Experimental Study of Helium Leakage Parameters in Flexible Composite

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ABSTRACT: The helium leakage characterizations of flexible composites are experimentally studied in this article. Three data processing techniques (Defining method, Fourier series method, and Fast series method) are used to evaluate the helium leakage parameters including the diffusion coefficient D, the solubility S, and the permeant rate k. The chamber pressure variation for helium permeation in flexible composite is measured by the differential pressure method. The results indicate that Fast series method is an effective technology in extracting the helium leakage parameters for flexible composites. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 116: 3562–3568, 2010

Key words: helium leakage parameters; flexible composite; differential pressure method; data processing

INTRODUCTION

Gas leakage is a common phenomenon in many polymer composite structures due to the diffusion of gas molecular and the damage of the material,[1] such as the helium leakage of flexible composite in aerobat,[2] the gas leakage in composite tank, etc. Therefore, the study of gas leakage characteristics is very important in airproof design of composite structures.

The gas leakages of composites have been widely studied via experiments. Yokozeki et al.[3] described some recent experimental techniques for leakage assessment in conjunction with permeability of composite laminates with matrix cracks used in cryogenic fuel tank structures. Kim and Lee[4] developed a new method to measure the gas leakage of adhesively bonded joints by the pressure decay rate. Kumazawa et al.[5] experimentally studied the gas leakage characteristics of damaged carbon fiber reinforced plastic (CFRP) laminates for three stacking sequences under in-plane biaxial loading. Tosti et al.[6] measured the permeance of inert gases (N_2 and He) through coated SiC based ceramic composites, also verified the effectiveness of coatings obtained by chemical vapour deposition (CVD). Mertiny and Gold[7] presented an experimental study about the leakage behavior of glass fibre reinforced epoxy tubular vessels. Tomohiro et al.[8] experimentally evaluated the fuel leakage related to the applicability of high-performance composites for cryogenic propellant tanks used in space launch vehicles. Choi and Sankar[9] investigated the effect of cryogenic cycling on the gas permeability of various composite laminates for cryogenic storage systems. Hino, et al.[10] investigated the change of helium gas permeability of a SiC/SiC composite under the thermal load. Toshio et al.[11] examined helium gas permeability of silicate clay (montmorillonite) particles/epoxy nanocomposites. Choudalakis and Gotsis[12] summarized the permeability of gas molecules in nanocomposite materials that consist of inorganic platelet-shaped fillers in polymeric matrices. Kumazawa and Whitcomb[13] developed a computational fluid dynamics (CFD) model to predict leakage through a damaged laminate. Although there are studies on the measurement of gas leakage rate for carbon fiber reinforced plastic (CFRP) composite laminates and other polymer composites, the gas leakage process is very lengthy in time, and the pressure curve is fair complicated for obtaining the leakage parameters. It is still a difficult task to obtain the leakage parameters including the diffusion coefficient D, the solubility S, and the permeation rate k.[14]

In this article, three kinds of data processing methods in obtaining the helium leakage parameters for flexible composites are described in detail. The important leakage parameters including the diffusion coefficient D, the solubility S, and the permeation rate k are extracted by the helium leakage test of flexible composites.

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PRINCIPLE OF DIFFERENTIAL PRESSURE METHOD

Differential pressure method

In differential pressure method[15] as shown in Figure 1, both a leak-tight master chamber and a measured chamber are simultaneously charged to a certain pressure and then closed from each other. If there is a gas leak in the measured chamber, the gas leak can be detected from the pressure difference between two chambers. This method is similar to pressure decay leak testing except the leak rate is calculated indirectly after measuring the air pressure difference between a fixed, identical reference test volume and air pressure in the part-under-test.

Because the dissolution, diffusion, and penetration of gas is a dynamic process with low velocity, it needs a long time to reach an equilibrium state. For the gas permeation inspection in differential pressure method, the relationships between the pressure or the leaking velocity and the pressurized time are usually obtained, but not directly characterize the permeation properties. Therefore it is necessary to process both the pressure curve and the time to obtain the important permeant parameters.

Permeating equation in differential pressure method

During the permeation measurement in differential pressure method, the testing vessel is separated into two chambers by the film material. In the high pressure chamber, the gas is under the room temperature with 1 atm, and the other chamber is under high vacuum state. Finally the gas pressure p permeated the specimen is measured using high precision gas piezometer and satisfies the following physical model[15]:

$$\frac{dp}{dt} = J\frac{A}{V} \tag{1}$$

where A is the effective area of the specimen with gas permeation, V is the volume of the measured chamber, J is the gas diffusion coefficient, and t is the gas diffusion time. When the dissolution, diffusion, and penetration of the gas reach an equilibrium state, the gas diffusion coefficient J is:

$$J = \kappa\frac{P_0}{h} \tag{2}$$

where κ is the permeant rate, which characterizes the gas permeant velocity of the specimen in equilibrium, h is the thickness of the specimen, P_0 is the pressure in the high pressure chamber. Then the relationship between the measured pressure and the permeant rate is:

$$\frac{dp}{dt} = \kappa\frac{A}{V}\frac{P_0}{h} \tag{3}$$

On the other hand, the dissolution and diffuse control equation of the gas is expressed as

$$\frac{\partial n}{\partial t} = D\frac{\partial^2 n}{\partial x^2} \tag{4}$$

Here D is the diffusion coefficient of the specimen. According to the law of Henry, the gas density n though the thickness of the specimen is

$$n = Sp \tag{5}$$

where S is the solubility coefficient. The corresponding boundary condition is

$$\begin{cases} x = 0, n = p_0 \\ t = 0, x > 0, n = 0 \\ t > 0, x = h, n = 0 \end{cases} \tag{6}$$

Here a vacuum initial environment in the measured chamber is assumed, x is though the thickness direction of the specimen which represents the leakage direction. Furthermore the pressure in the measured chamber can be obtained by solving the eq. (4).

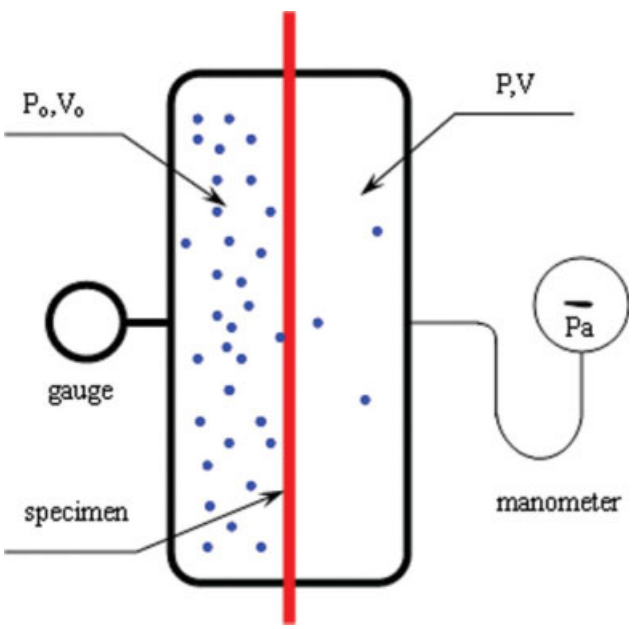

Figure 1 Basic principle of differential pressure method. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

DATA PROCESSING METHOD OF LEAKAGE PARAMETERS

It is known that the main leakage properties of the gas are the diffusion coefficient D, the solubility S, and the permeation rate k. These parameters can be

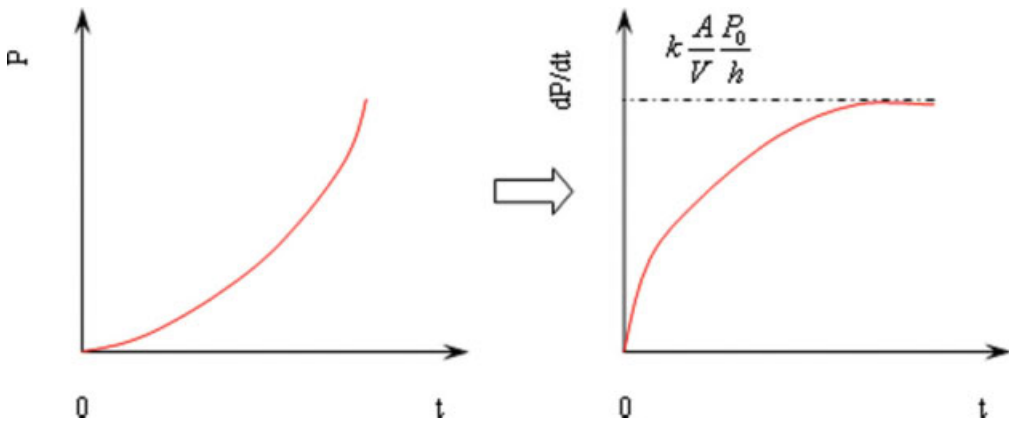

Figure 2 Defining method. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

obtained by means of processing the pressure-time curve. Here three data processing methods are described including Defining method, Fourier series method, and Fast series method.

Defining method

When the gas dissolution, diffusion, and penetration is in an equilibrium state, the permeant rate κ can be expressed as using eq. (3).

$$\kappa = \frac{dp}{dt}\frac{Vh}{AP_0} \tag{7}$$

In eq. (7), the permeant rate κ shows a direct ratio to the gas permeant velocity *dp/dt* under the given testing condition. The relationship between the gas permeant velocity *dp/dt* and the time *t* is obtained by differentiating the $p - t$ curve as shown in Figure 2. When the permeation is in an equilibrium state, the curve $dp/dt - t$ is a fixed value and it is the permeant rate κ. This method can not obtain the diffusion coefficient *D* and the solubility *S*, it also needs a long time to reach the equilibrium state, and it is of no practicability in testing analysis.

Fourier series method

For the film specimen with thickness *h*, the solution of eq. (4) can be expressed using Fourier series method.

$$\frac{d}{p} = p_0\frac{ADS}{Vh}\left(1 + \sum_{k=1}^{\infty} 2\cos k\,\pi\,\exp\left(-\frac{k^2\pi^2 D}{h^2}t\right)\right) \tag{8}$$

When the dissolution and the diffusion is in an equilibrium state, the gas pressure is

$$\frac{dp}{dt} = DS\frac{A}{V}\frac{P_0}{h} \tag{9}$$

Combining eq. (3) with eq. (9), the correlations among the permeant rate κ, the diffusion coefficient *D*, and the solubility *S* are

$$\kappa = DS = \frac{Vh}{A}\frac{1}{P_0}\frac{dp}{dt} \tag{10}$$

After permeating, the gas pressure in the measured chamber is obtained by the integration of eq. (8).

$$p = P_0\frac{ADS}{Vh}\left(t - \frac{h^2}{6D} - \frac{2h^2}{\pi^2 D}\sum_{k=1}^{\infty}(-1)^k\frac{\exp(-\frac{k^2\pi^2 D}{h^2}t)}{k^2}\right) \tag{11}$$

After leaking for a long time, the dissolution and the diffusion is in an equilibrium state, the relationship between the gas pressure and the measured time is regarded as approximately linear, the gas pressure in the measured chamber at any time is

$$p = P_0\frac{ADS}{Vh}\left(t - \frac{h^2}{6D}\right) \tag{12}$$

From eq. (12), it is clear that the $p - t$ curve is beeline as shown in Figure 3. If this beeline is extended, the intercept at the time and the pressure axes is t_c and p_c, respectively. Thus the diffusion coefficient *D* and the solubility *S* are:

$$D = \frac{h^2}{6t_c} \qquad S = -\frac{6V}{Ah}\frac{p_c}{P_0} \tag{13}$$

Finally the permeant rate κ is

$$\kappa = DS = -\frac{Vh}{AP_0}\frac{p_c}{t_c} \tag{14}$$

This kind of method can accurately obtain the permeant rate κ, the diffusion coefficient *D* and the solubility *S*, it is an efficient method. But the convergence of the pressure expression using Fourier series method is very slow.

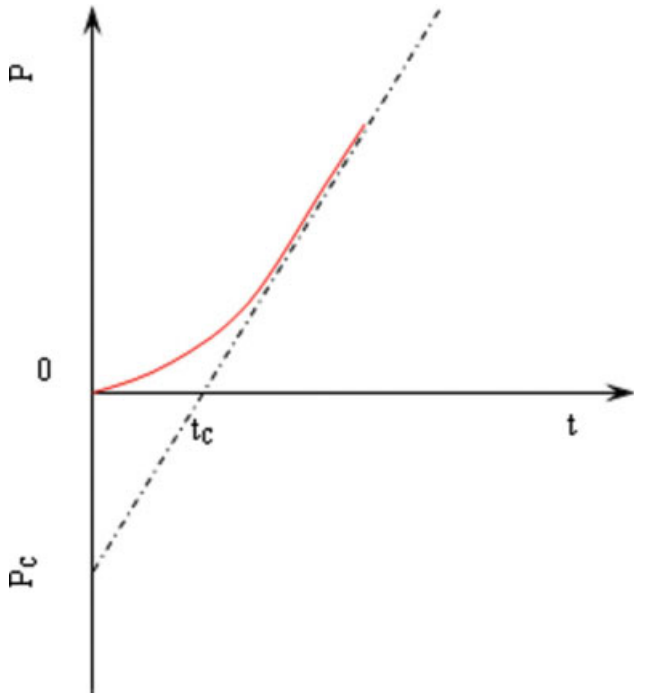

Figure 3 Fourier series method. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

Fast series method

From eq. (8), it can be obtained

$$\frac{dp}{dt} = 2P_0 \frac{AS}{V}\sqrt{\frac{D}{\pi t}}\sum_{k=1}^{\infty}\exp\left(-\frac{h^2}{4Dt}(2k+1)^2\right) \quad (15)$$

Comparing eq. (8)with eq. (15), it is found that the convergence of eq. (15) is faster than the convergence of pressure expression using Fourier series method. This is to say, fast series method shows the convergence in short time and tend to become a stabilization value.

$$\frac{dp}{dt} = 2P_0 \frac{AS}{V}\sqrt{\frac{D}{\pi t}}\exp\left(-\frac{h^2}{4Dt}\right) \quad (16)$$

Take natural logarithm of eq. (16),

$$\ln\left(\frac{dp}{dt}\sqrt{t}\right) = -\frac{h^2}{4D}\frac{1}{t} + \ln\left(2P_0\frac{AS}{V}\sqrt{\frac{D}{\pi}}\right) \quad (17)$$

Then the beeline $\ln\left(\frac{dp}{dt}\sqrt{t}\right) - 1/t$ can be obtained by the measured curve $p - t$ as shown in Figure 4, this slope is $\tan\alpha = -\frac{h^2}{4D}$, the transverse intercept is $1/t_f$. Thus the diffusion coefficient D and the solubility S are,

$$D = -\frac{h^2}{4\tan\alpha}$$

$$S = \frac{1}{P_0}\frac{V}{Ah}\sqrt{-\pi\tan\alpha}\exp\left(-\frac{\tan\alpha}{t_f}\right) \quad (18)$$

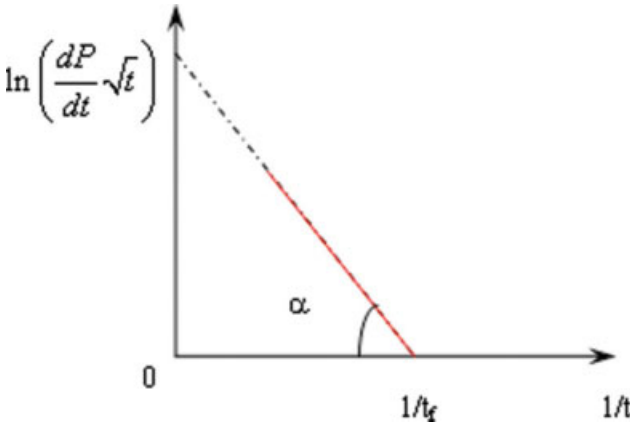

Figure 4 Fast series method. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

In fact, Fast series method is based on the Fourier series expanding method with some merits of Fourier series method, also its convergence is fast and can obtain the permeant properties of the measured specimen in short time.

MEASUREMENT OF LEAKAGE PARAMETER IN FLEXIBLE COMPOSITES

Experimental details

The basic principle of the pressure difference is shown in Figure 5. The chamber is separated into

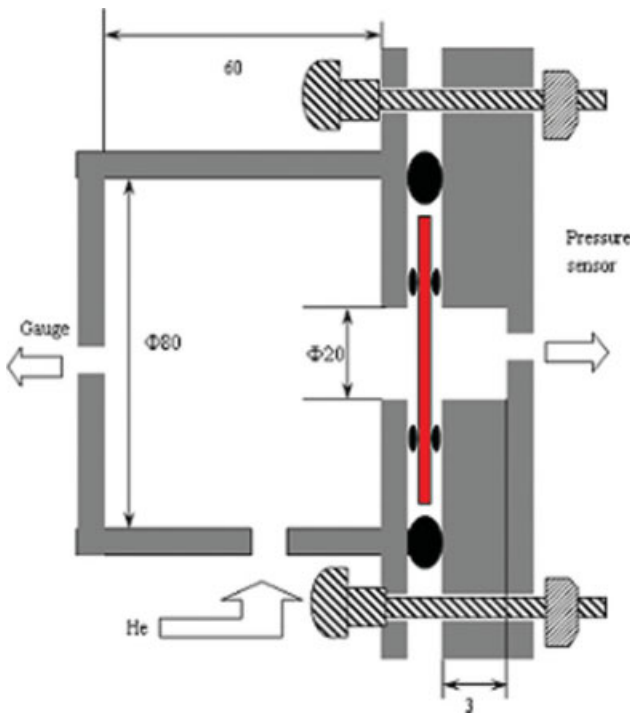

Figure 5 The basic principle of pressure difference method. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

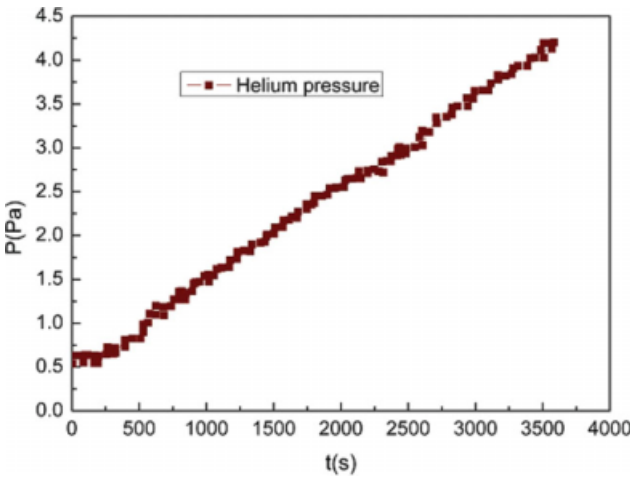

Figure 6 Curve p-t without damage in flexible composite. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

the high pressure chamber and the measured chamber by the specimen. Here, the resistance pressure transducer is used to record the helium gas pressure in the high pressure chamber; the capacitance absoluteness pressure transducer is used to record the helium gas pressure. From Figure 5, it is shown that the volume in the high pressure chamber is about 36 times of the measured chamber, which assures that the helium permeant tolerance has no effect on the gas pressure in the high pressure chamber and the pressure difference is kept stabilized. In this test, the effective penetrant area is a circle with diameter 20 mm.

1. Specimen preparation. The flexible composite with 0.25 mm thickness is clipped into roundness with a 60 mm diameter, which is less than the diameter of the O-type airproof outside ring. Then the specimen is cleaned by alcohol and placed in a dry box for 30 min. In this study, the intact flexible composite is named as skin structure in aerostats. This kind of skin structure is consisted of three different layers,[16] i.e., the protected layer, the load carrier, and the helium barrier, which have different structures and compositions. The main compositions of the helium barrier (Tedlar film) and the load carrier are TEDLAR and Vectran/PU adhesive, respectively. The major materials of the protected layer are TPU intermingling with inorganic nano-particles TiO_2.
2. Daubing the vacuum grease. The contact region between the specimen and the chamber is laid on the vacuum grease for avoiding the gas leakage from the brim of the specimen.
3. Deposited specimen. The specimen is placed in the middle position of the two chambers. Especially, the specimen is within the area of the O-type airproof ring avoiding the gas leakage from the brim of the specimen.
4. Two chambers are tighten by the bolt, and then vacuumed for 24 h.
5. When the vacuum tolerance of the chambers reaches the requirement, the baffle valve is closed. If the pressure in the measured chamber is not exceeded 5 Pa within 1 h, the airproof performance of the system reaches the test requirement.

After checking the airproof performance of the system, the chamber is again vacuumed. Then the baffle valve is closed, let the helium gas enter into the high pressure chamber with 1 atm, the pressure at the measured chamber is measured for almost 10 h. Finally the $p - t$ curve is processed to obtain the leakage parameters.

Extracting permeant parameters

To obtain the permeant rate κ, the diffusion coefficient D and the solubility S without damage the flexible composite, the $p - t$ curve is processed as shown in Figure 6 using Defining method, Fourier series method and Fast series method, respectively.

Defining method

If the curve $p - t$ is calculated using differential coefficient, the corresponding curve $dp/dt - t$ can be obtained as shown in Figure 7. Some fluctuations appeared in this curve, which is due to the low pressure and the inevitable instrument background noise during measurement.

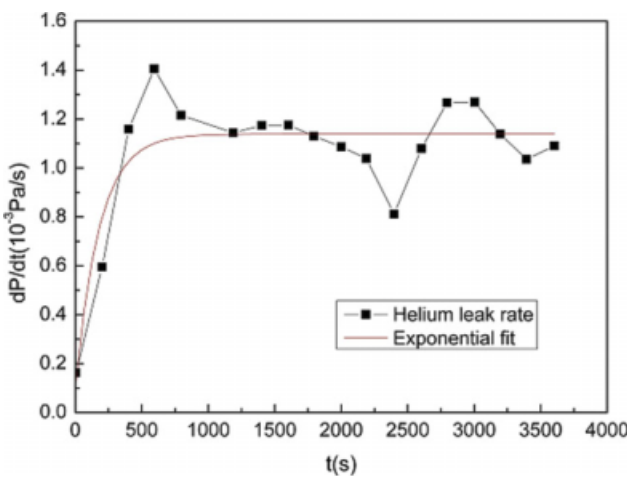

Figure 7 Leakage rate without damage in flexible composite. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

To obtain the stable value, *dp/dt*, in the equilibrium state, an attenuation exponent function is used to fit the curve $dp/dt - t$ in Figure 7.

$$y = A_1 \exp\left(-\frac{x}{t_1}\right) + y_0 \tag{19}$$

Here $A_1 = 1.16702 \times 10^{-3}$ Pa/s, the permeant rate κ is

$$\kappa = \frac{dp}{dt}\frac{Vh}{AP_0} = 8.67 \times 10^{-12}\,\mathrm{m^2/s} \tag{20}$$

Fourier series method

If the stable stage of the curve $p - t$ is linearly fitted, the curve using the Fourier series method is shown in Figure 8.

Based on the linear fitting of curve $p - t$, $p_c = -0.16198$ Pa and $t_c = 136.12$ s, the permeant rate κ, the diffusion coefficient D, and the solubility S without damage in flexible composite are

$$D = \frac{h^2}{6t_c} = 7.65 \times 10^{-8}\,\mathrm{m^2/s}$$

$$S = \frac{6V}{Ah}\frac{p_c}{P_0} = 1.15 \times 10^{-4}$$

$$\kappa = DS = -\frac{Vh}{AP_0}\frac{p_c}{t_c} = 8.79 \times 10^{-12}\,\mathrm{m^2/s}$$

Fast series method

If the curve $p - t$ is calculated using differential coefficient, the corresponding beeline $\ln\left(\frac{dp}{dt}\sqrt{t}\right) - 1/t$ is shown in Figure 9.

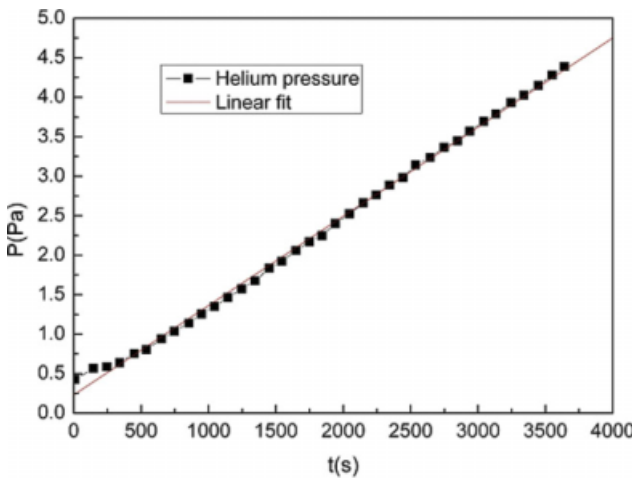

Figure 8 Curve p-t using the Fourier series method in origin specimen. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

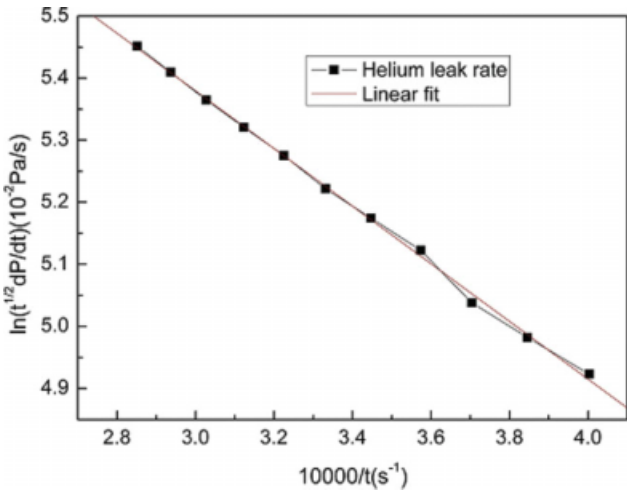

Figure 9 Fast series curve of the leakage without damage in specimen. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

After linearly fitting for Figure 9, $\tan\alpha = -0.21$, the permeant rate κ, the diffusion coefficient D, and the solubility S without damage in flexible composite are

$$D = -\frac{h^2}{4\tan\alpha} = 7.43 \times 10^{-8}\,\mathrm{m^2/s}$$

$$S = \frac{1}{P_0}\frac{V}{Ah}\sqrt{-\pi\ \tan\ \alpha}\exp\left(-\frac{\tan\alpha}{t_f}\right) = 1.17 \times 10^{-4}$$

$$\kappa = DS = 8.69 \times 10^{-12}\,\mathrm{m^2/s}$$

CONCLUSIONS

The analysis methods and the leakage characterization in flexible composites are studied. Some important conclusions are:

1. The basic principles of three data processing methods including the Defining method, the Fourier series method, and the Fast series method, are used for extracting the gas leakage parameters.
2. During the leakage test of flexible composites, permeant rate κ, the diffusion coefficient D, and the solubility S are obtained successfully by three kinds of data processing methods.
3. In the three data processing methods, the defining method can only obtain the permeant rate κ and it is of no practicability in actual tests. The Fourier series method can obtain the permeant rate κ, the diffusion coefficient D, and the solubility S, but the convergence is slow. The fast series method has the merit of the Fourier series method with fast convergence in short time for obtaining the leakage parameters.

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