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Technical Note

Thermophysical properties of monolithic carbon

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Abstract

The thermophysical properties (effective thermal conductivity k, permeability K, porosity x and effective specific heat C) of two types of monolithic activated carbons are investigated with the intention of designing a high performance generator for sorption refrigeration systems and heat pumps using ammonia as refrigerant. This paper is mainly focussed on the experimental results. Typical values obtained with one of the samples tested are: thermal conductivity = 0.44 W m^{-1} K^{-1}, limiting concentration = 0.36 kg NH_3/kg carbon and carbon specific heat = 1080 J kg^{-1} K^{-1} at 100°C. The permeability results are highly anisotropic. © 2000 Elsevier Science Ltd. All rights reserved.

1. Introduction

Most solid adsorbents exhibit poor heat transfer properties, mainly due to the high porosity of the material. The discontinuity of the solid material leads to lower density and lower thermal conductivity. Many approaches have been developed in order to improve the global heat transfer within the solid adsorbent. The use of consolidated material with highly conductivity binder such as graphite or metallic foam (copper or nickel) is the most usual method in order to increase both thermal conductivity and density [1,2]. For adsorption refrigeration applications, it can improve the specific cooling power by a factor of 6: typically from 0.060 to 0.350 kW/kg [3]. However, this method has both economic and technical limits. The use of highly conductivity material introduces an additional cost that could reach about 10 times the conventional material cost. The consolidation of the solid adsorbent with mixture material increases the resistance to the mass transfer of the refrigerant throughout the bed; the permeability is reduced significantly (up to 10,000 times) and this limits the rate of the sorption process itself. To improve the heat transfer and sorption process within the adsorbent in order to achieve a specific cooling power up to 1 kW/kg [3] with an environmentally friendly refrigerant, it is important to optimise the thermophysical properties such as thermal conductivity, specific heat, permeability, porosity and the contact heat transfer coefficient between the generator wall and the adsorbent. The monolithic carbon is certainly among the novel solid adsorbents that could lead to high performance.

The aim of this paper is to present the results of the experimental investigation of two types of monolithic carbon based on a 208C precursor from Sutcliffe Speakman Carbons Ltd. The two samples (denoted by LM127 and LM128) are monolithic carbon disks or blocs made with coarse and fine powders (compressed, pyrolysed and bound at high temperature), respectively.

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2. Thermal conductivity

The thermal conductivity is investigated by using the guarded-hot plate method based on BS-874 [4]. It is an absolute method for determining the steady state thermal conductivity of materials less than 2 W m $^{-1}$ K^{-1} over the mean temperature range −20–100°C. The determination of the effective thermal conductivity k (which includes the effect of thermal contact resistance) is based on the measurement of the average temperature gradient ΔT produced through the carbon disc sample (diameter: 76 mm; thickness: 10 mm) by a known axial heat flux ϕ under steady-state conditions. When the working conditions (heat flux, water flow rate, temperatures) are set up and the equilibrium is reached, the effective thermal conductivity k is given by the following expression:

$$k = \frac{\phi}{2S}\frac{\Delta z}{\Delta T} \tag{1}$$

where ϕ is the measured central plate heater heating power (W), Δz is average thickness of the carbon sample (m), ΔT is the average temperature drop through the carbon sample (K) and S is the effective heating area of the central plate heater (m^2).

A method of measuring the contact (carbon–aluminium) heat transfer coefficient h based on the measurement of surface-finish of the carbon using Taylor–Hobson analysis [5] and the evaluation of the total effective thermal conductivity k allows estimation of the intrinsic thermal conductivity of the carbon λ. The sample LM128 has the best surface-finish (because of the grain sizes) that leads to a maximum heat transfer coefficient h of about 1500 W m^{-2} K^{-1} while the minimum is about 450 W m^2 K. h is calculated, based on the conduction through the gas film whose mean thickness has been measured directly. With the sample LM127, the maximum is about 550 W m^{-2} K^{-1} while the minimum value is around 200 W m^{-2} K^{-1}. The effective thermal conductivity k decreases slightly with the increase of the sample mean temperature T regardless the sample [6,7]: it decreases from 0.450 to 0.400 W m^{-1} K^{-1} with the sample LM127 and from 0.350 to 0.300 W m^{-1} K^{-1} with the sample LM128 (when the sample mean temperature T varies from 20 to 120°C). The intrinsic thermal conductivity of the sample LM127 decreases from 0.600 to 0.500 W m^{-1} K^{-1} while with the sample LM128, it decreases from 0.400 to 0.350 W m^{-1} K^{-1}. The carbon LM127 has an intrinsic thermal conductivity which is about 50% higher than the carbon LM128. The intrinsic conductivity λ of monolithic carbon LM127 is nearly four times higher than with a granular carbon bed (k = 0.16 W m^{-1} K^{-1} [8]) because of the absence of large voids and the existence of an unbroken conduction path. The expressions of k and λ as a function of temperature (from linear regression of experimental data) and the equivalent mean value of the contact carbon–aluminium heat transfer coefficient h_E are presented in Table 1. The accuracies are about 7 and 13% for k and λ measurements, respectively.

3. Permeability

The axial and radial permeabilities of samples are measured by using a specially designed test rig [9]. The permeability test comprises measurements of the pressure drop ΔP (Pa) (axially or radially) across the sample of a carbon disc when a gas (air or argon) is flowing through it with q_v (m^3 s^{-1}) flow rate. The aim of the test is to establish the relationship between the pressure gradient across the sample (dP/dz or dP/dr) and the gas velocity v (m s^{-1}) and to determinate the intrinsic characteristic of the material. Since the samples to be tested are porous media with very low gas velocities, the Ergun model [10,11] which models both laminar and turbulent regimes is applicable:

$$-\frac{dP}{dz} = \frac{\mu}{K_a}v_a + B_a\rho v_a^2 \quad \text{(axial flow)} \tag{2a}$$

$$-\frac{dP}{dr} = \frac{\mu}{K_r}v_r + B_r\rho v_r^2 \quad \text{(radial flow)} \tag{2b}$$

where μ and ρ are the gas viscosity (Pa s) and density (kg m^{-3}), respectively; B_a and B_r are the material shape coefficients for axial and radial tests, respectively (m^{-1}); K_a and K_r are the axial and radial permeabilities, respectively (m^2); v_a and v_r are axial and radial

Table 1
Thermal conductivity (k, λ) and heat transfer coefficient h_E

Sample	Effective conductivity, k (W m^{-1} K^{-1})	Intrinsic conductivity, λ (W m^{-1} K^{-1})	Heat transfer coefficient, h_E (W m^{-2} K^{-1})[a]
LM127	$0.4455-0.0002 \times T$[b]	$0.6194-0.0008 \times T$[b]	350
LM128	$0.3490-0.0002 \times T$[b]	$0.3885-0.0003 \times T$[b]	800

[a] Equivalent mean value.

[b] T: temperature (°C).

Table 2
Expression of B, K, W and X function of gas flow configurations

Flow configurations	Axial	Radial converging	Radial diverging
B	$> B_a$	B_{rc}	B_{rd}
K	K_a	K_{rc}	K_{rd}
W	$\dfrac{(P_1^2 - P_2^2)S}{2RT\mu m_a \Delta z}$	$\dfrac{(P_2^2 - P_1^2)\pi\Delta z}{RT\mu m_{rd} \log(R_2/R_1)}$	$\dfrac{(P_1^2 - P_2^2)\pi\Delta z}{RT\mu m_{rd} \log(R_2/R_1)}$
X	$\dfrac{m_a}{\mu S}$	$\dfrac{(1/R_1 - 1/R_2)m_{rc}}{2\pi\Delta z\mu \log(R_2/R_1)}$	$\dfrac{(1/R_1 - 1/R_2)m_{rd}}{2\pi\Delta z\mu \log(R_2/R_1)}$

velocities, respectively (m s^{-1}); P is the gas pressure (Pa); r is the sample radius (m); z is the sample length (m).

The permeabilities (K_a and K_r) and the shape factors (B_a and B_r) of samples are identified by using experimental data. The intrinsic characteristic of the material is then calculated from the following expression which is derived from Eqs. (2a) and (2b):

$$W = \frac{1}{K} + BX \tag{3}$$

where W and X are defined in Table 2.

The carbon LM127 (coarse power) has an axial permeability (3.6×10^{-14} m^2) that is about six times higher than the carbon LM128 (fine powder) (0.6×10^{-14} m^2) as shown in Table 3. However, the radial permeability of the sample LM127 (36×10^{-14} m^2) is about 25 times higher than the sample LM128 (1.3×10^{-14} m^2). For the same sample, the radial permeability is about three (for LM128) to ten (for LM127) times higher than the axial permeability. Regarding the radial tests, there is no significant difference between the converging and diverging permeability measurements (less than 5% — order of magnitude of measurements errors) but the shape factor of the converging test is higher than the converging one (10 and 25% with LM127 and LM128, respectively). The anisotropic nature of the permeability is assumed to be caused by the manufacturing process which involves axial compression.

The use of these results is illustrated by a design calculation for the pressure drops ΔP in a monolithic carbon–ammonia generator assuming that there is no sorption process taking place:

$$\Delta P = \frac{RT\mu m_r}{2\pi\Delta z P_m}\log\left(\frac{R_2}{R_1}\right)\left[\frac{1}{K} + B\frac{(1/R_1 - 1/R_2)}{2\pi\Delta z \log(R_2/R_1)}\left(\frac{m_r}{\mu}\right)\right] \tag{4}$$

where $\Delta P = P_2 - P_1$ (Pa), $P_m = (P_2 + P_1)/2$ (Pa), $\mu = 10^{-5}(0.9268 + 0.0040T')$ is the ammonia gas viscosity (Pa s) with T' in °C; T is the bed temperature (K); R is the gas constant ($R \sim 488$ J kg^{-1} K^{-1}); Δz is the total bed length or thickness (m); m_r is radial gas mass flow rate (kg s^{-1}); R_1 and R_2 are the bed outer and inner radius, respectively (m); K is the permeability (m^2) and B is the shape factor (m^{-1}).

The pressure drop as a function of inner diameter ($D_1 = 2 \times R_1$) of the generator is given in Fig. 1 with the following conditions: $m_r = 0.001$ kg s^{-1} (which provides a specific cooling of about 1 kW kg^{-1} of adsorbent [3]), $D_2 = 2 \times R_2 = 50$ mm and $\Delta z = 1$ m. The bed pressures simulated are $P_m = 3$ bar with a bed temperature of 20°C and $P_m = 20$ bar with a bed tem-

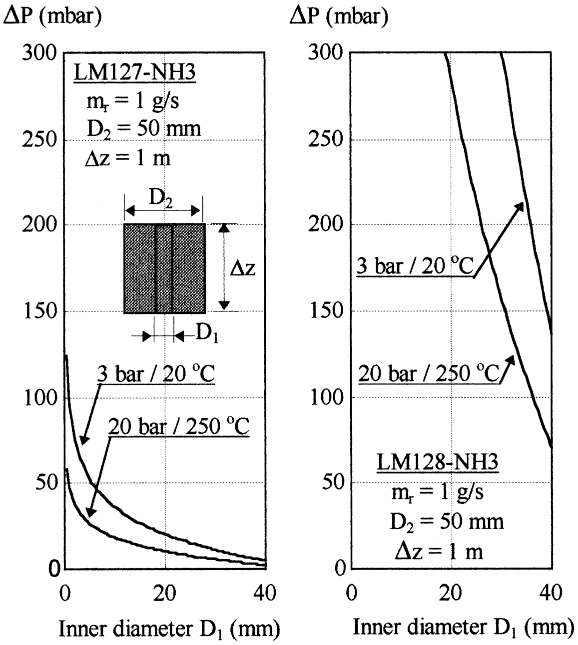

Fig. 1. Radial pressure drop through a monolithic carbon–ammonia (NH_3) bed.

Table 3
Permeability K and shape factor B

Sample	Gas	T_{am} (°C)[a]	T_g (°C)[b]	μ (Pa s) $\times 10^{-5}$	K (m^2) $\times 10^{-14}$	B(m^{-1}) $\times 10^8$
Axial						
LM127	Air	16.3	16.4	1.7838	*3.6065*	*6.6152*
	Argon	18.1	18.0	2.2088	*3.6486*	*5.2962*
	–	–	–	–	3.63[c]	5.95[c]
LM128	Air	19.4	19.0	1.7971	*0.5729*	*58.095*
	Argon	19.1	19.1	2.2150	*0.5658*	*85.815*
	–	–	–	–	0.57[c]	71.96[c]
Radial (converging)						
LM127	Air	18.6	18.6	1.7949	*34.9080*	*0.4863*
	Argon	15.9	15.9	2.1975	*36.4660*	*0.5004*
	–	–	–	–	35.69[c]	0.49[c]
LM128	Air	17.2	17.2	1.7882	*1.2584*	*6.5552*
	Argon	18.5	18.4	2.2110	*1.2901*	*8.1518*
	–	–	–	–	1.27[c]	7.35[c]

[a] T_{am}: ambient temperature (°C).
[b] T_g: gas temperature (°C).
[c] Combined value = mean value.

perature of 250°C (extreme working conditions). As expected, the results show that the pressure drop is higher with low gas pressures. The generator made with carbon LM127 offers the best performance: the radial pressure drop is about 50 mbar (hole diameter D_1 : 5 mm) compared with LM128 generator which has a pressure drop of about 1500 mbar with the same hole diameter.

4. Porosity

The ammonia concentration of the carbon samples is investigated by using the porosimeter developed at Warwick by Critoph [12]. The experimental procedure consists of weighing the mass of adsorbate gas within the sample (M_a) when its temperature varies from 50 to 200°C along five isobars corresponding to saturation temperatures of −15, 0, 15, 30 and 45°C. The ammonia concentration within the sample x is then calculated from the sample mass itself (M_c):

$$x = \frac{M_a}{M_c} \tag{5}$$

where x is the ammonia concentration (kg NH_3/kg Carbon); M_a is mass of adsorbate gas within the sample (kg) and M_c is the sample mass (kg).

The variation of the concentration x using a modified Dubinin–Radushkevich equation [12] is:

$$x = x_0 \exp\left[-K\left(\frac{T}{T_{sat}} - 1\right)^n\right] \tag{6}$$

where T is the sample temperature (K), x_0 is the ammonia concentration under saturation conditions corresponding to the maximum concentration (kg NH_3/kg carbon), T_{sat} is the saturation temperature corresponding to the gas pressure (K) and n is a constant.

The values of x_0, K and n are calculated from experimental data by minimising the sum of the squares of the differences in concentration predicted by Dubinin–Radushkevich equation (6) and those measured. The results for the two sample of carbon (LM127 and LM128) are given in Table 4. The two samples present nearly similar Dubinin coefficients because of the similarity of both base precursors. However, the concentration with monolithic carbon is about 30% higher

Table 4
Dubinin coefficients (x_0, K and n) with carbon–ammonia pair

Sample	x_0	K	n	SEE[a]
LM127	0.3629	3.6571	0.9400	0.0019
LM128	0.3333	3.6962	0.9900	0.0028

[a] SEE: Standard Estimated Error.

Table 5
Specific heat C_{pc} as function of temperature

Sample	C_{pc} (J kg^{-1} K^{-1})
LM127	$802.51 + 2.811 \times T^a$
LM128	$775.62 + 2.826 \times T^a$

[a] T: temperature (°C) — expression from experimental data.

Table 6
Comparison between monolithic and granular carbon beds

Sample	ρ (kg m^{-3})	λ (W m^{-1} K^{-1})	h_E (W m^{-2} K^{-1})	x_{max} (kg kg^{-1})	C_{max} (J kg^{-1} K^{-1})	K_r (m^2)	B_r (m^{-1})
LM127	750	0.60	350	0.36	8000	36×10^{-14}	0.44×10^{8}
LM128	715	0.38	800	0.33	8000	1.3×10^{-14}	5.42×10^{8}
Granular [8]	500	0.16	50	0.29	6500	–	–

than with ordinary granular carbon from the same precursors (208C): the maximum concentration is about 0.36 kg/kg with monolithic carbon (LM127) and about 0.29 kg/kg with granular carbon [12].

5. Specific heat

The effective specific heat of an adsorbent–adsorbate pair depends not only on the temperature and concentration but also on the boundary conditions that determine the heating/cooling process path. If desorption takes place, the effective specific heat will be much larger than if the process is along an isostere (constant concentration). For design purposes, the worst case is an isobaric process of adsorption or desorption and so this specific heat is of particular interest.

The effective specific heat C of samples with ammonia is calculated from the following expression:

$$C = C_{pc}(T) + xC_{pa} + H\left(\frac{\partial x}{\partial T}\right)_P \tag{7}$$

The first term C_{pc} is the sample specific heat (J kg^{-1} K^{-1}) and is given in Table 5. The measurements were carried out using a scanning differential calorimeter by Dr. G. Restuccia at CNR-TAE [13].

The second term $x \cdot C_{pa}$ is the ammonia adsorbed phase contribution with the concentration x from D-R model and the ammonia specific heat C_{pa} approximately constant ($C_{pa} \sim 4900$ J kg^{-1} K^{-1}). The third term $H(\partial x/\partial T)_P$ is the sorption process contribution at constant pressure P where H is the heat of sorption and is given by:

$$H = RA\frac{T}{T_{sat}} \tag{8}$$

where R is the gas constant ($R \sim 488$ J kg^{-1} K^{-1}), T is the sample temperature (K), T_{sat} (K) is the saturation temperature corresponding to the gas pressure P and A is a constant corresponding to the slope of the saturation curve on a plot of $\ln(P)$ vs. $-1/T_{sat}$ ($A = 2823.4$).

On the basis of the concentration test and the similarity of the specific heat of each sample (LM127 and LM128), as we expect, the effective specific heat of the two samples is very close. The maximum value of the constant pressure desorption effective specific heat is about 8000 J kg^{-1} K^{-1} at $P = 20$ bar and $T = 50$°C.

6. Conclusion

The experimental investigation of the conductivity, permeability, porosity, specific heat and heat transfer coefficient leads to the conclusion that the carbon LM127 bed (made with coarse powder) has better thermophysical properties compared with the carbon LM128 (made with fine powder) and ordinary granular carbon beds; the typical values of various properties of monolithic and granular carbon–ammonia beds are shown in Table 6. The reduction in volume from granular bed to monolithic bed is up to 50% and so must also lead to a substantial economic gain when designing and building a carbon–ammonia generator for sorption heat pump and refrigeration machine.

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