J. Chem. Eng. Data **1981**, *26*, 109-112

Measurements of Compressibility Factor and Vapor Pressure for Refrigerant 502

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The compressibility factors and vapor pressures have been measured on Refrigerant 502 by using a Burnett apparatus. Refrigerant 502 is a minimum-boiling azeotrope of Refrigerant 22 ($CHClF_2$) and Refrigerant 115 ($CClF_2CF_3$). The results on the compressibility factor cover the range of temperatures from 273 to 383 K and of pressures from 0.12 to 12.3 MPa corresponding to a density variation from 4 to 1035 kg/m^3. The experimental uncertainty of these 149 results was estimated to be ±0.2%. Twenty-five measurements of vapor pressure were made for temperatures from 273 to 354 K with an experimental uncertainty of ±0.05%. A correlation of vapor pressures was proposed for temperatures from 170 to 355 K. The available experimental *P-V-T* data are widely scattered by ~±2% around the data reported here. None of the equations of state represents the volumetric behavior of the present results.

Refrigerant 502 (R502) is a minimum-boiling azeotrope of 48.8 wt % chlorodifluoromethane (Refrigerant 22 (R22)) and 51.2 wt % chloropentafluoroethane (Refrigerant 115 (R115)). The critical constants of R502 were reported by Martin and Downing (*1*): P_c = 4.07 MPa, T_c = 355.3 K, and ρ_c = 561 kg/m^3. Owing to its superiority in applications to recent refrigeration systems, there have been some experimental studies of the volumetric behavior (*1-4*) and several vapor-pressure measurements (*1-6*). There are, however, considerable discrepancies among those data, which shows the necessity of new, accurate measurements to determine precisely the thermodynamic-state surface of R502. This paper reports 149 measurements of the compressibility factor with an experimental uncertainity of ±0.2% using a Burnett apparatus and 25 vapor pressures with an experimental uncertainity of ±0.05%. The composition of the sample of R502 used was analyzed (49.02 wt % R22 and 50.97 wt % R115) by gas chromatography.

Experimental Section

A Burnett apparatus, shown schematically in Figure 1, and the data reduction method used here were described in detail in a previous publication (*7*). A sample vessel (98-mm i.d., 122-mm o.d., and 500 cm^3 in inner volume) and an expansion vessel (78 mm, 98 mm, and 250 cm^3) were made spherically of 304 stainless-steel and were connected by a constant-volume valve. A differential pressure detector was connected with the sample vessel to separate the sample fluid from the nitrogen in the pressure-transmitting system by a stainless-steel membrane (30 μm thick and 16 mm in diameter). The apparatus was calibrated with 99.9999 mol % pure helium for two isotherms, 323 and 363 K, and the cell constant was determined to be N = 1.50495 ± 0.00018, which is in good agreement with the value of 1.50498 ± 0.00015 in the previous work (*7*).

The Burnett apparatus was completely immersed in a stirred fluid bath whose temperature was controlled within a fluctuation of ±5 mK. A 25-Ω platinum resistance thermometer (Chino: Model R800-1), calibrated with a precision of 5 mK on IPTS-68, was used for the measurement of temperature. The periodic observation of the resistance of the thermometer at the triple point of water showed no significant change. The uncertainity of the temperature measurements was less than ±5 mK.

Balancing the sample fluid with nitrogen by means of the differential pressure detector, we measured the pressure with three different pressure gages: an air-piston pressure gage (Futaba: Model AP-01) for pressures below 0.8 MPa, an oil-operated dead-weight pressure gage (Futaba: Model T) for those between 0.8 and 1.1 MPa, and another air-piston pressure gage (Ruska: Model 2470) for those above 1.1 MPa. The effective ram areas were calibrated as 199.93 ± 0.06, 24.977 ± 0.0013, and 8.386 96 ± 0.000 79 mm^2, respectively. Taking account of the uncertainity due to the temperature fluctuation in the bath, we estimated the uncertainity of the pressure measurements to be ±0.04%.

The compressibility factors were measured by both the Burnett-isothermal expansion method and the Burnett-isochoric coupled method. The measurements of vapor pressure were made under the coexistence of a liquid phase of the sample with its vapor phase in the sample vessel.

Results

Compressibility Factor. The unsmoothed 149 experimental data for the compressibility factor are given in Table I, and density values calculated by observed values of temperature, pressure, and compressibility factor are also tabulated. Here the molecular weight of 111.486 kg/kmol, which is the weighted mean value of each component, i.e., 86.469 kg/kmol for 49.02 wt % R22 and 154.467 kg/kmol for 50.97 wt % R115, without regard to that for the remaining 0.01 wt %, and the universal gas constant of 8.314 33 J/(K mol) recommended by the CODATA (*8*) were adopted for calculating the density values. Measurements along eight isotherms, i.e., 308.15, 323.15, 338.15, 353.15, 358.15, 363.15, 368.91, and 383.15 K, were made by the Burnett-isothermal expansion method, while the Burnett-isochoric coupled method was employed for 21 measurements in the gaseous phase along the 273.15, 283.15, 293.15, and 300.63 K isotherms and for 34 measurements in the dense liquid region for temperatures from 330 to 378 K. The uncertainity of these results is estimated to be no greater than ±0.2%, which is composed of an uncertainity of the pressure measurements, that due to the determination of the value at zero pressure of each isothermal expansion, i.e., less than 0.03%, and that resulting from the temperature fluctuation in the constant-temperature bath. The discrepancies of the two sets of measurements along the 363 K isotherm did not exceed 0.16%, which confirms the reproducibility of the present work.

Vapor Pressure. The present 25 results for the vapor pressure are given in Table II. Fifteen of them were obtained along the 490 kg/m^3 isochore near the critical density by a single series of vapor-pressure measurements, and the others marked with an asterisk in Table II were measured for 10 different densities from 80 to 1035 kg/m^3 during each series of compressibility-factor measurements. Several measurements repeated at three different temperatures, i.e., 326.33, 338.15, and 343.15 K, for various densities agree well with

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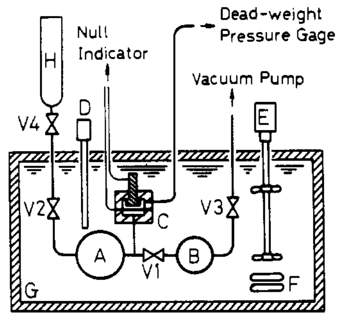
Table I. Compressibility Factors of Refrigerant 502[a]

T, K	P, MPa	Z	ρ, kg/m^3	P_{calcd},[b] MPa	dev,[c] %
273.15	0.136	0.9718*	6.884	0.136	0.0
	0.202	0.9582*	10.36	0.202	0.0
	0.298	0.9372*	15.59	0.297	0.3
	0.433	0.9056*	23.47	0.432	0.2
283.15	0.141	0.9737*	6.881	0.141	0.0
	0.210	0.9612*	10.36	0.210	0.0
	0.310	0.9422*	15.59	0.309	0.3
	0.452	0.9135*	23.45	0.451	0.2
	0.649	0.8702*	35.30	0.647	0.3
293.15	0.147	0.9758*	6.878	0.147	0.0
	0.218	0.9641*	10.35	0.218	0.0
	0.322	0.9467*	15.58	0.322	0.0
	0.472	0.9203*	23.44	0.471	0.2
	0.679	0.8807*	35.28	0.677	0.3
	0.953	0.8210*	53.10	0.952	0.1
300.63	0.150	0.9771*	6.876	0.150	0.0
	0.224	0.9660*	10.35	0.224	0.0
	0.331	0.9494*	15.57	0.331	0.0
	0.486	0.9249*	23.44	0.485	0.2
	0.702	0.8876*	35.27	0.700	0.3
	0.990	0.8322*	53.08	0.989	0.1
308.15	0.154	0.9782	6.873	0.154	0.0
	0.230	0.9679	10.34	0.230	0.0
	0.341	0.9524	15.57	0.340	0.3
	0.500	0.9290	23.43	0.499	0.2
	0.724	0.8940	35.26	0.722	0.3
	1.027	0.8418	53.06	1.024	0.3
	1.405	0.7655	79.86	1.402	0.2
323.15	0.254	0.9697	10.86	0.254	0.0
	0.376	0.9542	16.34	0.376	0.0
	0.552	0.9320	24.59	0.552	0.0
	0.802	0.8989	37.01	0.801	0.1
	1.141	0.8504	55.70	1.140	0.1
	1.576	0.7799	83.82	1.574	0.1
	2.064	0.6789	126.2	2.067	−0.1
338.15	0.185	0.9817	7.474	0.185	0.0
	0.276	0.9717	11.25	0.275	0.4
	0.409	0.9576	16.93	0.408	0.2
	0.602	0.9369	25.48	0.601	0.2
	0.876	0.9065	38.34	0.875	0.1
	1.254	0.8615	57.71	1.252	0.2
	1.744	0.7965	86.84	1.741	0.2
	2.321	0.7042	130.7	2.317	0.2
	2.868	0.5781	196.7	2.874	−0.2
	4.919	0.1885*	1035	5.065	−2.9
353.15	0.266	0.9775	10.34	0.266	0.0
	0.396	0.9655	15.56	0.395	0.3
	0.585	0.9482	23.42	0.584	0.2
	0.856	0.9227	35.24	0.855	0.1
	1.237	0.8852	53.04	1.234	0.2
	1.747	0.8310	79.82	1.742	0.3
	2.384	0.7537	120.1	2.377	0.3
	3.085	0.6479	180.8	3.075	0.3
	3.674	0.5128	272.1	3.668	0.2
	4.831	0.2056*	892.3	4.864	−0.7
	7.966	0.3011*	1004	7.848	1.5
	9.374	0.3442*	1034	9.240	1.5
358.15	0.134	0.9890	5.090	0.134	0.0
	0.201	0.9836	7.661	0.201	0.0
	0.300	0.9755	11.53	0.300	0.0
	0.446	0.9632	17.35	0.445	0.2
	0.659	0.9448	26.11	0.657	0.3
	0.963	0.9174	39.30	0.960	0.3
	1.386	0.8775	59.14	1.382	0.3
	1.948	0.8197	89.00	1.941	0.4
	2.641	0.7382	133.9	2.630	0.4
	3.382	0.6282	201.6	3.367	0.4
	3.986	0.4919	303.3	3.972	0.4
	4.257	0.3491	456.5	4.253	0.1
	4.379	0.2386	687.0	4.409	−0.7
	10.874	0.3938	1034	10.625	2.3
	4.196	0.3989*	393.9	4.189	0.2
	4.311	0.2723*	592.8	4.331	−0.5
	4.356	0.2444*	667.3	4.385	−0.7
	5.803	0.2435*	892.1	5.796	0.1
	9.345	0.3484*	1004	9.121	2.5
363.15	0.327	0.9751	12.39	0.327	0.0
	0.485	0.9616	18.64	0.484	0.2
	0.716	0.9424	28.05	0.714	0.3
	1.045	0.9141	42.22	1.043	0.2
	1.502	0.8728	63.54	1.497	0.3
	2.106	0.8134	95.62	2.099	0.3
	2.846	0.7302	143.9	2.834	0.4
	3.632	0.6193	216.6	3.615	0.5
	4.278	0.4847	325.9	4.258	0.5
	4.632	0.3487	490.5	4.628	0.1
	0.262	0.9801	9.881	0.262	0.0
	0.391	0.9700	14.87	0.390	0.3
	0.579	0.9552	22.38	0.577	0.3
	0.851	0.9324	33.68	0.847	0.5
	1.234	0.8990	50.69	1.229	0.4
	1.757	0.8503	76.29	1.748	0.5
	2.427	0.7806	114.8	2.414	0.5
	3.205	0.6850	172.8	3.186	0.6
	3.956	0.5618	260.0	3.933	0.6
	4.474	0.4221	391.3	4.452	0.5
	4.475	0.4196*	393.8	4.458	0.4
	4.752	0.2961*	592.6	4.782	−0.6
	4.877	0.2699*	667.2	4.919	−0.9
	6.790	0.2811*	891.8	6.729	0.9
	10.732	0.3947*	1004	10.392	3.3
368.91	0.134	0.9902	4.942	0.134	0.0
	0.201	0.9854	7.438	0.201	0.0
	0.301	0.9781	11.19	0.301	0.0
	0.448	0.9671	16.85	0.447	0.2
	0.663	0.9505	25.35	0.661	0.3
	0.972	0.9258	38.15	0.969	0.3
	1.405	0.8894	57.42	1.400	0.4
	1.989	0.8367	86.41	1.982	0.4
	2.730	0.7631	130.0	2.717	0.5
	3.570	0.6631	195.7	3.551	0.5
	4.362	0.5384	294.5	4.335	0.6
	4.933	0.4046	443.2	4.911	0.4
	5.495	0.2994	667.0	5.537	−0.8
	12.331	0.4466	1004	11.854	4.0
	4.789	0.4421*	393.7	4.765	0.5
	5.268	0.3232*	592.5	5.301	−0.6
	7.937	0.3236*	891.6	7.806	1.7
383.15	0.124	0.9921	4.387	0.124	0.0
	0.186	0.9878	6.602	0.186	0.0
	0.279	0.9818	9.936	0.278	0.4
	0.416	0.9727	14.95	0.415	0.2
	0.617	0.9592	22.50	0.616	0.2
	0.909	0.9391	33.87	0.907	0.2
	1.325	0.9096	50.97	1.321	0.3
	1.900	0.8667	76.70	1.893	0.4
	2.656	0.8053	115.4	2.647	0.3
	3.584	0.7220	173.7	3.566	0.5
	4.593	0.6149	261.4	4.565	0.6
	5.548	0.4936	393.4	5.515	0.6
	6.563	0.3880	592.0	6.586	−0.3
	10.823	0.4252	890.9	10.469	3.4
329.95	2.538	0.0996*	1035	2.771	−8.4
333.15	3.461	0.1346*	1035	3.667	−5.6
334.15	2.832	0.1130*	1005	2.993	−5.4
336.65	3.492	0.1383*	1005	3.634	−3.9
339.15	4.160	0.1636*	1005	4.274	−2.7
343.15	6.392	0.2414*	1035	6.460	−1.1
	5.237	0.2036*	1005	5.296	−1.1
348.15	7.878	0.2933*	1034	7.852	0.3
	6.595	0.2528*	1005	6.573	0.3
	3.879	0.1674*	892.5	3.933	−1.4
356.15	4.155	0.2344*	667.4	4.171	−0.4
373.15	5.018	0.4581*	393.6	4.989	0.6
	5.651	0.3428*	592.3	5.684	−0.6
	8.791	0.3544*	891.4	8.598	2.2
378.15	5.285	0.4762*	393.5	5.253	0.6
	6.106	0.3656*	592.2	6.135	−0.5
	9.805	0.3902*	891.1	9.533	2.9

[a] Values with an asterisk were measured by the Burnett-isochoric coupled method. [b] Values calculated by Martin–Downing equation (*1*). [c] $100(P - P_{calcd})/P_{calcd}$.



A: sample vessel B: expansion vessel
C: differential pressure detector
D: platinum resistance thermometer
E: stirrer F: electric heater
G: constant-temperature bath
H: sample bottle V2, V3, V4: valves
V1: constant-volume valve

Figure 1. Schematic diagram of the Burnett apparatus.

Table II. Vapor Pressures of Refrigerant 502[a]

T, K	P_s, MPa	P_{calcd},[b] MPa	dev,[c] %
354.30	3.986	3.985	0.03
351.43	3.766	3.767	−0.03
347.83	3.507	3.508	−0.03
343.96	3.247	3.248	−0.03
343.15	3.195*	3.195	0.00
343.15	3.194*	3.195	−0.03
340.18	3.008	3.008	0.00
338.15	2.887*	2.886	0.03
338.15	2.885*	2.886	−0.03
338.15	2.886*	2.886	0.00
338.15	2.886*	2.886	0.00
335.47	2.729	2.730	−0.04
331.24	2.498	2.497	0.04
329.15	2.390*	2.388	0.08
326.33	2.247*	2.247	0.00
326.33	2.247	2.247	0.00
323.18	2.096*	2.096	0.00
321.16	2.003	2.004	−0.05
314.75	1.732	1.731	0.06
308.59	1.495	1.495	0.00
302.90	1.299	1.300	−0.08
296.13	1.092	1.092	0.00
290.15	0.931*	0.931	0.00
283.93	0.782	0.782	0.00
273.15	0.567	0.567	0.00

[a] Values with an asterisk were measured under the vapor–liquid coexistence during each series of compressibility-factor measurements. [b] Values calculated by eq 1. [c] $100(P_s - P_{calcd})/P_{calcd}$.

Table III. Numerical Constants of Coefficients in Eq 1

$A = -6.97204$	$T_0 = 355.31$ K
$B = 2.1317$	$P_0 = 4.0645$ MPa
$C = -3.0939$	

each other within a discrepancy of ±0.03%, which shows not only that the reproducibility of the present measurements was satisfactory but also that the change of the compositions of the sample used was not detected for all measurments reported here. The uncertainity of the present results is estimated to be no greater than ±0.05%, considering both the uncertainity of pressure measurements and that resulting from the temperature fluctuation in the constant-temperature bath.

The results for the vapor pressure were fitted as a function of temperature to eq 1, with an average deviation of 0.02%.

$$\ln (P_s/P_0) = (T_0/T)[A(1 - T/T_0) + B(1 - T/T_0)^{1.7} + C(1 - T/T_0)^{2.5}] \qquad (1)$$

Here, P_0, T_0, A, B, and C are adjustable parameters, and their numerical values are given in Table III. The values calculated by eq 1 and the deviations of the present results from them are shown in Table II.

Discussion

Compressibility Factor. The present results for the compressibility factor were compared with the available experimental data (*1–4*). Martin and Downing (*1*) measured the $P-V-T$ properties of R502 (48.8 wt % R22 + 51.2 wt % R115) in the range of temperatures between 300 and 396 K to a maximum pressure of 13.6 MPa corresponding to 1093 kg/m^3, and most of their data agree with the present results within a difference of ±0.3% in the density range up to 600 kg/m^3. Their data in the dense liquid region are, however, largely scattered and deviate from the present results by ~±2%. Todorović (*2*) measured the $P-V-T$ properties of R502 (48.81 wt % R22 + 51.19 wt % R115) in the gaseous region between 293 and 362 K to a maximum density of 79 kg/m^3 corresponding to 1.7 MPa, and his data agree with the present results within a difference of ±1%. Döring (*3*) measured the $P-V-T$ properties of R502 (48.8 wt % R22 + 51.2 wt % R115) in the gaseous region between 275 and 367 K to a maximum pressure of 3.1 MPa, and his data are systematically smaller than the present results by ~0.3% on an average with a scatter of 0.5%. Wakamatsu (*4*) measured the $P-V-T$ properties of R502 (48.8 wt % R22 + 51.2 wt % R115) in the range of temperatures between 307 and 424 K to a maximum pressure of 8.6 MPa corresponding to 472 kg/m^3, and his data agree with the present results within a difference of ±0.3% in the temperature range from 350 to 383 K. The differences between the present results and Wakamatsu's data increase with decreasing temperature, and his data at 307 K are smaller than the present results by ~1%.

Equation of State. The present results for the compressibility factor were compared with the available equations of state (*1, 3, 9–11*). The pressure values calculated by the equation proposed by Martin and Downing (*1*) and the pressure deviations of the present results from their equation are shown in Table I. None of the equations except the Martin–Downing equation represents adequately the whole $P-V-T$ surface constructed by the present results. However the density region where the values calculated by the Martin–Downing equation are reasonably consistent with the present data is limited up to ~600 kg/m^3.

Vapor Pressure. The present experimental results for the vapor pressure were compared with the available experimental data (*1–6*). The measurements of Martin and Downing (*1*) are larger by ~1%, Todorović's data (*2*) are smaller by 1–2%, and Döring's data (*3*) are much smaller by ~2% with great scatter. Wakamatsu's data (*4*) agree with the present results within a deviation of ±0.5% except for five points. Long's data (*5*) overlap with the present results in the very limited region of temperatures from 273 to 282 K. The values reported by du Pont (*6*) agree with the present data within a deviation of ±1%, although the deviations decrease with increasing temperature.

The deviations of the experimental data from the values calculated by eq 1 are depicted in Figure 2. This figure shows that eq 1 represents not only the present results but also the whole behavior of the vapor pressure of R502 for temperatures between 170 and 355 K. The values calculated by eq 1 were also compared with the other equations (*1, 3, 9–11*). None of these equations except that proposed by Martin and Downing (*1*) represents adequately the behavior of the vapor pressure of R502. The values calculated by the Martin–Downing equation agree with those calculated by eq 1 within a deviation of ±0.2% between 325 and 355 K and within ±1.2% between 170 and 325 K.

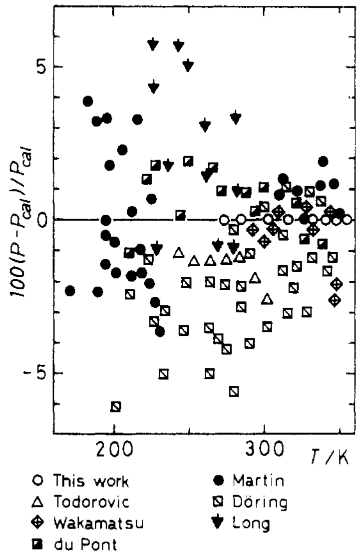


Figure 2. Comparison of experimental vapor pressures with eq 1.

Acknowledgment

We are greatly indebted to Mitsui Fluorochemicals, Co., Ltd., Tokyo, for kindly furnishing the sample of R502. We are also grateful to the National Research Laboratory of Metrology, Ibaraki, for their calibration of the thermometer. H. Kobayashi and M. Suzuki have provided valuable assistance in the experiments.

Glossary

A, B, C	coefficients in eq 1
P	pressure in MPa
P_c	critical pressure in MPa
P_s	vapor pressure in MPa
P_0	coefficients in eq 1
T	temperature in K
T_c	critical temperature in K
T_0	coefficients in eq 1
Z	compressibility factor

Greek Letters

ρ	density in kg/m^3
ρ_c	critical density in kg/m^3

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Received for review February 26, 1980. Accepted November 10, 1980. Financial support was provided by the Fukuzawa Memorial Research Fund of Keio University.

Equilibrium Adsorption of Oxygen, Nitrogen, Carbon Monoxide, and Their Binary Mixtures on Molecular Sieve Type 10X

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Pure-gas adsorption isotherms of oxygen, nitrogen, and carbon monoxide on molecular sieve type 10X have been determined at 32, −50, and −150 °F. Binary adsorption data for all pairs of these adsorbates on the same adsorbent have been collected at the same temperatures and a pressure of 760 mmHg.

Introduction

Adsorption data for the oxygen-nitrogen-carbon monoxide system on molecular sieve type 10X have been reported previously from our laboratory. Danner and Wenzel (*1*) provided the pure-component isotherms at −200 °F and the binary-mixture data at the same temperature and 760 mmHg. All of these data were collected on adsorbent samples taken from the same batch of adsorbent and thus provide the most comprehensive data set available in the literature on gas-mixture adsorption equilibria.

Experimental Section

Experimental Apparatus. The apparatus used was of the volumetric type. The total quantity of each gas admitted to the system and the amount of each gas in the vapor remaining after adsorption equilibrium was established were determined by appropriate $P-V-T$ measurements and analysis by a gas chromatograph. The adsorbed-phase parameters were determined by the difference between the quantities of admitted and remaining gases. In the case of gas-mixture studies, the vapor phase was recycled through the adsorbent bed in order to obtain equilibrium.

The temperature was measured with a copper-constantan thermocouple which had been calibrated with a certified platinum resistance thermometer. Pressures were read from a 60-in., U-type mercury manometer. The estimated accuracies are ±0.2 °F, ±0.3 mmHg, and ±0.5% for the total volume adsorbed and ±0.6 mol % for the phase compositions. Details of the equipment and the operating procedures are described in the previous publication of Dorfman and Danner (*2*).

Materials. The adsorbent used was Linde molecular sieve type 10X in the form of $^1/_{16}$-in. diameter pellets of $\sim ^1/_4$-in. length. The adsorbent pellets were a mixture of 80% crystalline zeolites with 20% inert clay bonding material. This is the same batch of sieves used for the previously reported data (*1*, *2*). The nitrogen isotherm data given in Table I at liquid-nitrogen temperature (−318.7 °F) agree within 2% with the data reported by Danner (*3*). From these data the surface area of this adsorbent was calculated by the n-layer BET method (*4*) to be 672 m^2/gm. Before the adsorbent was weighed into the sample holder, it was regenerated at 600 °F under a vacuum of 50 μmHg for a period of more than 12 h. Between adsorption runs

0021-9568/81/1726-0112$01.25/0