$

$$\theta_d \equiv \frac{1}{4N_p}\left(\frac{\sigma_d}{\sigma_p}\right)^2 d_s \tag{9}$$

Table 1. Summary of Assigned Input Values(a)

Variable:	Units:	Values assigned:	Comments:
T	°C	50;150;250;350;450 323;423;523;623;723	The assigned values span the range over which effects on adsorption of activation energy, mass loading, adsorption time, etc., can be demonstrated.
E_1	$kcal \cdot mol^{-1}$	10	See discussion, this section.
E_{-1}	$kcal \cdot mol^{-1}$	40	See (a) and discussion, this section.
E_2	$kcal \cdot mol^{-1}$	0	See discussion, this section.
E_{-2}	$kcal \cdot mol^{-1}$	30	See discussion, this section.
σ_p	cm	$5x10^{-5}$, $5x10^{-4}$, 5×10^{-3}	Corresponds to particle diameters of 1; 10 and 100 micronmeters.
M_p	$g \cdot cm^{-3}$	10^{-7}; 10^{-6}; 10^{-5}	Corresponds to fly ash mass loadings of 0.1; 1; and 10 grams per cubic meter.
d_o	$g \cdot M^{-3}$	10^{-5}	Must be converted to units of molecules per cubic centimeter for use in equations appearing in this report.
a_o	$g \cdot M^{-3}$	$10^2 d_o$	See discussion, this section.
σ_d	cm	$\cong 4.24x10^{-8}$ cm	(a)
σ_a	cm	σ_d	Arbitrary assignment; see discussion, this section.
m_d	g	321.8	(a)
m_a	g	178.2	See discussion, this section.

(a) Definitions of additional variables and values assigned are presented in (ref.10).

$$\theta_a >> \theta_d \tag{10}$$

The subscript, o, used in equations (6) and (7) above denotes initial values of the concentrations of a and d prior to chemisorption. If the following definitions are made:

$$a_1 \equiv k_2 a_o \tag{11}$$

$$a_2 \equiv -\left\{\frac{k_2 a_o}{4N_p}\left(\frac{\sigma_a}{\sigma_p}\right)^2 + k_2 + k_{-2}\right\} \tag{12}$$

$$a_3 \equiv \frac{k_2}{4N_p}\left(\frac{\sigma_a}{\sigma_p}\right)^2 \tag{13}$$

Equations (6) through (13) can be used to write equation (5) in the form

$$\dot{a}_s \cong a_1 + a_2\, a_s + a_3\, a_s^2 \tag{14}$$

The physically meaningful solution to this equation can be shown to be (ref.11):

$$a_s = \frac{C_1}{C_o}\,\frac{\left[1 - e^{-t_a j}\right]}{\left[e^{-t_a j} - \frac{C_1}{C_2}\right]} \tag{15}$$

The symbol, t_a, denotes time for chemisorption of a. Below, j, C_o, C_1 and C_2 are defined:

$$j \equiv (a_2^2 - 4a_1 a_3)^{1/2} \tag{16}$$

$$C_o \equiv 2a_3 \tag{17}$$

$$C_1 \equiv a_2 - j \tag{18}$$

$$C_2 \equiv a_2 + j \tag{19}$$

At equilibrium ($t_a \rightarrow + \infty$), equation (15) has the solution:

$$(a_s)_{EQ} = -\frac{C_2}{C_o} \tag{20}$$

At equilibrium the forward and reverse rate components of the R.H.S. of equation (4) can be equated to each other, and it follows from equation (10) that:

$$\left(\frac{d_s}{d}\right)_{EQ} \cong \left(\frac{k_1}{k_{-1}}\right)\left[1 - (\theta_a)_{EQ}\right] \tag{21}$$

This can be written as:

$$\left(\frac{d_s}{d}\right)_{EQ} \cong \left(\frac{k_1}{k_{-1}}\right)\left[1 - \frac{1}{4N_p}\left(\frac{\sigma_a}{\sigma_p}\right)^2 (a_s)_{EQ}\right] \tag{22}$$

Rearrangement of equation (15) provides the following solution:

$$t_a = \frac{1}{j}\,\ell n\left[\frac{1 + a_s\left(\frac{C_o}{C_1}\right)}{1 + a_s\left(\frac{C_o}{C_2}\right)}\right] \tag{23}$$

The time required to achieve 99% of the equilibrium value of chemisorbed a_s is found from equations (20) and (24) shown below to be:

$$a_s = 0.99\ (a_s)_{EQ} \tag{24}$$

$$t_a(99\%) = \frac{1}{j}\,\ell n\left[10\left\{1 - 0.99\,\frac{C_2}{C_1}\right\}\right] \tag{25}$$

using the definition:

$$y \equiv \frac{1}{4N_p}\left(\frac{\sigma_a}{\sigma_p}\right)^2 \tag{26}$$

The expressions which have been developed above can be used to show that:

$$\dot{d}_s \cong k_1 d_o\ (1 - ya_s) - [k_{-1} + k_1\ (1 - ya_a)]d_s \tag{27}$$

Near equilibrium,

$$a_s \cong (a_s)_{EQ} \tag{28}$$

If the time required for a_s to reach an equilibrium value is significantly shorter than the time required for d_s to reach an equilibrium value, equation (28) above may be regarded to be exact.

Defining

$$Q \equiv k_1 d_o \; [1 - y \; (a_s)_{EQ}] \qquad (29)$$

$$P \equiv [k_{-1} + k_1 \; \{1 - y \; (a_s)_{EQ}\}] \qquad (30)$$

It can be shown that near the equilibrium value of d_s that:

$$d_s \cong \frac{Q}{P} \; (1 - e^{-Pt}) \qquad (31)$$

This equation can be rearranged to show that near the equilibrium value of d_s:

$$t \cong - \frac{1}{P} \; \ell n \; (1 - \frac{P}{Q} \; d_s) \qquad (32)$$

To determine the time required to achieve 99% of the equilibrium value of chemisorbed d_s, first calculate a value for $(a_s)_{EQ}$. Use $(a_s)_{EQ}$ and equations (6) and (22) to calculate a value for $(d_s)_{EQ}$ set:

$$d_s = 0.99 \; (d_s)_{EQ} \qquad (33)$$

Use this value of d_s to calculate t_d (99%) in equation (32).

A more approximate value for t_d (99%) may be developed using equation (41) of Appendix C of (ref.10). If the activation energy for chemisorption E_A of d_s is non-zero (ref.6,12):

$$\overset{\text{approx}}{t_d}(99\%) \cong - \frac{4N_p}{Z_p^d a_o} \left(\frac{\sigma_p}{\sigma_a}\right)^2 \ell n \left[1 - \frac{0.99 (d_s)_{EQ}}{4N_p} \left(\frac{a_o}{d_o}\right) \left(\frac{\sigma_a}{\sigma_p}\right)^2\right] e^{+E_A/(RT)} \qquad (34)$$

This equation explicitly illustrates the dependence of the time to achieve near-equilibrium chemisorption upon the activation energy for adsorption.

When performing calculations of $(a_s)_{EQ}$ and $(d_s)_{EQ}$ utilizing the above equations, it is important to determine that the results obtained are physically meaningful.

For example, since the available number of surface adsorption sites is limited, the calculated values for $(a_s)_{EQ}$ and $(d_s)_{EQ}$ should be reset to the values $(a_s)_{max}$ and $(d_s)_{max}$ (these are upper limit values for a_s and d_s) respectively, if:

$$(a_s)_{EQ} > (a_s)_{max} \qquad (35)$$

$$(d_s)_{EQ} > (d_s)_{max} \qquad (36)$$

The values for $(a_s)_{max}$ and $(d_s)_{max}$ may be calculated from the following relationships which are valid assuming equilibrium chemisorption (ref.7) and $a_o \geq (a_s)_{max}$ and $d_o \geq (d_s)_{max}$:

$$\left(\frac{\theta_d}{\theta_a}\right)_{max} \cong \left(\frac{\theta_d}{\theta_a}\right)_{EQ} = \frac{k_1 k_{-2} d_o m_a}{k_{-1} k_2 a_o m_d} \equiv h \qquad (37)$$

$$(\theta_d)_{max} + (\theta_a)_{max} = 1 \qquad (38)$$

Equations (37) and (38) yield the results:

$$(\theta_d)_{max} \cong h\ (1 + h)^{-1} \qquad (39)$$

$$(\theta_a)_{max} \cong 1 - (\theta_d)_{max} \qquad (40)$$

These equations can be used with equations (8) and (9) to show that

$$(d_s)_{max} \cong 4N_p \left(\frac{\sigma_p}{\sigma_d}\right)^2 (\theta_d)_{max} \qquad (41)$$

$$(a_s)_{max} \cong 4N_p \left(\frac{\sigma_p}{\sigma_a}\right)^2 (\theta_a)_{max} \qquad (42)$$

In equation (37) above, m_d and m_a are the respective molecular weights of d and a.

In addition, if there are more adsorption sites for a and d than initial gas phase species, i.e.,

$$(d_s)_{max} > d_o \qquad (43)$$

$$(a_s)_{max} > a_o \qquad (44)$$

then if $(a_s)_{EQ} > a_o$ and $(d_s)_{EQ} > d_o$, the calculated values for $(a_s)_{EQ}$ and $(d_s)_{EQ}$ should be reset to a_o and d_o respectively, instead of to $(a_s)_{max}$ and $(d_s)_{max}$ respectively.

The rate constants for adsorption and desorption of dioxins and other competitively adsorbed species are determined as described in (ref.10), except that in the following section the presumption is made that there is a non-zero activation energy E_1, for adsorption of dioxins on fly ash, i.e.:

$$k_1 = Z_p^d \, e^{-E_1/(RT)} \tag{45}$$

The superscript, d, denotes collisions of dioxins with particulates.

MODEL INPUT PARAMETERS

Values for input parameters to test the model are reported in Table 1 of this report or in (ref.10). The parameter values have been chosen to illustrate the dependence of the chemisorption process upon key variables. In general the assigned values for parameters are approximations (ref.10).

E_1, the activation energy for chemisorption of a dioxin molecule, has been assigned a value of 10 $\text{kcal}\cdot\text{mol}^{-1}$ based on the reported observation (ref.12) that significant chemisorption of dioxins on fly ash occurs at temperatures above about 100°C. It is reported (ref.12) that thermal desorption of dioxins is apparently irreversible even at temperatures up to about 300°C. Additionally, estimates of the activation energy for thermal desorption of dioxins from fly ash based upon models for catalytic dioxin formation have been made (ref.6). It appears that the activation energy for thermal desorption of chemisorbed dioxins may be large, perhaps in excess of 40 $\text{kcal}\cdot\text{mol}^{-1}$. The value $E_{-1} = 40\ \text{kcal}\cdot\text{mol}^{-1}$ is used in calculations presented in this report.

Other species besides dioxins are present in the hot gas streams of thermal processing units (ref.1-6). Many of these species may be capable of being chemisorbed on fly ash (ref.1-7, 11,12). In the calculations discussed in this report, it is assumed that all chemical species undergoing competitive chemisorption have the same molecular diameter. This is equivalent to stating that the total number of adsorption sites is fixed and that adsorption at any site is non-specific to species identity. This assumption also implies the mathematical simplification of grouping all competitively adsorbed species together and treating them as one species with an arbitrarily assigned molecular weight. In this report, the molecular weight for the competitively adsorbed species is assigned a value typical of a three-ringed polycyclic aromatic. Inspection of the model equations suggests that chemisorption results do not depend strongly upon molecular weight. The relative ratio of chemisorbed species to dioxins is assigned a value of one hundred. Other values may be tested by the reader.

The particulate mass loading values and particle sizes which have been assumed for the fly ash are representative of the range of values for these parameters during thermal processing. Unique rather than distributed values for these parameters, have been assumed to avoid mathematical complications. These approximations make it easier to recognize the dependence of chemisorption of dioxins upon parametric variations of these variables.

The values assigned to the activation energies for chemisorption and desorption of competitively adsorbed species correspond closely to values that have been assigned to polycyclic organic molecules (ref.11). In particular, the zero value assigned to the chemisorption activation energy provides strong contrast to the value of 10 $\text{kcal}\cdot\text{mol}^{-1}$ assigned to the chemisorption energy for dioxins. This provides a test of the dependence of competitive chemisorption of dioxins upon differences in chemisorption activation energies between dioxins and other chemisorbed species.

RESULTS OF MODEL CALCULATIONS

Calculations, based on the constructed model and the input data presented in this report and in (ref.10), have been made to test the dependence of chemisorption upon variations in several input parameters. The results of these calculations are presented in Figs. 1a through 1c and in Table 2.

In Figs. 1a through 1c, the y-axis variable is the base ten logarithm of the ratio of chemisorbed dioxins to dioxins which are present in the gas phase. The larger the numerical value of this y-axis variable, the greater the amount of dioxins which are adsorbed on the fly ash compared to the amount of dioxins which are present in the gas phase. To assure successful containment of dioxins by chemisorption on fly ash, it is necessary to determine conditions which maximize the value of this y-axis parameter. Since it is presumed that it may be desirable to collect particulates laden with chemisorbed dioxins, no modeling of chemisorption onto particles smaller in size than one micrometer was carried out. Particles smaller in size than this are more difficult to contain with air pollution control equipment than are larger particles.

All of the results presented in Figs. 1a through 1c are based on the assumption of passage of sufficient time for development of dynamic equilibrium partitioning of dioxins between the gas and entrained fly ash. Table 2 suggests approximate transit times required for development of dynamic equilibrium between gas and fly ash at typical load conditions. If the configuration of the flow in a thermal processing unit is insufficient to allow time for development of dynamic equilibrium, the reader can recalculate the partitioning using the time dependent equations which have been presented in the model development section of this report. If the existing gas flow configuration is found insufficient to achieve substantial dioxin containment by entrained fly ash chemisorption, an alternative may be to attempt reconfiguring the flow stream to allow for packed bed chemisorption. The reader is cautioned that uncertainties in approximations made in this report dictate necessary testing to select materials, configurations and operating conditions for optimal chemisorption.

Table 2. Chemisorption Times for Dioxins and Other Species[(a)]

T °C	t_a (99%) seconds	t_d (99%) seconds
50	2.04×10^{-2}	$7.06 \times 10^{+4}$
150	1.99×10^{-2}	$1.69 \times 10^{+3}$
250	1.20×10^{-2}	$1.59 \times 10^{+2}$
350	5.82×10^{-3}	$4.00 \times 10^{+1}$
450	4.77×10^{-4}	8.90×10^{-1}

(a) Pocket calculator results based on equations in the text and using the values: $\sigma_p = 5 \times 10^{-4}$ cm, $M_p = 10^{-5}$ g·cm^{-3}. These results illustrate the sensitivity of adsorption times to activation energies for chemisorption.

SUMMARY AND CONCLUSIONS

The results developed in this report suggest that containment, through chemisorption of dioxin emissions from refuse fired thermal processing units, may be technically feasible. Suggestions for research objectives and full scale tests of concepts developed in this report are outlined below:

- Based on the hypothesized model, the most critical factor affecting the required contact time for efficient adsorption of dioxins at any temperature is the energy of adsorption, E_A. The lower E_A is, the shorter the required contact time for efficient adsorption. The higher the temperature of the adsorbent, the shorter the contact time required for efficient adsorption. If the value of E_A is greater than zero, the adsorbent must be heated to effect efficient dioxin containment. Laboratory investigations should be carried out to determine E_A for candidate adsorbents, e.g., by type (and composition): carbonaceous (carbon black, graphite, charcoal), ferromagnetic, mineral (sand, fly ash, Zeolites), etc. Mixtures of adsorbents should be similarly tested. Different adsorbents can be field tested. Fly ash may be a desirable adsorbent since it is a by-product of refuse fired thermal processing operations and is therefore readily available on site.

- There may be a thermal "window" for efficient chemisorption of dioxins, which is principally defined by the energy of desorption, E_D on the high temperature side and by E_A on the low temperature side. The larger E_D is, the higher the upper limit cutoff temperature for efficient dioxin adsorption will be. The smaller E_A is, the smaller the lower limit cutoff temperature for efficient adsorption of dioxins will be.

- Since adsorption is likely to be inefficient at furnace gas exit temperatures, it will be necessary to drop the gas temperature after the gas exits from the furnace of the thermal processing unit. In the example presented in this report, the temperature should be dropped to below about 350°C. This implies that incinerators equipped for energy recovery may be desirable, since the process of energy recovery can be used to control the gas temperature and assure that the required temperature drop is achieved.

- Increasing the mass loading of fly ash or other adsorbent with which the thermal processing unit gas stream is in contact, may significantly increase the amount of dioxins that are irreversibly adsorbed by the adsorbent.

- Decreasing the particle size of a given adsorbent, or replacing low surface area adsorbents with high surface adsorbents may significantly increase the amount of dioxins that are irreversibly adsorbed on the adsorbent.

- If E_A for a given adsorbent is large, the contact time for efficient dioxin adsorption may be long. In some instances, this may necessitate usage of packed bed adsorbers - for providing longer contact times and higher surface area (mass loading of adsorbent) per unit volume. In addition adsorbent beds would likely have greater thermal inertia and therefore may afford more uniform control of the adsorbent temperature. Adsorbent beds may also provide better particle size control.

- Competitive (multicomponent) adsorption may limit the mass of dioxins that is adsorbed. Research should be conducted to identify adsorbents which are inexpensive and which preferentially (selectively) adsorb dioxins and similar compounds, e.g., furans.

- The logic presented here for potential control of dioxin emissions should also be applicable to control (containment) of other toxic substances. Conditions for optimal containment may be determined by research, development and testing as suggested above. The economics of applications of the suggested methodologies for dioxin containment have not been determined. However, sorbent cleanup of polycyclic organic matter emissions through filtering of flue gas at low temperature through a bed of activated charcoal is a common practice in a number of coal fired power plants in the western United States (ref.11).

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