



Design and development of radioactive xenon gas purification and analysis system based on molecular sieves



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ABSTRACT

The dynamic adsorption of xenon on molecular sieve packed columns was investigated. The modified Wheeler-Jonas equation was used to describe adsorption parameters such as adsorption capacity and adsorption rate coefficient. Different experimental conditions were accomplished to study their effects and to touch appropriate adsorbing circumstances. Respectable consistency was reached between experimental and modeled values. A purification and analysis setup was developed for radioactive xenon gas determination. Standard sample analysis results approved acceptable quantification accuracy.

1. Introduction

Xenon is a noble gas that is present in ambient air in a concentration of 0.087 parts per million (ppm). It has wide applications in various fields of science, technology and medicine and in most of them, it should be accumulated and isolated (Dresel et al., 2008). In almost all environmental xenon applications, this requirement can be most effectively met by adsorption processes using various adsorbents (Saey, 2009). Fixed beds of granular activated charcoal has been used successfully to remove xenon from gas streams (Abeetal, 2012; Bowyer et al., 2002). From the early studies to recent, different methods have been employed by some research groups for xenon adsorption and separation (Adams et al., 1959a; Charles et al., 2014). Excellent characteristics of 5 Å molecular sieve (MS5A) make it useful for gas adsorption and separation. The MS5A provides great selective retention, which improves the separation of argon, oxygen, and nitrogen. It is a perfect choice for analyzing the trace impurities in inert gases (Munakata et al., 1999). Outstanding features of MS5A provide long term regeneration with remaining adsorption characteristics. While environmental samples likely to have certain amount of oxygen that may cause damaging effects on use of the carbon absorbers, molecular sieve selected as an appropriate adsorbent in this work. Wheeler-Jonas equation is the simplest and most widely used equation that permits prediction of the breakthrough time of vapors on carbon absorbers (Wood and Moyer, 1989). In this work the applicability of the modified Wheeler-Jonas equation broadened as it used to predict breakthrough curves of xenon on molecular sieves. Moreover calculation of adsorption parameters has been performed based on the modified W-J

equation. The effects of temperature and flow rate on these parameters have been investigated completely. The exact amount of MS5A required to develop a setup to isolate and enrich certain amount of xenon gas in a helium mixture was calculated by using adsorption parameters. Afterwards, a gas purification and quantification setup was equipped with three appropriate MS5A packed column absorber eliminating usage of activated carbon and the performance of the setup was investigated based on the standard sample analysis.

2. Theory

In dynamic adsorption processes at low concentrations, the non-linear Langmuir is displayed in Henry equation form as:

$$q = k_d C_{in} \quad (1)$$

The MS5A equilibrium adsorption capacity, q , is described by k_d that can be derived from mass balance of a packed bed absorber (Adams et al., 1959b; Siegworth et al., 1972). This parameter can be calculated approximately from breakthrough curve as below (Chongyang et al., 2010):

$$q = \frac{F t_{0.05} C_{in}}{W} \quad (2)$$

Where F , $t_{0.05}$, W and C_{in} are the inlet gas volumetric flow rate, the equilibrium time (defined as the time when the outlet concentration is at 50% of the inlet concentration), MS5A mass in grams and equilibrium concentration of inlet gas, respectively.

Several models have been proposed to predict the breakthrough

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curves for adsorption of organic vapors based on a mass balance. These models assume that the quantity of vapor entering the bed equals the mass of vapor adsorbed plus the mass of vapor leaving the bed (Mecklenburg, 1930; Wheeler, 1969; Jonas and Rehrmann, 1973; Wood and Stampfer, 1993; Yoon and Nelson, 1984). Among these models the modified Wheeler-Jonas equation has the form (Wood, 2002):

$$t_b = \frac{q \cdot W}{C_{in} \cdot F} - \frac{q \cdot \rho_B}{k_v \cdot C_{in}} \ln \left(\frac{C_{in} - C_{out}}{C_{out}} \right) \quad (3)$$

Where:

t_b = breakthrough time (min) at selected penetration fraction (C_{in}/C_{out})

C_{in} = bed inlet concentration (g/cm^3)

C_{out} = breakthrough concentration (g/cm^3)

W = weight of the adsorbent bed (g)

q = equilibrium adsorption capacity (g/g MS)

F = volumetric flow rate (cm^3/min)

$\rho_B = W/V$, bulk density of the adsorbent bed (g/cm^3), V is the volume of the packed bed

k_v = adsorption rate coefficient (min^{-1})

The equilibrium adsorption capacity, q , and adsorption rate coefficient, k_v , should be determined, either by extrapolation from measurements or by experiments using a reference adsorbent. The adsorption capacity is usually calculated from an adsorption isotherm equation (Wood and Lodewyckx, 2003). Efficient prediction of adsorption rate coefficient by models would improve the estimation of breakthrough time, especially in the lower range, when using the W-J equation. k_v can be calculated from a plot of $\ln[(C_{in}-C_{out})/C_{out}]$ vs. Breakthrough time t_b for varying C_{in}/C_{out} ratios (Wood and Moyer, 1989). Many factors, such as temperatures, flow rates and concentrations can affect q and k_v . Equation (3) yields the following form:

$$t_b = A - B \ln \left(\frac{C_{in} - C_{out}}{C_{out}} \right) \quad (4)$$

Where, $A = \frac{qW}{C_{in}F}$ and $B = \frac{q\rho_B}{k_v C_{in}}$. According to equation (2) $A = t_{0.05}$. If one plots the breakthrough time (t_b) as a function of $\ln[(C_{in} - C_{out})/C_{out}]$, the theory predicts that a straight line results, where the intercept (A) and slope (B) permit calculation of the adsorption capacity and overall rate coefficient as:

$$q = \frac{AC_{in}F}{W} \quad (5)$$

$$K_v = \frac{AF}{BV} \quad (6)$$

Equation (2) can be used to calculate the breakthrough time at different flow rates based on known k_v and q .

3. Material and methods

The chromatographic adsorption column filled with a MS5A that was purchased from Merck Company (Darmstadt, Germany). Using the data for the calculation of Brunauer–Emmett–Teller (BET) and Barret–Joyner–Halenda (BJH), specific surface area, pore volume and pore diameter was accomplished in accordance with literature reports (Rouquerol et al., 1994) and the results are given in Table 1. The MS5A was dried in an oven (Memert GmbH, Germany) at 473 K for 2 h to remove moisture and then it was cooled in a desiccator to room temperature. Purified helium (99.999%) used as carrier gas, was purchased from Air Products, China. The pure xenon gas (99.999%) was procured from Technical Gas, U.A.E. The mixture of xenon with 1000 ppmv concentration in helium equilibrium gas for dynamic adsorption study was provided by domestic company. Isolation and transfer of xenon gas was completed using a stainless steel column, 7 cm long with 3 mm

Table 1
Characteristics of MS5A obtained from BET analysis.

BJH Method Pore Diameter (nm)	BJH Method Pore Volume (CC/g)	BJH Method ($\text{m}^2 \text{g}^{-1}$)	BET Surface Area ($\text{m}^2 \text{g}^{-1}$)
1.22	15.22	83.96	455.6

diameter confined 0.87 g of carbogenic molecular sieve (CMS).

3.1. Xenon dynamic adsorption apparatus

A simplified schematic of the adsorption system used to study xenon dynamic adsorption on MS5A column is illustrated in Fig. 1. MS5A with mass of 21 g was properly packed into a stainless steel column, 100 cm long with the internal volume of 22.68 mL. A glass wool filter (Merck, Darmstadt, Germany) was placed at both ends to prevent loss of MS5A during sample loading stage. Before first adsorption experiment, MS5A packed column was conditioned by heating up to 443 K for at least 2 h in the oven and helium gas was provided at a volumetric flow rate of 60 mL min^{-1} till the column cooled to the room temperature. This procedure repeated after each adsorption experiment to regenerate the column. The MS5A packed column was placed in a controllable temperature water bath. The stream of pure helium gas and helium-xenon gas mixture cylinders was directed through the column using a solenoid valve. At first step whole system pipes evacuated using vacuum pump (VP25, PRO-SET, USA) and then filled with helium. The adjusted gas flow of carrier gas was controlled by a digital mass flow controller (FC-2900 V, Mykrolis, TX, USA). Then the mixed gas transferred to a six port valve sampler of gas chromatographic system passing through the column (680, Buck scientific, USA). 1 mL of eluted xenon output of the adsorption column was injected to GC column every 5 min using sampling loop placed on the rotary valve. A stainless steel tube packed with Propak Q (80–100 mesh) adsorbent was used as GC column. All eluted xenon gases were analyzed by using a thermal conductivity detector (TCD). The GC oven temperature was adjusted to be 473 K. TCD temperature was fixed at 473 K and pressure of GC carrier gas at the column was adjusted to be 21 Psi. The apparatus makes it feasible to regulate several adsorption parameters such as the gas flow-rate, pressure, carrier gas, temperature, adsorbent length and type. The experiments were designed to study the effect of temperature and mixed gas flow-rate. So the desired temperatures was measured and controlled by means of water bath thermocouple and different flow-rates were controlled by flow controller's needle valve. The experimental xenon dynamic adsorption procedure consists of two sets of measurement runs: 1- Fixed temperature (Isotherm), 2- fixed volumetric flow-rate.

The first flow rates were 50, 100, 150, 215, 300 mL min^{-1} keeping the temperature constant at 298 K. The second run was designed at constant flow rate of 180 mL min^{-1} as temperature varied in range of 268–328 K: (268, 273, 278, 288, 298, 308, 328 K).

3.2. Radioactive xenon purification and analysis setup

The schematic diagram of purification and analysis setup that was used to isolate and identify xenon content of gas mixtures is shown in Fig. 2. The main goal of this setup was to transfer radioactive xenon content in a gas mixed cylinder to a radiation detector. This process was accomplished in three major steps. In the first step, the helium gas and possible air content in the gas cylinder was separated. This step completed by using a 28 cm stainless steel column that was filled with MS5A. The column physical characteristics were carefully chosen based on the results of xenon adsorption studies. To provide proper pressure difference at both sides of column, helium gas was added to mixed gas cylinder (archive bottle) till the inside pressure of cylinder reaching 3 bar. Furthermore, a vacuum pump was connected to the end of

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