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Analysis on the Elasto-Plastic Peel Test in a Cu/Cr/Polyimide System

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Cu/Cr/Polyimide 계의 탄-소성 필 테스트에 대한 해석

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초 록 Cu/Cr/polyimide 계에서 금속박막 두께와 폴리이미드 표면의 플라즈마 전처리 조건에 따른 필 테스트 결과로부터 Park과 Yu[1]의 X-선 측정에 의한 방법과 Moidu 등[2,3]의 이론적 방법을 통해 Cr/polyimide 계면균열의 계면파괴에너지를 구했다. 두 방법으로 구한 박막의 소성일과 계면파괴에너지는 대부분의 경우에 대해 서로 잘 일치하였으며, 이와 같은 실험적 방법과 이론적 방법 모두 계면파괴에너지의 측정에 유용함을 알 수 있었다. 계면파괴에너지는 박막 두께에 거의 무관하였으며, 0.03, 0.036 그리고 0.05 W/cm²의 rf 플라즈마 밀도에 대해 각각 46.8±17.8, 170.3±42.9 그리고 253.9±44.4 J/m²의 계면파괴에너지를 얻었다.

Abstract The interfacial fracture energies, Γ, of the Cu/Cr/polyimide system were deduced under varying Cu film thickness and pretreatment conditions based on two methods; X-ray measurement by Park and Yu[1] and theoretical methods by Moidu et al..[2,3] The two methods showed reasonable agreement for most cases, imparting validity for both approaches. Estimated Γ were quite independent of the metal film thickness and increased with the rf plasma power density of polyimide pretreatment as expected. Estimated values of Γ were 46.8±17.8, 170.3±42.9 and 253.9±44.4 J/m² for the rf plasma power density of 0.03, 0.036 and 0.05 W/cm², respectively.

1. Introduction

Peel tests have been widely used to measure the adhesion strength of various materials with thin films. During the peel test extensive plastic deformation occurs, and the peel strength does not represent the interfacial fracture energy because the measured value includes not only the debonding energy of the interface but also the work expenditure during peeling.[1~8]

Since the peel strength, P, is the sum of the interfacial fracture energy, Γ, and the work expenditure during the peel test, Ψ, estimates of Γ were made by subtracting Ψ from the measured values of P. Theoretical analysis by Kim and Aravas[4] highlighted the importance of the plasticity effect and showed that the peel strength depends sensitively on the thickness and yield stress of the film. However, values of the maximum curvature, K_B, or base angle, θ_B, which are critical to calculating Ψ are generally difficult to know. Improvements to the theory were made by Kinloch et al.[5] and Moidu et al.[2,3] who modeled the attached part of flexible adherend as an elasto-plastic beam on an elastic foundation[9~15] and presented a method to calculate θ_B and ultimately Ψ numerically. On the experimental side, Park and Yu[1] made an X-ray analysis of the mechanical effects of the peel test in a Cu/Cr/polyimide system. Through a careful calibration of the X-ray peak broadening, plastic strain in peeled metal films was measured, and the maximum curvature was deduced. The estimated work expenditure was close to measured peel strength for most cases, which indicated that the measured peel strength was more a measure of the plastic deformation during peel test than a measure of the true interfacial fracture energy.

In the present analysis, the interfacial fracture energy of a Cu/Cr/polyimide system is deduced at varying metal layer thickness and the pretreatment conditions of polyimide by using two different methods; one based on the X-ray measurements by Park and Yu[1] and the other on the theoretical method by Moidu et al..[2,3] Then, the two methods were compared each other and the effects of metal film thickness and the pretreatment conditions were discussed.

2. Procedures to estimate the interfacial fracture energy

2.1 Experimental Method

The 90° peel test were conducted using multi-films/alumina substrate system, and the film structure consists of electroplated Cu/sputtered Cu/sputtered Cr/spin coated polyimide/sputtered Cr in the order of increasing proximity to the alumina substrate as shown in Fig. 1. It was the electroplated Cu layer whose thickness was varied, and it was near the Cr/polyimide interface where cracking occurred. The Cr layer was introduced to enhance the adhesion of the Cu/polyimide and polyimide/alumina interfaces which exhibit intrinsically low adhesive strength. Copper layers of varying thickness (electroplated Cu plus sputtered Cu) were prepared, and the polyimide surfaces were pretreated by Ar^+ rf plasma under varying plasma density before the subsequent Cr deposition onto polyimide. Uniaxial tension tests were conducted with specimens of electroplated Cu/sputtered Cu at all thickness levels, and the resultant stress-strain curves (cf. Fig. 4 of reference 1) were fit either to the elastic-perfectly plastic[4] or bilinear hardening constitutive relations.[5] The peel strength data of Cu/Cr/polyimide specimens under varying Cu film thickness, t, and rf plasma density, ρ, are presented in Fig. 2.[1]

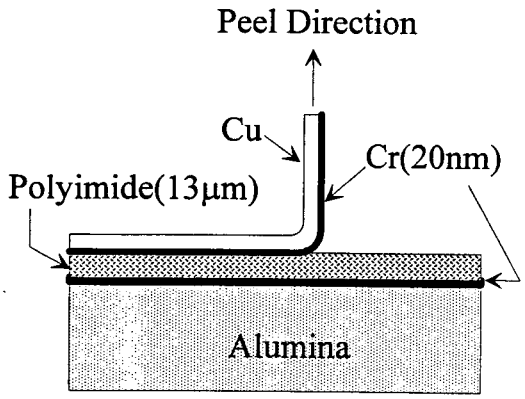

Fig. 1. A schematic diagram showing the 90° peel test and the multi-layered film structure.

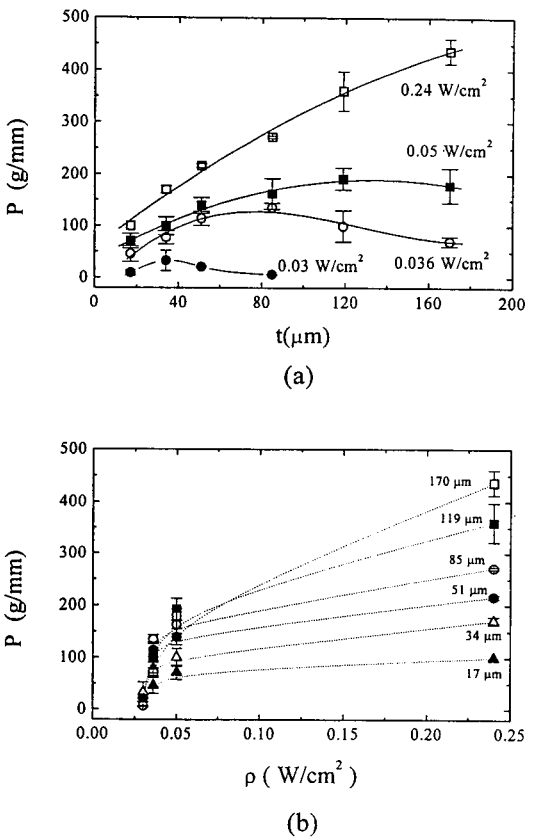

Fig. 2. The peel strength P as a function of (a) the metal layer thickness t for various pretreatment conditions, and (b) the rf plasma power density ρ for various metal layer thickness levels.

Work expenditure, Ψ, during the peel test was estimated from the X-ray peak broadening measurement of peeled metal films. The full width at half maximum, FWHM, of the Cu (331) peak was measured before and after the peel test, and the plastic strain in peeled films was found from a calibration plot relating the shift in FWHM to those of uniaxial tension specimens. From the measurement of the plastic strain, the maximum curvature K_B was found as in eq. (10) of reference 1, from which Ψ was calculated.

For an elastic-perfectly plastic material, the work expenditure of the adherend metal film due to bending is given by

$$\Psi = M_0 K_e \left[2\frac{K_B}{K_e} + \frac{10}{3}\frac{K_e}{K_B} - 5 \right] \quad (1)$$

where K_B is the maximum curvature in the moment-curvature curve which occurs at the crack tip, M_0 is the fully plastic moment, and K_e is the elastic limit curvature defined in eq. (8) of reference 1.

Similarly, the work expenditure for a bilinear hardening material is given by

$$\Psi = M_0 K_e \left[\frac{2K_e(1-\alpha)}{3K_B(1-2\alpha)} (5-14\alpha+12\alpha^2-4\alpha^3) - (1-\alpha)(1+4(1-\alpha)^2) + 2(1-\alpha)^2(1-2\alpha)\frac{K_B}{K_e} + \frac{4}{3}\frac{K_B^2}{K_e^2}\alpha(1-\alpha)^2 \right] \quad (2)$$

where α is the work-hardening parameter defined in reference 5.

Once Ψ was estimated, the interfacial fracture energy Γ could be deduced from the energy balance equa-

tion[4] which states that

$$P = \Gamma + \Psi \tag{3}$$

where P is the steady state peel strength and Ψ is the work expenditure per unit advance of interfacial crack per unit width.

2.2 Theoretical Method

Here, we follow the methods developed by Kinloch et al.[5] and Moidu et al.,[2,3] and numerically calculate the maximum curvature K_B and the base angle, θ_B according to the following procedure;

(a) If the detached part of an adherend is treated as a slender beam,[4] conditions of force and moment equilibrium become;

$$P(1-\sin\theta_B) = K_e M_0\left[\frac{8}{3}\frac{K_e}{K_B} - 4 + 2\frac{K_B}{K_e}\right] \tag{4}$$

for an elastic-perfectly plastic material and

$$P(1-\sin\theta_B) = K_e M_0\left[\frac{8}{3}\frac{(1-\alpha)^4}{(1-2\alpha)k_B} - 4(1-\alpha)^3 + 2(1-\alpha)^2(1-2\alpha)k_B + \frac{\alpha}{3}k_B^2(1+4(1-\alpha)^2)\right] \tag{5}$$

for a bilinear hardening material.

(b) If the attached part of an adherend is treated as an elasto-plastic beam on an elastic foundation,[2,3] a nonlinear equation involving two unknown parameters K_B and θ_B follows from the deflection equation of the attached part of adherend and a set of boundary conditions (cf. equations (28) - (34) of reference 2).

Then, the two equations with two unknowns (K_B and θ_B), one from the force and moment equilibrium and the other from the deflection equation, can be solved numerically, from which Ψ and Γ can be estimated as in eqs. (1) - (3).

3. Results and discussion

The peel strength data of Park and Yu[1] are presented here as a function of the metal film thickness t and the rf plasma density ρ in Fig. 2. Note that the peel strength was strongly affected by the thickness of metal film and the pretreatment condition of polyimide in a synergistic way, and that the peel strength showed a maximum at an intermediate thickness level.

In Fig. 3, estimated Ψ values of an elastic-perfectly plastic and a bilinear hardening material are shown for a specimen treated under the plasma power density of

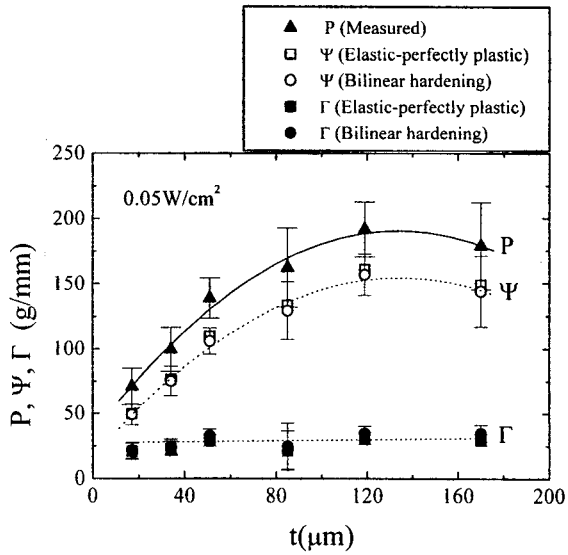

Fig. 3. Measured peel strength P and estimated work expenditure Ψ from numerical calculation for the elastic-perfectly plastic and bilinear hardening materials. The peel strength data were from specimens treated under 0.05W/cm², and Γ was the subtraction of Ψ from P.

0.05 W/cm². As expected from the stress-strain curves of copper films (cf. Fig. 4 of reference 1), the two constitutive models (cf. eqs. (1) and (2)) differed very little in Ψ and the same was true of Γ which is the balance between the measured peel strength and the estimated work expenditure. This provides a basis for choosing only the elastic-perfectly plastic material behavior and neglecting the bilinear hardening behavior in the following analysis for the sake of simplicity. It is interesting to note that the interfacial fracture energy Γ was around 25 ~ 30 g/mm for the pretreatment plasma power density of 0.05W/cm² and virtually independent of the metal film thickness. Considering the large variations of measured P and the errors involved in the numerical method, this is a remarkable result because the Cr/polyimide interface energy should be independent of the film thickness.[16]

In Fig. 4 (a), values of the work expenditure deduced from the X-ray measurements and from purely numerical calculations are compared under varying metal film thickness and plasma treatment conditions. The Ψ values obtained by the two methods agreed reasonably well for most cases (except for the 85 and 119㎛ samples treated under 0.036 W/cm²) suggesting the validity of the numerical method by Moidu et al..[2,3] If errors in X-ray measurements or constitutive relations can be corrected, discrepancies between the two methods are expected to be smaller.

Interfacial fracture energy was estimated by sub-

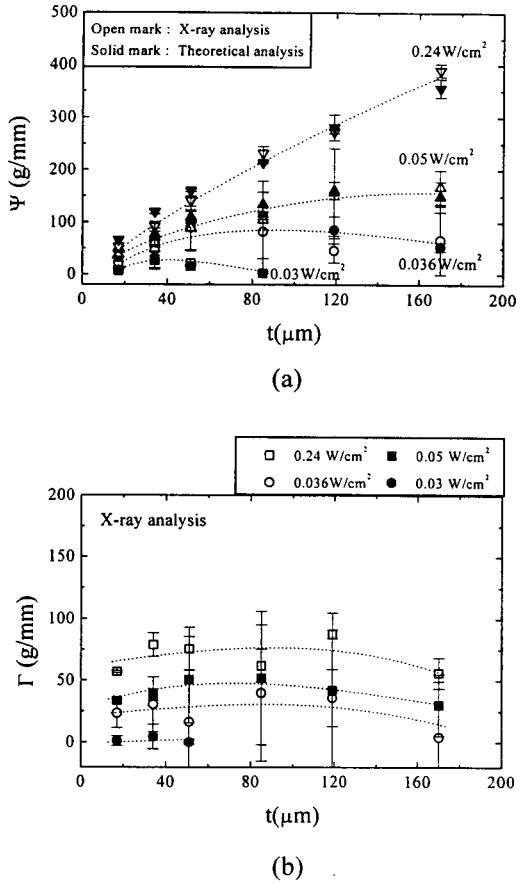

Fig. 4. (a) Work expenditure Ψ estimated from the X-ray and the theoretical analyses and (b) the interfacial fracture energy Γ from the X-ray analysis as a function of the metal layer thickness.

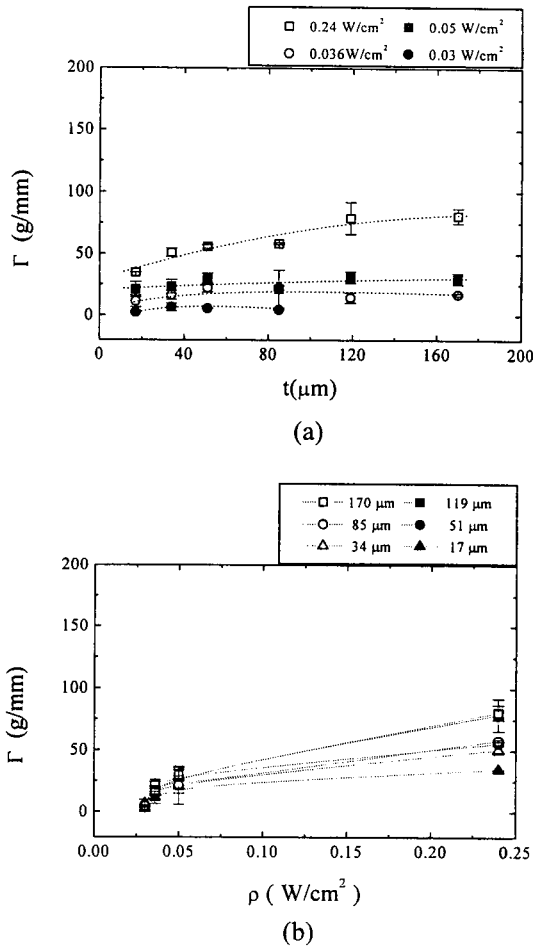

Fig. 5. The interfacial fracture energy Γ from the theoretical analysis as a function of (a) the metal layer thickness and (b) the rf plasma density.

tracting Ψ from the peel strength, and Γ values were deduced from the X-ray measurements and from the numerical calculations. Even though values of Γ derived from the experiment show substantial scattering over the film thickness for most cases except the 0.03 W/cm² data as shown in Fig. 4 (b), it can be generally stated that Γ values fall within a certain range despite widely varying metal film thickness and corresponding peel strengths. For example, specimens treated under the plasma density of 0.24 W/cm² show Γ values of 63 ±20 g/mm even though P varies from 100 to 450 g/mm. Note that Γ accounts for 10 ~ 55 % of P for those samples leaving the rest to the work expenditure during the peel test. On the other hand, scatter bands in Γ values derived from the numerical calculations were quite smaller except for the specimens treated under 0.24 W/cm² with typical interfacial fracture energies of 4.8±1.8, 17.4±4.4 and 25.9±4.5 g/mm for the plasma treatments of 0.03, 0.036 and 0.05 W/cm², respectively as shown in Fig. 5 (a). Considering the large variations in peel strengths shown in Fig. 2 (a) and general agreement of Γ values deduced from the experiment and theory, we believe that the interfacial fracture energy is independent of the film thickness, per se. Since the work expenditure of eqs. (1) and (2) includes the plastic dissipation of the adherend due to bending only, the near tip plasticity is included in Γ. Therefore, nature of the crack tip singular fields and the measure of the mode mixity, the phase angle, seem to be quite independent of the film thickness. The reason why numerically deduced Γ increases gradually with the film thickness (Fig. 5 (a)) while experimentally deduced Γ does not (Fig. 4 (b)) for the case of 0.24 W/cm² specimens is not clear at the moment. Possibilities include inaccurate description of the real stress-strain curve by the elastic-perfectly plastic curve, etc.

Variations of numerically calculated Γ with the rf plasma power density ρ are shown in Fig. 5 (b). As in the case of the peel strength (Fig. 2 (b)), the interfacial fracture energy increases with ρ with a rate that increases with the film thickness. Pretreating the polyim-

ide surface by rf plasma before the sputter deposition of Cr is known to increase the peel strength by activating the C and/or O bonds of polyimide to form more carbide-like or oxide bonds with Cr.[17,18] In fact, it is the Cr /polyimide interface energy and Γ which are most directly affected by the pretreatment as shown in Fig. 2 (b), and a comparison with Fig. 5(b) shows that a small change in Γ causes a large change in the peel strength. The point is well illustrated in Fig. 6 which shows correlation between the interfacial fracture energy and the peel strength. Overall, P increases with Γ ($P \approx 3.1 \sim 7.5\Gamma$) and that is directly related to the fact that the work expenditure Ψ increases with Γ. In detail, two things are noteworthy; Firstly, there is a clear thickness effect as illustrated in a group of samples with $20 < \Gamma < 25$ g/mm. For that level of Γ, peel strength increases with thickness twofold. Secondly, data belonging to pretreatment power density of 0.24 W/cm^2 show different correlation, however, P still increases with Γ ($\Delta P \approx 4.4 \sim 7.4\Delta\Gamma$) implying a systematic error to be involved in the calculation of Γ as mentioned previously.

Now, we are going to explain quantitatively the maximum peel strength phenomenon observed in Fig. 2(a) where the maximum peel strength is reached at an intermediate thickness level, t_{max}. For the sake of comparison, two pretreatment conditions; 0.24 and 0.036 W/cm^2 are taken, and quantitative explanations are presented on the basis of the $M-K$ curve. Peel strength and numerically calculated Ψ are presented in Fig. 7(a) and $M-K$ curves corresponding to arrows in the figure are presented in Fig. 7(b) and (c). It can be seen that the maximum curvature K_B is large but corresponding moment is small for thin films ($t < t_{max}$), thereby making the area under the $M-K$ curve, i.e. work expenditure Ψ, small. However, for thick films ($t > t_{max}$), the reverse is quite true due to the difficulty of bending the film, which again makes Ψ small. Therefore, maximum Ψ and the peel strength are observed at intermediate thickness, t_{max} with intermediate K_B and moment. This is consistent with the qualitative explanation by Park and Yu[1] that the crystal volume effect and the thickness effect (or compliance effect) lower the amount of plastic deformation at both ends.

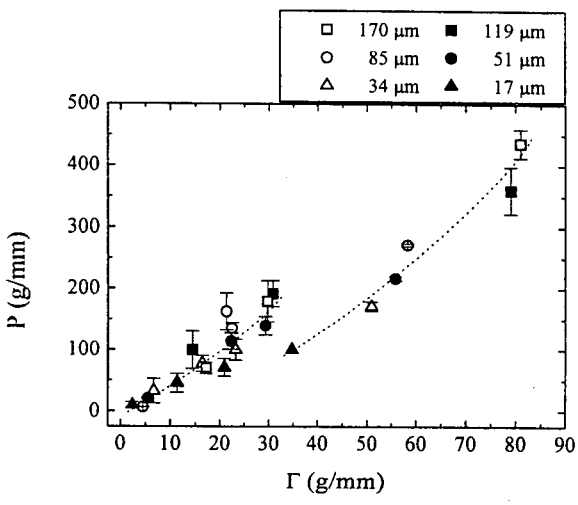

Fig. 6. Correlation between the interfacial fracture energy and the peel strength for all the specimens studied.

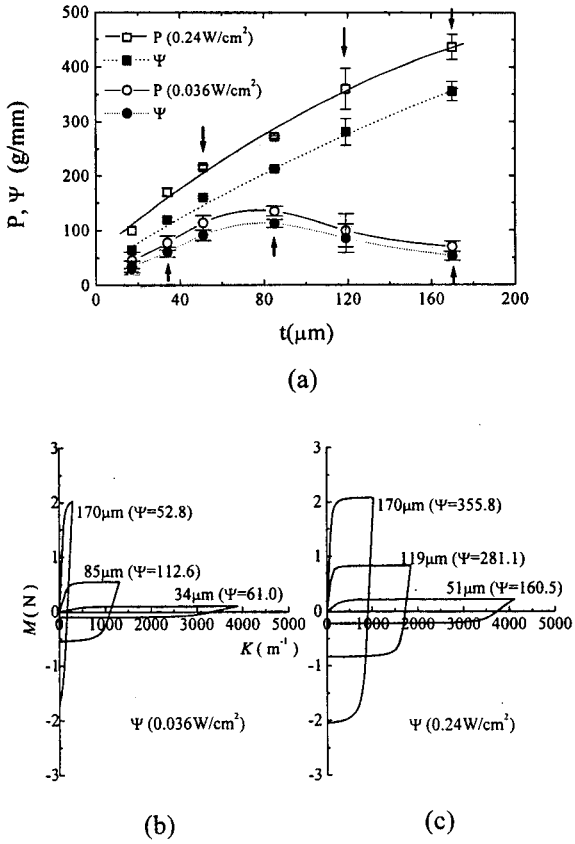

Fig. 7. (a) Variations of peel strength and work expenditure with the metal layer thickness for specimens pretreated under the rf power density of 0.036W/cm^2 and 0.24W/cm^2. Moment-curvature curves corresponding to arrows in (a) for specimens pretreated under (b) 0.036W/cm^2, and (c) 0.24W/cm^2, respectively.

4. Conclusions

1) The interfacial fracture energies of the Cu/Cr/polyimide systems are deduced by subtracting the work expenditure Ψ from the peel strengths. Two methods were used to obtain Ψ; one based on the X-ray measurement of plastic strain by Park and Yu[1] and the other based on the numerical calculation of Moidu et al..[2,3]

2) The two methods yield reasonably consistent Ψ

for most cases, imparting the validity to the experimental and theoretical methods used here.

3) Interfacial fracture energies of the Cr/polyimide interface were found to be quite independent of the film thickness. Estimated values of Γ were 46.8 ± 17.8, 170.3 ± 42.9 and 253.9 ± 44.4 J/m^2 for the rf plasma pretreatments under 0.03, 0.036 and 0.05 W/cm^2, respectively. This provides another support to the methods used here because the interface energy is independent of the film thickness, per se. The behavior of specimens treated under 0.24 W/cm^2 is not well understood yet.

4) As the rf plasma power density activating the polyimide surface was increased the interfacial fracture energy of the Cr/polyimide interface increased, which in turn increased the peel strength. Overall, the peel strength correlated well with the interfacial fracture energy showing $P \approx 3.1 \sim 7.5\Gamma$.

5) Occurrence of the maximum peel strength at some intermediate thickness levels was clearly demonstrated in M-K curves.

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