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Tomohiro Yokozeki , Takahira Aoki & Takashi Ishikawa
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The effect of matrix cracks on gas permeability through CFRP laminates

TOMOHIRO YOKOZEKI [1,*], TAKAHIRA AOKI [2] and TAKASHI ISHIKAWA [1]

[1] *Advanced Composite Evaluation Technology Center, Institute for Space Technology and Aeronautics, Japan Aerospace Exploration Agency, 6-13-1 Osawa, Mitaka 181-0015, Japan*
[2] *Department of Aeronautics and Astronautics, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*

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Abstract—Diffusion-controlled gas permeability through CFRP laminates was experimentally investigated as fundamental research on the feasibility of composite propellant tanks. Using helium gas and a helium leak detector, through-the-thickness gas permeability in CFRP laminated tubes with or without matrix cracks was measured at room temperature. The effect of loadings on the *in situ* gas permeability was also clarified. It is shown that, although gas permeability through CFRP laminates increases on account of the existence of matrix cracks and tensile loadings, these effects turned out not to be crucial in comparison to the leakage through multi laminar matrix cracks, which is three or four orders higher than the diffusion-controlled permeation. These results suggest that the existence of no less than one intact layer is important for the feasibility of composite propellant tanks. Finally, a diffusion model including the combined effects of damages and loads is applied to the experimental results and a successful characterization of gas permeability is presented.

Keywords: Laminates; matrix cracks; gas permeability; diffusivity; continuum damage mechanics.

1. INTRODUCTION

Composite laminates are commonly used as various structural components and as the major candidates for reducing the structural weight of future reusable launch vehicles (RLV). The application of CFRP laminates to cryogenic propellant tanks is especially one of the most sought after but challenging technologies for achieving drastic weight reduction of RLV. Recent basic studies on the feasibility of composite liquid propellant tanks indicated that matrix crack onset and its accumulation is inevitable when applying the conventional high performance composites to the cryogenic tanks and multi-laminar matrix cracks may induce crucial propellant

*To whom correspondence should be addressed. E-mail: yokozeki@chofu.jaxa.jp

leakage [1, 2]. Thus, adequate guidelines for the possible application of CFRP laminates to propellant tanks are necessary from the leakage and damage tolerance point of view.

In this study, diffusion-controlled gas permeability through CFRP laminates is investigated at room temperature using helium gas and tubular specimens. Helium diffusion properties through undamaged laminates are obtained to provide basic information of permeability. In order to evaluate the effects of damage and loads on gas permeability, helium permeation is measured under three conditions: (i) under tensile or compressive loadings without matrix cracks, (ii) with matrix cracks alone, and (iii) under tensile or compressive loadings with matrix cracks. For the evaluation of damage and/or stress induced gas permeability, a diffusion model based on thermodynamics and continuum damage mechanics [3, 4] is applied. A successful characterization of gas permeability through damaged laminates is presented.

2. EXPERIMENTAL PROCEDURE

The material system used in this study is IM600/Q133, an intermediate modulus carbon fiber and toughened epoxy system. This system shows high strength and resistance at cryogenic temperature and the temperature-dependent material properties were obtained [1]. Ply thickness was about 0.15 mm. All tubular specimens were 200 mm long with 50-mm CFRP end-tabs, leaving a 100-mm gauge section, and the inner radius was 30 mm. Four types of specimens with 90-degree layers in the inner or outer layers were prepared: [45/−45/90]s, [45/−45/90_2]s, [90_2/−45/45]s and [90_2/0/90_2].

The test procedures are summarized as follows:

1. Helium permeation tests using undamaged specimens.
2. Helium permeation tests using undamaged specimens subjected to tensile or compressive loadings without inducing damage.
3. Static tensile tests of specimens to induce matrix cracks.
4. Helium permeation tests using cracked specimens.
5. Helium permeation tests using cracked specimens subjected to tensile or. compressive loadings without inducing further damage.

All permeation tests were conducted at 22°C. The details of helium permeation tests and tensile tests are described below.

2.1. Helium permeation test

Tensile loading fixtures with flow path were bonded to tubular specimens and connected to the helium leak detector with a vacuum pump. Helium gas was supplied from outside the gauge sections wrapped with polyethylene film. The differential helium pressure was set to be 1 atm. The helium leak flux into the inside

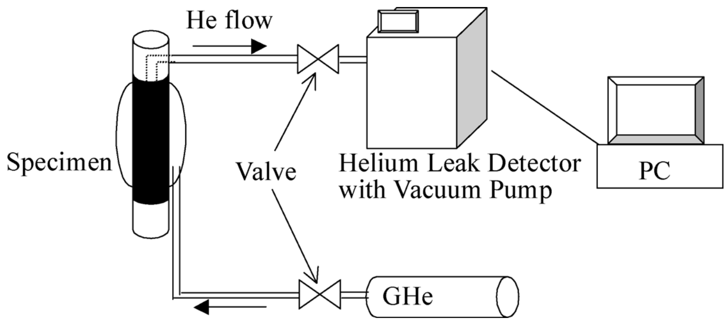


Figure 1. Helium permeation test apparatus.

of tubular specimens was measured with the helium leak detector. The schematic of the test apparatus is shown in Fig. 1.

The gas diffusion properties were obtained using the time-efficient measuring method based on the one-dimensional Fickian model of homogeneous solids [5]. Although it was reported that this model cannot be used to explain the moisture diffusion in polymer matrix composites [6], preliminary studies on gas diffusion in CFRP tubes with or without matrix cracks suggested that the Fickian model can be applied with relatively low errors. One solution of Fick's law with appropriate boundary conditions can be expressed in the form of an infinite series as

$$Q = Q_0\sqrt{\frac{4h^2}{\pi Dt}}\sum_{m=0}^{\infty}\exp\left(-\frac{h^2}{4Dt}(2m+1)^2\right), \quad (1)$$

where Q and Q_0 denote helium leak flux at time t and at steady state, and h and D are the plate thickness and diffusion coefficient, respectively. In the earlier stage of gas diffusion,

$$Q \cong Q_0\sqrt{\frac{4h^2}{\pi Dt}}\exp\left(-\frac{h^2}{4Dt}\right), \quad (2)$$

is employed as an approximation. With simple transformation,

$$\ln\left(Q\sqrt{t}\right) \cong -\frac{h^2}{4D}\frac{1}{t} + \ln\left(Q_0\sqrt{\frac{4h^2}{\pi D}}\right), \quad (3)$$

is used for the extrapolation of measured data by plotting $\ln(Qt^{0.5})$ *versus* $1/t$. Thus, the diffusion coefficient D and steady-state leak flux Q_0 can be determined. For extrapolation, the measured data between 1 h and 2 h after test initiation were used in this study.

For measurement of helium permeation under tensile or compressive loadings, helium was diffused through tubular specimens without loadings to the steady state at first, and then tensile or compressive loadings were applied to the specimens using an Instron 4505 Testing Machine. The steady state is defined in this study as that of less than 1% of leak flux increase during 30 min. Tensile or compressive loadings were applied in incremental steps of 10 min intervals for keeping the loads constant. Thus, the helium leak flux was measured as a function of applied loads.

3. EXPERIMENTAL RESULTS

Measured helium leak flux through a [45/−45/90]s tubular undamaged specimen as a function of time is shown in Fig. 2. Using the extrapolation method, the helium diffusion coefficients of undamaged CFRP laminates were obtained for all specimens. The diffusion coefficients *D* turned out to be independent of stacking sequence and the obtained values of this system at 22-degree centigrade are 2.0×10^{-11} m/s^2.

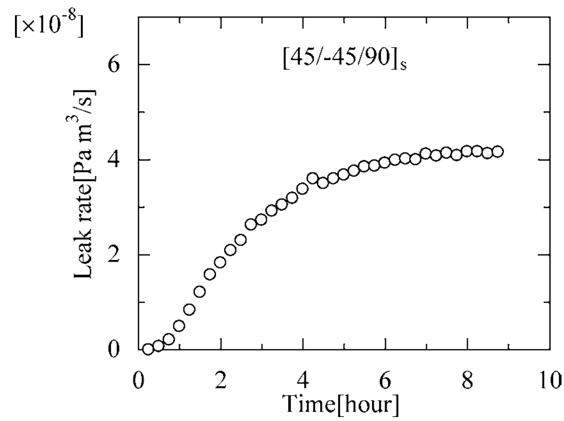


Figure 2. Helium leak flux as a function of time through undamaged [45/−45/90]s tube.

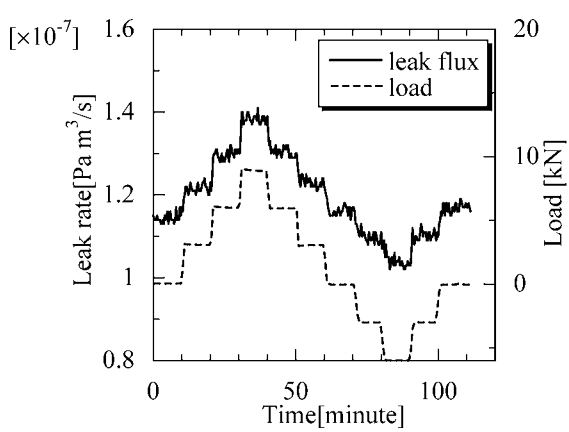


Figure 3. Relationship between applied loadings and helium leak flux of a [90_2/0/90_2] tube.

3.1. Effect of loads on gas permeability

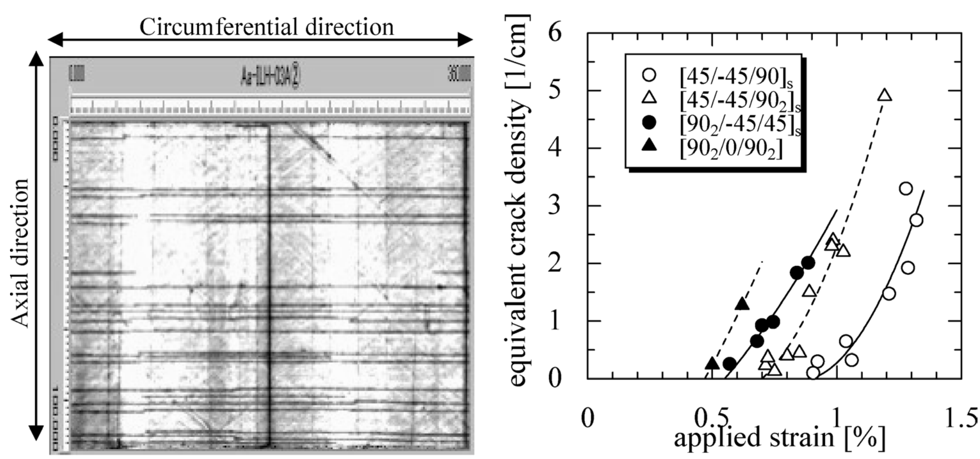
After helium was diffused through undamaged tubular specimens without loadings to the steady state, the effect of applied loadings on helium leak flux in undamaged specimens was investigated. Figure 3 shows the relationship between the applied loadings and helium leak flux of a $[90_2/0/90_2]$ tube. In conjunction with the increase in applied loads, the helium leak flux increases, whereas leak flux decreases under compressive loadings. The helium leak flux reverts to the same value when the applied loadings are removed, even though it exhibits some hysteresis. Considering the case of multi-laminar matrix cracks in CFRP laminates, which induce crucial increase (three or four orders higher) of leak flux [1, 2], the effect of applied loadings on diffusion-controlled gas permeability is negligible.

3.2. Effect of matrix cracks on gas permeability

Figure 4a shows the deployed ultrasonic image of a $[45/-45/90_2]$s specimen with matrix cracks in 90-degree layers. Matrix cracks can be clearly observed, some of which do not spread fully in the circumferential direction. In this study, equivalent crack density is defined as

$$\rho_E = \frac{\sum w_i}{LW}, \tag{4}$$

where w_i and W denote the lengths of the ith matrix crack and the tubular specimen in the circumferential direction, and L is the length of gauge section in the axial direction. In the case of $[90_2/-45/45]$s and $[90_2/0/90_2]$s specimens, observed matrix cracks are assumed to be equally distributed in two surface cracked layers. The



(a) (b)

Figure 4. Matrix cracks in tubular specimens: (a) deployed ultrasonic image of a $[45/-45/90_2]$s specimen at 0.98% strains, (b) relationship between crack densities and applied strains.

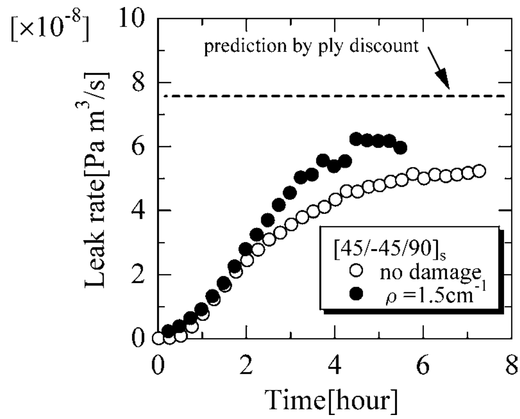


Figure 5. Comparison of helium leak flux between uncracked and cracked [45/−45/90]s tube.

relationship between measured crack densities and applied strains is summarized in Fig. 4b. It is shown that matrix cracks are susceptible to initiation in thicker 90-degree layers and in surface layers.

Measured helium flux through a [45/−45/90]s specimen with matrix cracks is compared with that through an undamaged tube in Fig. 5. Both the leak flux at steady state Q_0 and diffusion coefficient D increase on account of the existence of matrix cracks. It should be noted that the leak flux increase due to matrix cracks is lower than the predicted value using the ply discount method. Thus, the effect of matrix cracks on the gas permeability turned out not to be crucial, unless the specimens have multi-laminar cracks. This result suggests that the existence of no less than one intact layer is important for leakage prevention.

3.3. Combined effect of matrix cracks and loads on gas permeability

The helium leak flux through cracked specimens subjected to tensile or compressive loadings was measured after the steady state without loadings. The relationship between applied strains and helium leak flux of a cracked specimen is shown in Fig. 6 in comparison with the data of an undamaged specimen. The existence of matrix cracks leads to an increase in gas permeability under the same loading conditions and higher hysteresis. It is clarified that gas permeability is doubly affected due to both matrix cracks and loads. However, the effect is not crucial compared with the multi-laminar matrix cracks.

4. ANALYSIS

4.1. Theoretical basis

In order to evaluate diffusion characteristics including the combined effects of matrix cracks and loads, a diffusion model based on thermodynamics incorporated

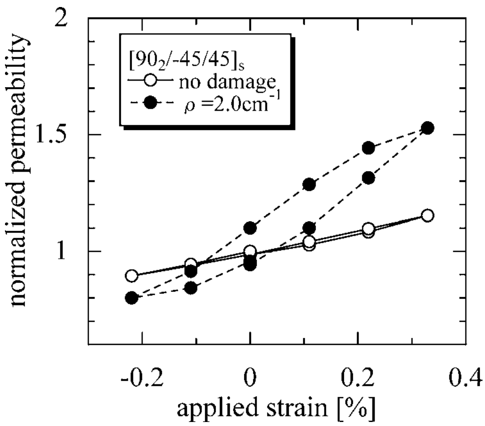


Figure 6. Relationship between applied strains and leak flux of a $[90_2/-45/45]$s tube with and without matrix cracks.

with continuum damage mechanics [3, 4] is applied to through-the-thickness gas permeability. Thus, laminates with matrix cracks in some layers are regarded as continuum materials with damage entities as internal state variables [7].

The chemical potential of gas in the laminate is given by

$$\mu = \frac{\partial G}{\partial C}, \tag{5}$$

where C is the gas concentration and G is the Gibbs free energy, which is a function of stress, damage, temperature, and C. In the absence of stress, damage, and temperature gradients, gas flux in one-dimensional term is assumed to be the form

$$Q = -D\frac{\partial \mu}{\partial x} \cong -D\frac{\partial \mu}{\partial C}\frac{\partial C}{\partial x}, \tag{6}$$

where x is the coordinate along the thickness direction. With the conservation of diffusing mass, these equations give the following governing equation.

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial \mu}{\partial C}\frac{\partial C}{\partial x}\right). \tag{7}$$

The Gibbs free energy for solids of orthotropic symmetry can be expanded as a polynomial function of invariant variables given by [3, 4, 7]. Also, damage entities can be expressed as average variables in tensorial form related to the displacement and normal vectors of damages [7]. On the assumption that matrix cracks occupy the whole thickness and the whole length along fibers of cracked layers, as well as appear with regular spacings and affect the overall response only by normal displacements of the crack surfaces, the damage tensor can be expressed as [7]

$$D_{ij} = \frac{a t_c \rho}{h \sin\theta} n_i n_j, \tag{8}$$

where a is average crack surface displacement, n_i is unit normal vector of crack surfaces, t_c is the thickness of cracked plies, and θ is the fiber angle of cracked layers in the global sense.

In the case of a laminate with matrix cracks only in 90-degree layers subjected to uniaxial loadings in the 0-degree direction, the only non-zero component of D_{ij} denoted as d is given by

$$D_{11} = d = \frac{at_c\rho}{h}, \tag{9}$$

and the Gibbs free energy can be expressed in the polynomial of applied stress σ and d [3]. In this study, axial strain ε is used instead of σ. Thus, the polynomial is restricted to quadratic terms of ε and d, as $\varepsilon, d \ll 1$. With α_i and β_i as the functions of temperature and gas concentration, the polynomial can be expressed as

$$G = \left(\alpha_0 + \alpha_1\varepsilon + \alpha_2\varepsilon^2\right)\left(\beta_0 + \beta_1 d + \beta_2 d^2\right). \tag{10}$$

From equations (7) and (10), the diffusion equation including the effects of matrix cracks and loads can be obtained, which is nearly identical to Fick's law but has a different diffusion coefficient.

For cracked laminates without loadings,

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(\bar{D}\frac{\partial C}{\partial x}\right), \quad \bar{D} = D_0 + D_2 d + D_5 d^2, \tag{11}$$

and for undamaged laminates subjected to uniaxial loadings,

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(\bar{D}\frac{\partial C}{\partial x}\right), \quad \bar{D} = D_0 + D_1\varepsilon + D_3\varepsilon^2, \tag{12}$$

are derived. Also, the expression including the combined effects of damage and loads can be obtained similarly.

$$\bar{D} = D_0 + D_1\varepsilon + D_2 d + D_3\varepsilon^2 + D_4\varepsilon d + D_5 d^2 + D_6\varepsilon^2 d + D_7\varepsilon d^2 + D_8\varepsilon^2 d^2. \tag{13}$$

The coefficients D_i are functions of temperature and gas concentration and the coincidence of D_i in equations (11) to (13) is valid only under the condition of uniform distribution and concentration-independence of stress, damage and temperature.

4.2. Application to experimental results

Equations (11) to (13) are applied to the measured diffusion coefficients. For load-dependent permeability, only the data of saturated helium leak flux was obtained in this experiment. Thus, it is assumed that gas solubility is independent of loads and saturated flux is proportional to the gas diffusion coefficient.

In this analysis, the damage parameter expressed as equation (9) is applied with $\rho = \rho_E$, which is defined in equation (4). For laminates with surface cracked layers, the sum of ρ_E in each cracked layers is used. Although the crack surface

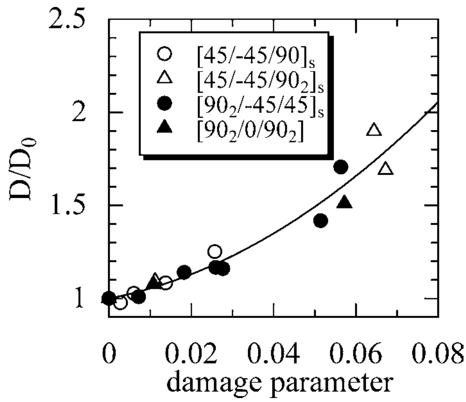


Figure 7. Relationship between normalized diffusion coefficients and damage parameters of cracked laminates without loadings.

displacement a in equation (9) can be determined if the solution to the associated problem of crack surface displacements can be solved, it is assumed that a is proportional to the thickness of cracked layers but the proportional coefficient in the surface cracked layers is twice as high as that in inner cracked layers. Thus, for tractability, $a = kt_c$ is used with $k = 1$ for inner cracked layers whereas $a = 2kt_c$ is used for surface cracked layers.

Figure 7 shows the relationship between the damage parameter defined in equation (9) and the diffusion coefficients, which are normalized with that of undamaged laminates without loadings. For all data of four layups, the identical fitting curve can be obtained. This curve corresponds to equation (11) and is expressed as

$$\frac{\bar{D}}{D_0} = 1 + 4.28d + 1.11 \times 10^2 d^2. \tag{14}$$

Also, for equations (12) and (13), the experimental results can be analogously fitted. It is concluded that through-the-thickness diffusion-controlled gas permeability, including the combined effects of matrix cracks and loads, can be evaluated using this analytical method based on thermodynamics and continuum damage mechanics.

5. CONCLUSIONS

Diffusion-controlled gas permeability through CFRP tubular specimens was experimentally investigated at room temperature as fundamental research of the applicability of composites to propellant tanks. It was shown that the existence of matrix cracks and loads affects the gas permeability. A diffusion model based on thermodynamics and continuum damage mechanics was applied to the experimentally obtained diffusion coefficients, which revealed that gas permeability, including the combined effects of damages and loads, can be evaluated using this model.

However, these effects turned out not to be crucial in comparison with the leakage through multi-laminar matrix cracks, which is three or four orders higher than the diffusion-controlled permeation. This suggests that the existence of no less than one intact layer is important for the suppression of crucial propellant leakage. Key issues for the feasibility of composite propellant tanks were obtained.

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