CHAPTER 10

REMOVAL OF GASES AND VAPORS

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10.1 INTRODUCTION

Why spend money to clean the air we breathe when outdoor air is free and (we hope) uncontaminated? One reason is that air that appears clean even to the EPA may still be too polluted to use, for example, in museums and libraries, where even trace levels of contaminants can do irreparable harm. Zoos, restaurants, and funeral homes, to give another set of examples, may produce their own air pollutants in unacceptable concentrations. And finally, we produce our own pollution—we release odors into the ambient air, and it may be more economical to cleanse the indoor air and recycle a certain fraction of it than heating (or cooling), and possibly humidifying or dehumidifying the fresh air from the outside.

Unwanted vapors can be removed by many different procedures, but only adsorption has found widespread use. Other procedures—including absorption by liquid sprays, condensation, catalytic combustion, photocatalysis, plasma-induced reactions, and pressure swing adsorption, may be used for specialized purposes, such as by the military, where the outside air could be highly contaminated and exceptional treatment is needed. Additionally, the development of low-temperature catalyses may lead to their particular uses, including treating air *brought into* the passenger space of automobiles. Also, at high altitudes (e.g., 60,000 ft), ozone can enter the troposphere from the stratosphere through a process of tropopause folding. For this reason catalytic procedures are commonly used to remove ozone from the ventilation air of high-flying aircraft. This chapter reviews current developments in these areas, with emphasis on the most common procedure: adsorption.

10.2 ADSORPTION

Adsorption is in one respect a very simple process; to remove gases and vapors by adsorption, the air is passed through an adsorption bed such as shown in Fig. 10.1. This procedure is used, for example, in commercial buildings (office buildings, beauty salons, print shops, restaurants, retail stores), fitness centers, health care facilities (including hospitals, convalescent homes, extended-care facilities, physicians' offices, laboratories), airports, schools and libraries, industrial plants, social halls, and residences (odors from pets, entertainment, painting, hobbies). Adsorption on activated carbon is especially useful in removing diesel fumes, hydrocarbons, tobacco smoke, body odor, cooking odors, and volatile organic compounds. Impregnated adsorbents are used to remove low-molecular-weight compounds, such as ammonia, hydrogen sulfide, and formaldehyde, which are poorly adsorbed on the unimpregnated adsorbent. The primary disadvantage in this process is that the adsorption beds eventually become saturated with contaminant and must be replaced. As a rough approximation, the time between replacement depends inversely on the concentration of the contaminant passing into the adsorption bed. Thus the lower the concentration of contaminant, the less expensive the cleansing procedure, making this process well suited for use at the low concentration of contaminants usually found in indoor air.

But the use of activated carbon can also be quite complex. Some have called its application "black magic." The purpose of this section is to take away as much as possible of the mystery in its use.

A brief description of the manufacture of activated carbon is a good starting point in gaining an understanding of this unique material (Ecob, undated). In one manufacturing
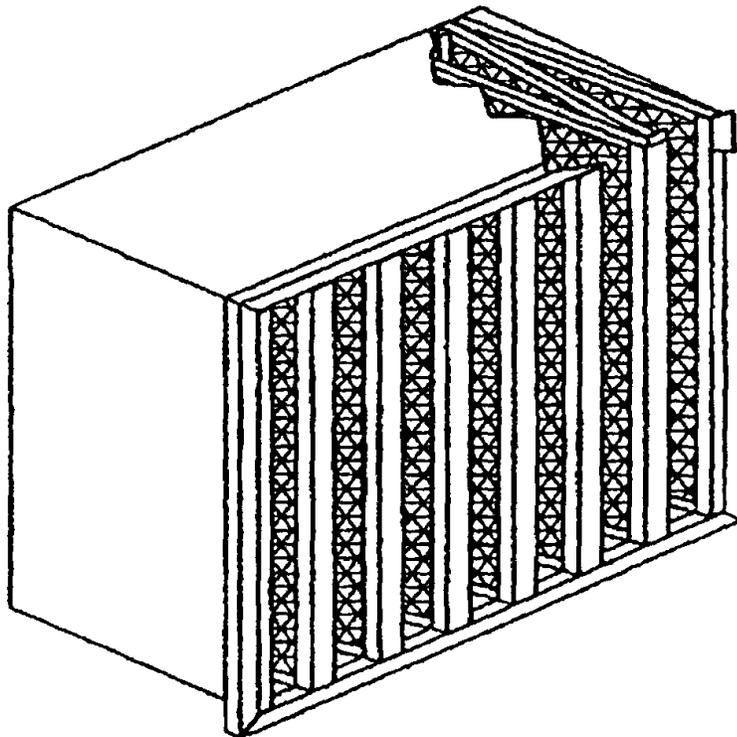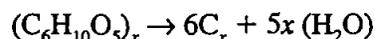

FIGURE 10.1 V-configuration adsorption bed. [*Figure taken from a diagram in commercial literature by Barnebey & Sutcliffe Corp. (ca. 1970).*]

process, a vegetable material rich in cellulose (e.g., coconut shell) is charred in the absence of air to convert the cellulose to carbon. The chemical reaction giving the carbon base is

$$(C_6H_{10}O_5)_x \rightarrow 6C_x + 5x\,(H_2O)$$

In a second step the carbon base is slowly oxidized in the presence of steam, and through this chemical reaction, some carbon is removed:

$$H_2O + C \rightarrow H_2 + CO$$
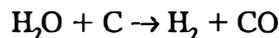
This second step creates the micropores in the carbon, giving the very high surface area necessary for a good adsorbent. The product is activated carbon. Although this process involves chemical reactions, it is known as *physical* activation. Some important facts about activated carbon are

- Activated carbon's structure is like a natural sponge, having a network of pores extending throughout the bulk of the material. As in a natural sponge, there are a wide variety of pore diameters, with the pores smaller than 2 nm termed *micropores,* the pores between 2 and 50 nm termed *mesopores,* and those greater than 50 nm termed *macropores.* Virtually all pores are invisible without a microscope. Pores with diameters less than 5 nm account for most of the adsorptive capacity.
- Internal surface areas of good quality activated carbons are in the range of 1100 to 1200 m^2/g. With this degree of activation, approximately *half* the carbon atoms in the adsorbent are on the internal surface of the pores and available to adsorb contaminants.

Activated carbon can be made from a variety of carbonaceous materials, including coconut shell, wood, peat, and coal—all of plant origin. Most gas-phase filter applications use coconut shell- or coal-based carbon. There is a shift to larger pores as one passes from coconut shell to coal to peat to wood. Activated carbons having small-diameter micropores (microporous carbons) are better adsorbers of small molecules, and are also the most retentive. High-grade coconut-base charcoal removes most volatile organic compounds at a capacity of 33 percent by weight, and is hard enough not to readily dust while in service. However, shell-based carbons are not best for all applications. For example, at relative humidities greater than RH 95%, a coal-based carbon may perform better than a shell-based carbon.

A relatively new material, activated-carbon fibers, is under development, and may supplant granular activated carbon in some of the latter's traditional uses. [See, for example, Mori et al. (1997) and Li et al. (1997).]

Once the activated carbon has been manufactured, how is it best used to purify room air? Some basic problems that face a user of activated carbon include those described in the following subsections.

Selecting the Appropriate Commercial Grade of Carbon

There are many commercial grades of carbon available, and without guidance, one might (quite literally) select an activated carbon more appropriate for barbecuing than for air purification.

The American Society for Testing and Materials (ASTM) has developed basic standards for determining the quality of activated carbon. The first step in working intelligently with activated carbon is to become familiar with these standards, which include the following:

D2652. Standard Terminology Relating to Activated Carbons.   This standard gives the basic definitions required to apply the test procedures that follow. These definitions are particularly important to vendors and consumers of activated carbon, for they define the basic terminology used in contracts to buy and sell activated carbon.

D2854. 'Apparent Density of Activated Carbon.   Without knowledge of the bulk density of the activated carbon, one would not know the *weight* of activated carbon needed to fill an adsorption bed of a given size. The weight of activated carbon present is key to longevity in a given application. In this test the bulk density is determined by dropping the activated carbon at a known rate and distance into a graduated cylinder and weighing the charcoal that is in the cylinder after it is filled.

D2862. Particle Size Distribution of Granular Activated Carbon.   In practice, the charcoal granules can be either too large or too small for the adsorbent to be most effective. If the granules are too small, the pressure drop across a bed will be too high; conversely, if the granules are too large, there is reduced contact between the adsorbent and the air passing through it, and removal efficiency is poor. This test places the adsorbent ' over a set of wire cloth sieves, with the largest mesh sieve placed at the top, and the subsequent sieves placed, going from top to bottom, in order of decreasing mesh size. These sieves are then vibrated for a fixed period of time at a predetermined intensity, and the amount of activated carbon on each screen is determined by weighing. The finer the granules, the greater the percent of the activated carbon that will be found on the lower sieves. Typically a coarse activated carbon may have a mesh size of 4×8; and a finer-mesh–activated carbon a mesh distribution of 16×30. The numbers given here refer to the U.S. sieve series, with the *smaller* numbers referring to the larger mesh sieves.

D2866. Total Ash Content of Activated Carbon.   Activated carbon should contain a minimum of inorganic contaminants. To determine the quantity of inorganic material present, a sample of the adsorbent is autoclaved at 650°C, oxidizing away the carboniferous component, and the resultant ash is weighed.

D2867. Moisture in Activated Carbon.   Activated carbon readily picks up environmental moisture. Activated carbon should be purchased on the basis of *dry* weight; otherwise one is purchasing water at a very high cost. To determine the percent of moisture, a sample is weighed, then dried for 4 h at 150°C, and reweighed. The percent loss of weight is taken to represent the water content.

D3467. Carbon Tetrachloride Activity of Activated Carbon.   Without a large micropore volume, the adsorbent will rapidly saturate with adsorbed material. The uptake of CCl_4 from a nearly saturated stream of CCl_4 is proportional to the available micropore volume. This test describes a standard procedure for such a test, including the quantity of carbon to use, the flow rate of the CCl_4, and the times at which the adsorbent is weighed. Although not mentioned in the standard, the volume of micropore space, in terms of milliliters per hundred grams of adsorbent, is approximately the measured carbon tetrachloride activity divided by the specific gravity of liquid carbon tetrachloride (1.594 at 25°C). Activities for good-quality activated carbons range from 55 to 90 percent, although for some specific uses, such as the control of radioactive gases, a lower activity may be preferable.   ⸓

D3802. Ball Pan Hardness of Activated Carbon.   The activated carbon in an adsorption bed is subject to attrition from the vibrations induced, among other factors, by the air passing through it. As this occurs, the adsorbent is reduced to a powder; and if this occurs rapidly, the activated carbon will settle (causing air to bypass the adsorbent), and the loose carbon will soil the surroundings of the adsorption unit. Here the hardness is determined by shaking a sample of the adsorbent, under carefully controlled conditions, with stainless-steel balls, and the amount of dust that is formed in this process is

weighed. The percent of the adsorbent *that is not* reduced to powder is reported as the "hardness" of the sample. In actuality this is a test for resistance to attrition. A good quality activated carbon may have a hardness of 98 percent.

Determining the Capacity of the Adsorbent for the Contaminant

An adsorption isotherm gives the relationship between the concentration of contaminant and the amount of that contaminant—at a given temperature—that will be adsorbed at equilibrium on the adsorbent. Figure 10.2 shows two typical adsorption isotherms. In this figure, the vertical axis gives the concentration of contaminant in the air and the horizontal axis, the uptake of contaminant on the adsorbent. This information is crucial; unless it is known, for example, that so many pounds of activated carbon will remove some quantity of material, the effectiveness of the adsorbent really is not known. The chemical literature contains literally hundreds of isotherms, and there are excellent reference books describing their use (Smisek and Cerny 1970, Flood 1967). Some isotherms, like the Langmuir isotherm, are appropriate for some uses, but rarely to the user of activated carbon. The two isotherms that a user of activated carbon should be aware of are the BET and the Dubinin–Radushkevich isotherms.

The Brunauer–Emmett–Teller (BET) Isotherm. The BET isotherm is very useful in describing the absorbtion of light gases, especially at low temperatures, such as at the temperature of liquid nitrogen. This isotherm is

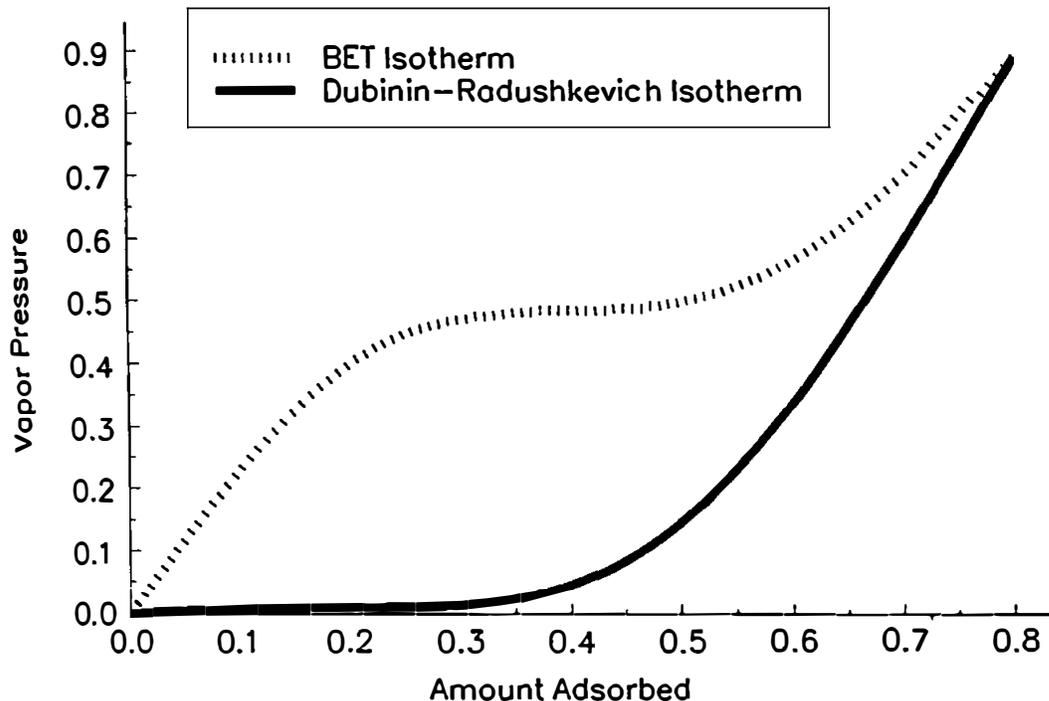$$\frac{x}{V(1-x)} = \frac{(C-1)x}{V_m C} + \frac{1}{V_m C} \qquad (10.1)$$

FIGURE 10.2   The BET and the Dubinin–Radushkevich isotherms.

where C = constant

    V_m = volume of adsorbed gas required to cover the surface of adsorbent with a single layer of molecules (a monolayer)

    V = volume of adsorbed gas

    x = ratio of vapor pressure of adsorbate to vapor pressure of solid or liquid adsorbate at the test temperature (e.g., if the vapor pressure of the compound in the air were 10 mmHg, and the vapor pressure of the pure compound were 100 mmHg, then $x = 0.10$)

The BET isotherm is also important because it allows the surface area of an adsorbent to be determined. If the function of V and x on the left-hand side of Eq. (10.1) is plotted against x, a straight line would be obtained with a slope of $(C - 1)/V_m C$ and an intercept of $1/V_m C$. Thus, from the slope and y intercept of this line, V_m can be determined, and finally, if the surface covered by an individual molecule is known, the surface area of the adsorbent can be calculated from V_m. In these measurements, nitrogen is commonly used as the adsorbate and the area covered by a nitrogen molecule (depending slightly on the temperature) is 16.2 Å^2. The surface area of an adsorbent is an important measure of the quality of the adsorbent. One important fact in using the results of this procedure is that it has an inherent error of about ±10 percent. In purchasing an activated carbon, the buyer should be aware that differences between surface areas less than 10 percent may be meaningless.

The Dubinin–Radushkevich Equation. Our interest is generally in the adsorption of larger molecules, however, such as volatile organic compounds. For these substances, the Dubinin–Radushkevich equation has been found to give better results. The basis of this equation is the assumption that the activated carbon contains micropores in which the adsorbed organic vapors can condense out in the form of a liquid (Astakhov et al. 1969, Dubinin and Astakhov 1971). This equation is

$$M = V_0\, \rho \exp\left[-k\,(RT \ln x)^b\right] \tag{10.2}$$

where M = weight of adsorbed material, mL/kg

    V_0 = micropore volume of the adsorbent, cm^3/kg

    ρ = density of the liquid contaminant, g/cm^3

    k = the first structural constant, mol$^b \cdot$ cal^{-b}

    b = the second structural constant, dimensionless

    R = ideal-gas constant, 1.9872 cal/(mol $\cdot$ K)

    T = absolute temperature, K

    x = as defined in Eq. (10.1)

For the basic assumption of the Dubinin–Radushkevich equation—that the adsorbate is held as a liquid in the micropores of the charcoal—to be valid, the adsorbate must be at or below its critical temperature T_c, or else the liquid phase cannot form. Thus at room temperature the Dubinin–Radushkevich would be appropriate for the adsorption of n-butane ($T_c = 152°C$), but not for methane ($T_c = -82.1°C$), on activated carbon.

One strong point of Eq. (10.2) is that the factors in it can be correlated with physical properties of either the adsorbent or the adsorbate. The factors V_0, ρ, P, and P_0 are associated with the adsorbate; whereas the structural constants (k and b) are characteristic of the adsorbent. One reason for using a highly activated charcoal is that the pore volume (V_0) increases as the activation of the carbon is increased.

The structural constants describe the distribution of pore sizes in the activated carbon. In earlier studies it was commonly assumed that the value of b was equal to 2; but values

of b differing from $b = 2$ have been found for many commercial activated carbons (Richter and Schütz 1991). Note that $b = 2$ gives the most commonly used version of the Dubinin–Radushkevich equation and that $b = 1$ gives the Freundlich isotherm. In practice, b usually lies between 1 and 2. The other structural constant k may change slightly as the adsorbate is changed. Sansone and Jonas (1981) give a procedure using the index of refraction to determine the factor k for one adsorbate if the factor k is known for another adsorbate on the same activated carbon.

Determining the Efficiency of the Bed

After the adsorption capacity for the air contaminant is known, for example, in pounds of contaminant per pound of adsorbent, what will be the practical efficiency of the adsorption bed?

As a stream of polluted air passes through an initially clean adsorption bed (Fig. 10.3), there is a band of partially saturated adsorbent between the clean absorbent and the saturated adsorbent. Only when this band reached the exit of the adsorption bed does contaminant pass through the adsorption bed (see line 1 in Fig. 10.4). A goal of good bed design is to keep this band as narrow as possible so that the entire adsorption bed will be near saturation before breakthrough occurs. Should the band be wide, then the breakthrough curve will resemble line 2 of Fig. 10.4. This line shows an appreciable contamination of the air passing through the adsorber before the capacity of the adsorber was spent.
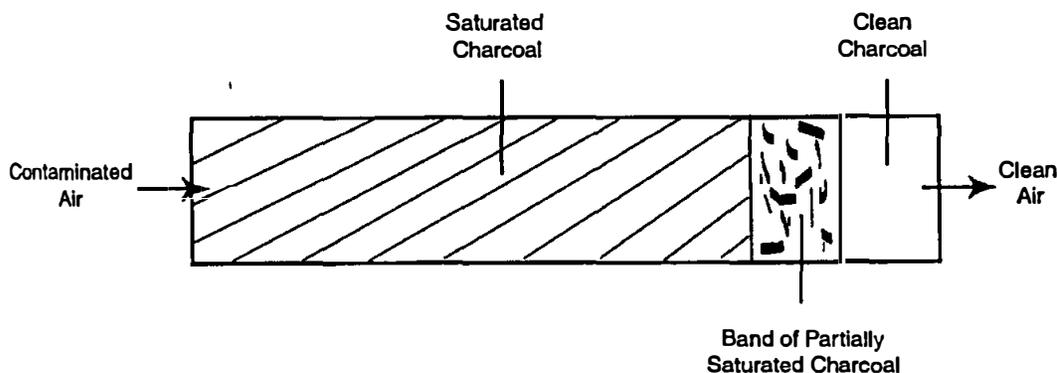

FIGURE 10.3   The adsorption wave in an adsorption bed.

- - - - - -   Narrow band of partially saturated charcoal (Line 1)

——————   Broadband of partially saturated charcoal with early breakthrough (Line 2)
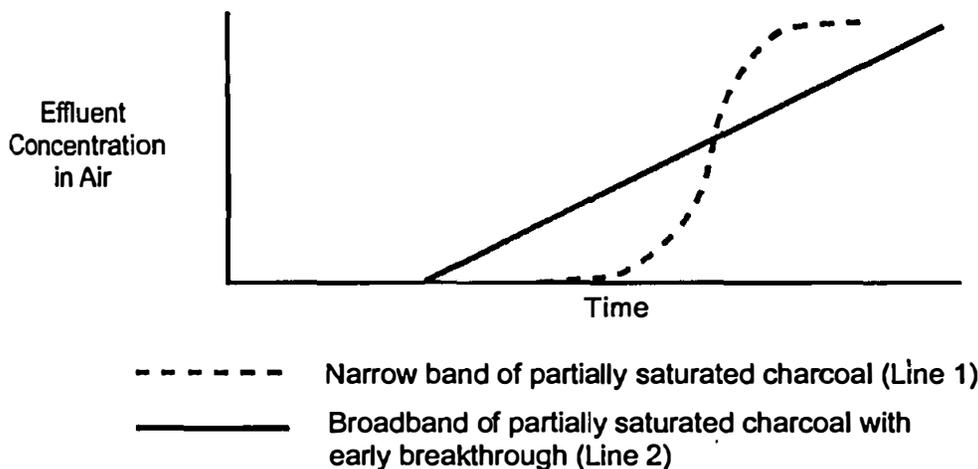
FIGURE 10.4   Two typical breakthrough curves.

Very often a mathematical curve called the J function is fitted to the breakthrough data (Perry 1984), and the parameters that describe this function are then used to characterize the breakthrough curve. The J function has the form

$$J(N, Nt') = 1 - \int_0^N \exp\left(-Nt' - z\right) I_0 \left(2\sqrt{Nt'z}\right) dz \qquad (10.3)$$

where I_0 = modified Bessel function of the first kind
N = number of theoretical plates, dimensionless
t = time from beginning of test challenge, h
t_h = ideal holdup time, h
$t' = t/t_h$

The factors in Eq. (10.3) are easily defined. The ideal holdup time t_h is the time required to saturate the adsorbent bed if the bed took up all the contaminant that passed into it. The number of theoretical plates N describes the steepness of the breakthrough curve. The higher the number of theoretical plates, the closer the breakthrough curve approaches the highly efficient breakthrough curve (line 1 in Fig. 10.4). It has been a common procedure for engineers to study the effect of bed design, temperature, airflow velocity, contaminate concentration, relative humidity—in fact all the variables that might affect the performance of an adsorption bed—and determine how these factors change the mean holdup time and the number of theoretical plates. If the adsorption bed has too few theoretical plates (say, below 10), its performance will be poor.

Fitting the J function to data was once commonly done using a nomograph, but today it can be quickly done by microcomputer (Forbes and Underhill 1986). There are many other possible equations which can be fitted to experimental data, but the J function is not only one of the simplest functions for analyzing the dynamic performance of adsorption beds but also the most commonly used—therefore the results obtained using this function are understandable by engineers working on similar problems.

The semiempirical Wheeler equation (Jonas and Rehrmann 1974),

$$t_b = \frac{v_e}{C_{in}Q}\left\{M_c - \left[\frac{\rho_c Q}{k_v} \ln\left(\frac{C_{in}}{C_{out}}\right)\right]\right\} \qquad (10.4)$$

where t_b = breakthrough time, min
v_e = equilibrium mass adsorbed, g/g activated carbon
$C_{in,out}$ = inlet and outlet concentrations, g/cm^3
Q = volumetric flowrate through bed, cm^3/min
M_c = mass of carbon in bed, g
ρ_c = density of bulk carbon, g/cm^3
k_v = adsorption rate constant, min^{-1} (= $14.41\ U^{0.5}d_p^{1.5}$) ·
U = superficial air velocity, cm/min
d_p = carbon granule diameter, cm

proved useful in the study of the performance of thin adsorption beds, such as respirator cartridges. It is not as versatile as the J equation, but it is easier to use in calculations.

Ensuring Removal of Contaminant by Absorbent in Presence of Water Vapor

Water vapor is always present at far higher concentrations than is any contaminant. For example, at 25°C air saturated with water contains 25,000 ppm of water vapor. How can

one be certain that the adsorbent will remove the contaminants present in concentrations of a few parts per million in the presence of these much higher concentrations of water vapor?

Charcoal can adsorb most organic vapors in the presence of very high concentrations of water vapor for one basic reason—most organic molecules are larger than molecules of water and are more strongly attracted to the activated carbon than is water vapor. It would be very helpful if we knew how relative humidity affects the adsorption of complex mixtures, such as the complex mixture that makes up polluted air. But at this time the best experimental data that are available are for the effect of relative humidity on the adsorption of a single organic component from air. Different results have been seen for the effect of relative humidity on the adsorption of water-soluble organic compounds and on water-immiscible compounds (Underhill 1987, Kawar and Underhill (1999). Figure 10.5 demonstrates the effect of relative humidity on different concentrations of trichloroethylene (Werner 1985). The strong reduction in the adsorption at high relative humidities is apparent; what is not yet well understood is how water vapor interferes with the adsorption of the complex mixture that comprises indoor air contaminants at the low concentrations in which they are found in the ambient air. At this time we can state with confidence that the relative humidity, especially if it is over 55%, is detrimental.

Adsorption of Mixtures

In most cases, indoor air pollutants are a complex mixture of very low, often highly variable (e.g., with occupancy), concentration of compounds, many of which may be unknown at the time that the adsorption unit is selected. The hundreds of volatile compounds present in cigarette smoke illustrate this, and cigarette smoke may be only one of the components present in contaminated indoor air. This is in contrast with what is found in industry, where adsorption is often applied to constant concentrations of one (or at most a few) known components. The unfortunate fact is that the adsorption of complex mixtures at low concentrations is not
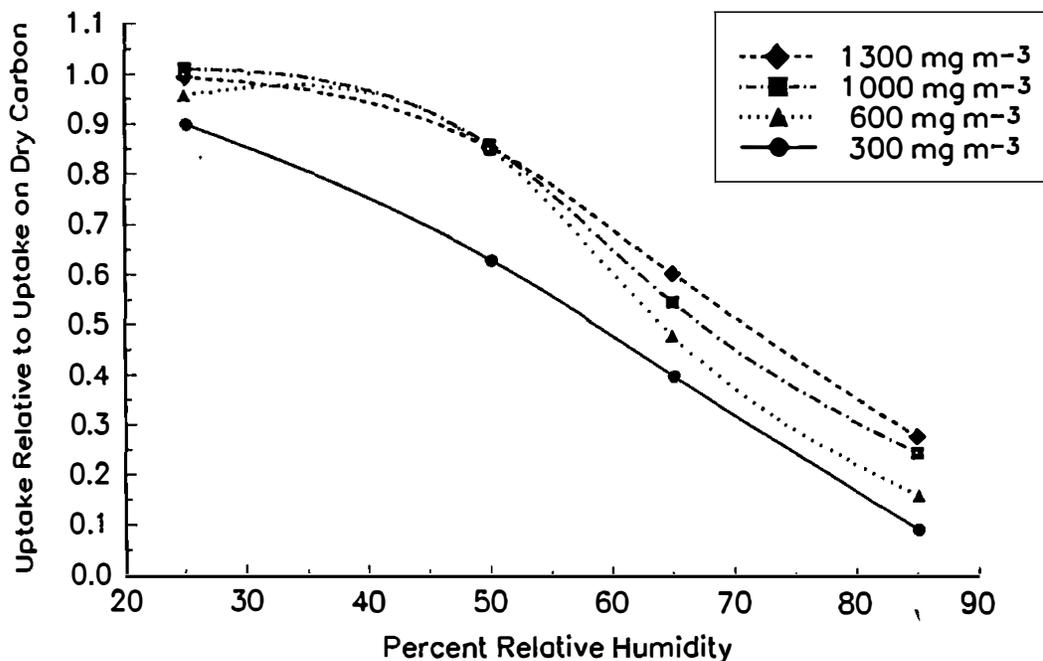

FIGURE 10.5   Effect of relative humidity on the adsorption of trichloroethylene. [*Information taken from Werner (1985). Figure redrawn by Dwight Underhill.*]

well understood. As an illustration, the EPA published test results in which a mixture of the vapors of three components (2-ethoxyethyl acetate, toluene, and propanone) was passed through an adsorption bed. Figure 10.6 shows the result. The lightest component, propanone, passed the most rapidly through the bed. There was also displacement of lighter components by heavier components. The real world is far more complex. Cigarette smoke alone contains hundreds of compounds, and the interactions of these compounds in an adsorption bed is largely unknown. What is known is that activated carbon can adsorb large quantities of indoor air pollutants, even at the low concentrations of these substances found in indoor air. As an example, it is not uncommon for activated carbon to adsorb as much as 20 to 30 percent of its weight in contaminants on exposure to indoor air pollutants before losing its ability to adsorb additional contaminant. One reason for this high adsorption capacity is that many chemical reactions take place between the reactive materials that are adsorbed (aldehydes, organic acids, ketones, sulfides, amines, etc.), and the resultant high-molecular-weight compounds that are formed are relatively nonvolatile. These compounds become so tightly bound to the carbon that the only way to remove them is to reactivate the carbon. But the rate of these reactions, as well as the extent with which they can go forward, is still not well understood. The rate of these reactions is probably far higher at high relative humidities; this is another area where more needs to be learned.

The Basic Design Parameters in an Adsorption Bed

1. *Contact time:* The air must be in contact with the activated carbon for a time—generally at least 0.1 to 0.2 s—for the adsorption to near completion. This contact time t_c is
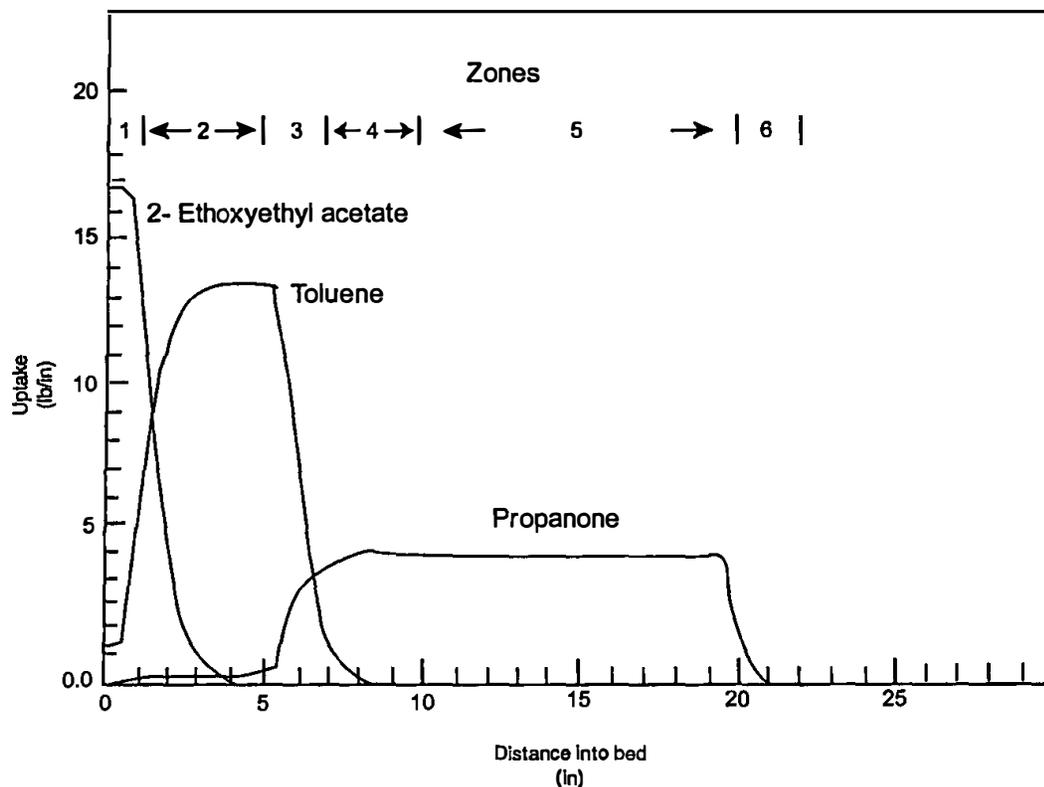

FIGURE 10.6   The adsorption of a three-component mixture. [*Figure taken from MSA Research Corp. (1973).*]

$$t_c = \frac{V_b}{Q}$$ (10.5)

where V_b = volume of activated carbon, m³
Q = airflow, m³/s
t_c = contact time, s

2. *Pressure drop across the unit:* Knowing the pressure drop is important—after all, it is a part of the ventilation system. The first point about the pressure drop is that it is (roughly) directly proportional to the airflow velocity, and generally can be estimated from graphs supplied by the vendor.

The second point is that there are two conflicting factors in bed design that involve the pressure drop; we want to use a large volume of adsorbent, yet to pass the air through a thick bed (Fig. 10.7a) containing this carbon may give an unacceptably high loss of pressure. To get the desired low-pressure drop, either partial bypass (Fig. 10.7b) or extended surfaces are used (Fig. 10.7c–f). The adsorption bed shown in Fig. 10.1 uses a number of V sections placed in parallel. In commercial V banks, there may be 30 to 40 lb of carbon per 1000 cfm of air. Also commercial adsorption beds are available in sizes that match standard ductwork, such as 2 × 2 or 1 × 2 ft. Additional details on commercial designs are available in the literature (ASHRAE 1983, Godish 1989), but this information is subject to change from competition in the marketplace. The most important fact to remember is that in buying an adsorption bed one is purchasing both adsorbent carbon and a unit to hold this adsorbent and place it in close contact with the air passing through the unit. The more adsorbent and the lower the pressure drop across the unit, the better the performance will be over time. It costs less to build a unit that has either a higher pressure drop or contains less carbon, so the buyer must not base a purchase solely on the criterion of the lowest possible cost.

Factors to Consider in Installation of an Adsorption Bed

At the time of installation, important factors to consider are:

1. Is there sufficient vacuum (or pressure) to maintain the desired flow? Often backfitting of existing facilities is requested. A common example is for a hospital in which a heliport has been installed (or in which the weekly testing of emergency power generators) creates unacceptable diesel fumes. If this installation is an upgrade, is there sufficient space and carrying capacity to support the added volume and weight of the unit? Can the adsorption bed be placed in an area permitting easy access to change the adsorber, or to check for leaks? Even small leaks can compromise the effectiveness of a large unit. A rapid, and often effective, procedure for locating leaks in a large instillation is to place a floodlight inside the unit at night, and look for light leaking either outside the unit or across the adsorption bed.

2. Can the unit be placed far enough away from fans or other mechanical devices so that vibration will not cause settling and/or attrition of the activated carbon?

3. Should the unit include either a prefilter or an afterfilter? This depends on the dustiness of the inflowing air, and the degree with which attrition of the carbon can be tolerated.

4. Is the influent air either too humid or too warm for satisfactory performance? If so, can the air be conditioned, perhaps by mixing with outdoor air? If the excess humidity (or even water droplets) is from an air-conditioning unit, can the adsorber (or the air conditioner) be relocated?
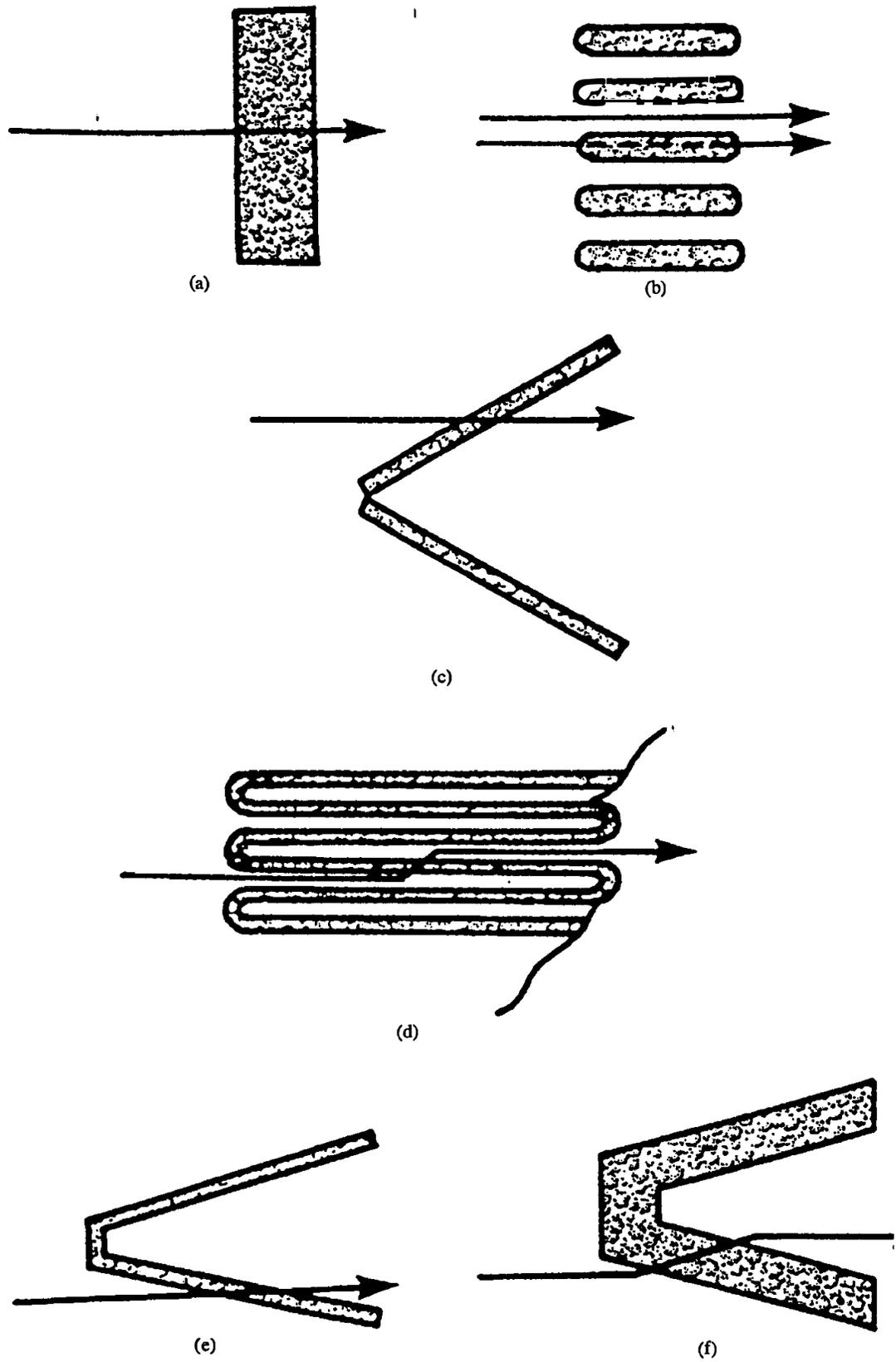
FIGURE 10.7   Adsorption bed configurations. (*a*) Thick bed tray; (*b*) partial bypass; (*c*) trays in V configuration; (*d*) serpentine configuration; (*e*) intermediate bed depth V module; (*f*) thick bed extended surface area module. [*Modified from ASHRAE (1983)*.]

5. The unit must be transported to the installation site carefully—If horizontal trays are to be used (and a horizontal orientation is generally preferred to reduce bypassing through settling), then they should be transported horizontally to prevent settling that will leave airgaps in the adsorber. In a new installation the adsorber should not be operated until it is needed, otherwise contaminants from degassing (from paints, resins, furniture, plastics, etc.) will cause the unit to lose capacity before it is used.

How Long the Adsorbent Can Be Used Before Replacement

It is difficult to tell, other than by loss of performance, when the adsorbent has lost its ability to adsorb additional contaminant. Unlike the pressure drop in filters for particulate matter, the pressure drop across an adsorbent bed does not change as the carbon collects adsorbate (contaminant). There is no way of ensuring continued satisfactory performance other than taking out a sample of adsorbent and testing it, or by replacing the adsorbent before its *predicted* lifetime has been reached. A standard of 4.5 lb/1000 ft^3 per year has been a design guideline for many years, but it may overstate the amount of carbon needed. The time that a unit can remain in service depends on the rate at which organic vapors are generated in the space being deodorized.

Another old guide to estimate the effective use time is the *Odor Index* developed by Barnebey-Cheney Co. From this odor index (see Table 10.1) in typical usage, one pound of charcoal will purify the following cubic feet of space for one year: type A, 2000 ft^3; type B, 800 ft^3; type C, 300 ft^3; type D, 100 ft^3. The difficulty remains that the accuracy of these estimates depends on the release of consistent amounts of adsorbable vapors from similar sources, and, to cite examples, whether we can assume that all funeral homes (or bars) are similar. The idea of ranking areas to estimate their probable need for replacement carbon has merit, and it would greatly advance the field if vendors could find a way to pool their data so that the *Odor Index* could be updated and expanded.

Few industrial test data are appropriate to the indoor environment. For example, tests of the effectiveness of an adsorption bed, challenged with high concentrations of toluene (a stable chemical that generally does not react while adsorbed), may give very different results from the simultaneous adsorption of low concentrations of many reactive compounds. Given the variability in the concentration of contaminants, it will always be challenging to estimate the service life of an adsorption bed in a given situation. But such selection could be facilitated if there were a consistent set of performance standards in which the performance of adsorption beds could be compared under care-

TABLE 10.1 Odor Index for Type of Space (Condensed)

C—aircraft	B—department stores	B—offices
D—air-raid shelters	C—drugstores	C—photo dark works
D—animal rooms	D—funeral homes	D—pollution control
A—apartment buildings	A—homes	C—public toilets
C—apple storage	C—hospitals	C—reception rooms
B—auditoriums	B—hotels	B—restaurants
C—bars	C—kitchens	C—schools
C—beauty shops	C—locker rooms	B—supermarkets
A—churches	D—meat-packing plants	C—telephone exchanges
C—conference rooms	C—morgues	B—theaters

Source: Taken from Barnebey-Cheney Company (ca. 1970).

fully controlled conditions of flow velocity, temperature, relative humidity, challenge gas composition and concentration, among other factors. An example of a performance standard is the German National Standard, DIN 71460, which permits adsorptive filters for automobile cabins to be tested under standard conditions for their effectiveness in removing four very different challenge agents (toluene, butane, sulfur dioxide, and nitrogen dioxide).

10.3 CHEMISORPTION

Chemisorption on Impregnated Activated Carbon

Activated carbon is not effective in removing sulfur dioxide, low-molecular-weight aldehydes and organic acids (especially formaldehyde and formic acid), nitric oxide, and hydrogen sulfide, but it can be impregnated with chemicals (impregnants) that do react with these contaminants. In this case the activated carbon serves as a carrier of the impregnants, enhancing the reaction rate by providing a large surface on which the chemical reaction(s) can transpire. An example of a chemisorbent is as follows. Activated carbon is immersed in a solution of sulfur dissolved in carbon disulfide; the carbon is then dried, leaving the elemental sulfur coating the surface of the carbon. The treated carbon is especially effective in removing mercury vapors from an airstream because the sulfur reacts with mercury vapor to form mercuric sulfide.

Other impregnated carbons have uses as diverse as the removal of hydrogen sulfide, formaldehyde, acid gases, radioactive iodine, ammonia, chlorine, war gases, arsine, phosphine, chlorine, nitrogen dioxide, and sulfur dioxide (Anonymous 1994). The patent literature is an excellent guide to the compounds that have been found to be useful as impregnants. In general, there is no difference between the sorption beds used to contain impregnated versus unimpregnated carbons. However, because a chemical reaction is involved, the contact time must be sufficiently long to allow the chemical reaction to near completion—a time that in practice can vary greatly (e.g., from 0.01 to 0.4 s). Another difference is that increased relative humidity and increased temperature can speed up the rate of the chemical reaction, whereas for physical adsorption these factors are always detrimental.

Many chemical defense systems are based on the use of Whetlerite, an activated carbon impregnated by an ammoniacal solution of copper(II) and chromium(VI) salts. Disadvantages of this chemisorbant are (1) its short lifespan—it may lose 75 percent of its effectiveness in a year; (2) release of the impregnating compounds, ammonia, which causes breathing discomfort, and chromium(VI), known to be carcinogenic; and (3) the need for replacement of the adsorbent immediately after any challenge.

Chemisorption on Permanganate–Impregnated Alumina

Many reactive compounds can be removed from air by chemisorption on potassium permanganate-impregnated alumina (Purafil undated, 1993). Table 10.2 gives comparisons between the adsorption capacities of activated carbon and potassium permanganate–impregnated aluminum oxide for some important air contaminants. In Table 10.2, the adsorptive capacities are percent uptake by weight observed at the point of 95 percent breakthrough of contaminant through the adsorption bed.

From Table 10.2, the potassium permanganate–impregnated aluminum oxide was superior in the removal of most highly reactive compounds (nitric oxide, sulfur dioxide

TABLE 10.2 Comparison of the Capacity (in Weight Percent) of a Chemisorbent and Unimpregnated Activated Carbon Material

Capacity, weight %	Nitrogen dioxide	Nitric oxide	Sulfur dioxide	Formaldehyde	Hydrogen sulfide	Toluene
$K_2MnO_4^+$ Al_2O_3	1.56	2.85	8.07	4.12	11.1	1.27
Activated carbon	9.15	0.71	5.35	1.55	2.59	20.96

Source: Taken from Muller and England (1995).

formaldehyde, and hydrogen sulfide). It was *not* as good as the activated carbon for removing nitrogen dioxide, which is to be expected, as potassium permanganate is a strong oxidizing agent, and nitrogen dioxide is in such a highly oxidized state that it cannot be oxidized further. In the same context, the chemisorbent was very effective in the removal of hydrogen sulfide, an easily oxidized compound. And as would be expected, the chemisorbent had poor capacity for the removal of toluene, an organic compound that is resistant toward oxidation.

In estimating the use time from data such as in Table 10.2, one must remember that the bulk density of permanganate-impregnated alumina is about 50 lb/ft^3, whereas that for granular activated carbon ranges between 30 and 32 lb/ft^3 in most applications. Therefore, nearly twice as much (by weight) of permanganate-impregnated alumina can be placed in the same volume as granular activated carbon. An uptake of 10 percent by weight of contaminant on a system containing permanganate-impregnated alumina may give the same capacity as a 16% uptake on granular activated carbon.

Permanganate-impregnated alumina may initially contain 4 to 8% potassium permanganate by weight. Higher loadings may not give an effective use of the oxidant because of the tendency of potassium permanganate to form crystals at higher loadings. Once the permanganate-impregnated alumina has been put into use, it will gradually lose this initial activity, and the question rises concerning the residual activity of the potassium permanganate. This assay is usually carried out by wet chemistry, in which the steps are (1) eluting the potassium permanganate from the adsorbent, (2) adding an excess of dissolved iodine to the extract, and (3) backtitration with sodium thiosulfate. Results can be obtained far more quickly—and possibly as reliably—by a colormetric analysis of the purple extract, in which the extract is compared to standardized solutions of potassium permanganate.

Standardized tests for permanganate-impregnated alumina are not available from ASTM, and as a rule, the ASTM test procedures for granular activated carbon are not useful for permanganate-impregnated alumina.

Aluminum oxide is naturally hydrophilic, and on normal contact with the atmosphere will pick up 15 to 25% by weight moisture. This moisture is sufficient to maintain a high fraction of the potassium permanganate impregnate in an aqueous solution covering the internal surface of the adsorbent. In this form, the potassium permanganate is highly reactive. If, however, the relative humidity is reduced, say, to <30% for long periods of time, or <5% for short periods of time, then the water film will begin to evaporate, and the potassium permanganate will form small crystals, which are far less reactive. If permanganate-impregnated alumina is to be used in a dry atmosphere, it is advised that pilot tests first be run to determine whether the chemisorbent can be effective. On the other hand, one should avoid conditions where high relative humidity will leach out the impregnant.

Even though chemisorption and physical adsorption—the removal processes on permanganate-impregnated alumina and granular activated carbon, respectively—are quite different, the design of adsorption beds using these materials is essentially the same. The underlying reason is the desire for a short contact time with a low pressure drop. A typical contact time in a permanganate-impregnated alumina bed is 0.01 s, corresponding to 500 fpm passing through a 1-in.-deep bed. A typical removal efficiency of such a bed for reactive contaminants is 85 percent. Such beds operating in series may give adsorption efficiencies of >98 percent.

As Table 10.2 shows, there are airborne species for which the permanganate-impregnated alumina outperforms granular activated carbon and other airborne species for which permanganate-impregnated alumina outperforms granular activated carbon. To try to get the optimal adsorbent where both species are present, it is a common practice to use mixtures with granular activated carbon, usually in a 50/50 mixture by volume. One observation with such mixtures has been that the granular activated carbon often outlasted the permanganate-impregnated alumina, sometimes by factors as great as 3 or 4. The use of heavily impregnated alumina (e.g., containing 8% potassium permanganate by weight) has helped narrow this performance gap.

Applications include airports, commercial office buildings, health care facilities, hotels, industry (including pharmaceutical and microchip manufacturing), museums, restaurants, restaurant cooking grills, university facilities, and zoos.

The time for which permanganate-impregnated alumina may be used before losing its activity is highly variable. In treating outdoor air, a 1-in.-thick bed permanganate-impregnated alumina having a 500 fpm face velocity may last for a year, giving a removal efficiency of 85 percent averaged over this period. But this projected use time varies enormously with respect to the concentration of pollutants, and this, in turn, varies not only from city to city, but also with the location in the city. Small changes, such as taking air in from a high floor rather than at street level, can make a substantial difference in use time.

10.4 OTHER PROCESSES

Catalysts

Ozone is a hazard to flight personnel and passengers in commercial aircraft flown at high altitudes. At 60,000-ft altitude, the mean ozone concentration in the winter months can be 3 ppm, 30 times the FAA (Federal Aviation Administration) exposure limit of 0.1 ppm in the airplane cabin zone. Abatement of this ozone is technically feasible through a number of procedures, but thermal catalytic decomposition using palladium on an alumina substrate is particularly effective (Heck and Farrauto 1995). Catalytic beds for ozone removal are also in common use in xerographic copiers. These catalytic beds have a finite use time, and after they fail, the odor of ozone near the copy machine can become quite intense.

The current patent literature reveals that there is intensive development of new catalytic materials that remove several important air contaminants at or near room temperature. Especially interesting are catalysts that remove carbon monoxide and nitric oxide, as these are key components of automobile exhaust, and thus major pollutants in garages, tunnels, automobile passenger cabins, and other areas. The obvious advantage of room-temperature catalysts over adsorbents is that the former do not become saturated with contaminants.

Pressure-Swing Adsorption

In pressure-swing adsorption, two adsorption beds are operated in parallel (Skarstrom 1972). While the first bed is purifying a stream of compressed air, the second bed uses a fraction of the clean air output from the first bed to backflush from it, at atmosheric pressure, any adsorbed contaminants. Eventually the first bed nears saturation with contaminant and the second bed is cleansed of contaminants. Then the roles of the beds are reversed; the compressed air to be cleansed is passed through the second bed, and a fraction of the effluent air from the second bed is then used to backflush the first bed. The air passing through the bed under compression *not used to backflush* the other bed is the cleansed air that is the product of this process. To give an example of this process, assume that 1 L of air can be passed through the first bed at 10 atm pressure before breakthrough occurs. This 1 L can be decompressed to give 10 L of air at atmospheric pressure. If only 5 L of air is needed to backflush the bed at atmospheric pressure, then the remaining 5 L of purified air are available for other purposes, including air supply to personnel. The cycling time in these systems can be very short, permitting a high flow of purified air. Attractive features of this process include (1) the adsorbent is constantly renewed, and (2) very high purification factors are readily attainable. Pressure-swing adsorption seems especially attractive for supplying air to military personnel in confined spaces such as tanks, where the power required for the pressure cycle is readily available and a high decontamination factor is required.

Plasma Destruction and Photocatalytic Destruction

Plasma treatment of contaminated air has the potential to treat large flows of air with exceptionally high decontamination factors for a wide variety of toxic materials, especially at low concentrations, such as may be released from paint shops, on-site remediation, and pharmaceutical manufacture (Nunez et al. 1993). But the corona discharge can produce by-product nitrogen oxides in sufficient concentrations to make the treated air unbreathable, and thus a second system, perhaps a catalytic converter, may be needed to make the air breathable. There is also the question of cost. Plasma treatment may be too expensive for the routine treatment of low concentrations. One cost estimate is that to remove 5 ppm of CH_2O in 5 h requires a discharge power of ≈ 5 to 20 W/m^3 of occupied space (Storen and Kushner 1993). There are many variations on the plasma model; some systems use the plasma in conjunction with a solid adsorbent on which the decomposition occurs.

A newer approach is the use of ultraviolet (UV) light in conjunction with a titanium dioxide adsorbent (Hisanaga and Tanaka 1995, Raupp and Junio 1993, Peral and Ollis 1997). There is considerable variation between compounds in the degree with which the adsorbent and the UV light interact to cause decomposition of the airborne compounds. Also it is known that some compounds, such as oxygenated compounds can be decomposed by near-UV radiation. A basic difficulty with this new process appears to be obtaining the required minute volume through the air cleaner.

Ozone

Ozone is a very reactive compound easily generated in highly toxic concentrations in air passing through an electrostatic field. A recent review should put to rest the 100-year-old (1900-1999) controversy regarding the use of ozone as an air purifier (Boeniger 1995). There it was reported that ozone, through its own odor, might mask some odors, but this effect is small. Ozone also may react with odiferous organic compounds, but the rate of

reaction is generally very low; for instance, it would take more than 880 years to reduce the concentration of toluene by 50 percent on exposure to 100-ppb ozone. Furthermore, the reaction products formed are often less desirable than the unreacted compounds—for example, the reaction products of ozone with toluene include aldehydes that have a far more unpleasant odor (and are far more toxic) than the parent toluene. Even less desirable is the presence of the unreacted ozone, which at the relatively low concentration of $\cong 120$ ppb can cause eye irritation, visual disturbances, headaches, dizziness, dry mouth and throat, chest tightness, insomnia, and coughing (Sittig 1991). The danger to the general public from ozone generators increased by their use in public areas (such as hotels) without warning to those so exposed. Some vendors try to disguise the fact that their so-called "air-purifying device" generates ozone, so that the person using it will not be aware of the health risks involved. Also, as there are no standards for commercial ozone generators, there is the danger that a particular generator may give a far greater output of ozone than was originally intended. The writer of this chapter once used an ozone generator designed for indoor use to expose laboratory guinea pigs to *lethal* concentrations of ozone. The important point is that despite some misconceptions to the contrary, ozone is a very toxic compound. The harmful effects of human exposure to ozone are of current concern to the American Lung Association, the Environmental Protection Agency, and the Federal Trade Commission.

10.5 RESOURCES FOR FURTHER READING

Essentially all information available in the scientific literature regarding cleansing of indoor air published since 1992 can be found in *Chemical Abstracts* in the general subject index under the recently added heading of *Air Purification.* Under this heading in the 13th collective general subject abstract are approximately 2000 references, including many patents. An older, but still very useful review of the removal of organic vapors from the indoor environment, is *Indoor Air Pollution* (Godish 1989). The basic reference for engineers interested in the removal of either gases or particulates remains *The Chemical Engineers' Handbook,* which is updated periodically. Finally, a comprehensive—yet very readable—review of current adsorption theory has been presented by Manes (1998), who pioneered many developments in this area.

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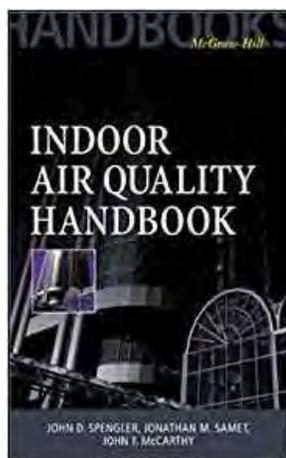
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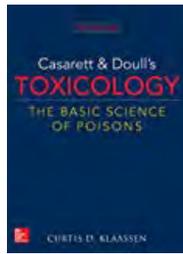## Customers who bought this item also bought

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