Journal of the Korean Ceramic Society
Vol. 48, No. 5, pp. 458~462, 2011.

http://dx.doi.org/10.4191/kcers.2011.48.5.458

X-ray Diffraction Analysis of Residual Stress in Laminated Ceramic

Young Ho Jin† and Dong Yoon Chung

Department of Weapons and Mechanical Engineering, Korea Military Academy, Seoul 139-799, Korea

(Received May 27, 2011; Revised July 21, July 25, 2011; Accepted July 28, 2011)

ABSTRACT

The strength of ceramic was improved by lamination by suppressing the propagation of cracks with compressive residual stress in the face layer of the laminate. Hot pressed SiAlON+SiC/SiC/SiAlON+SiC laminate discs were fabricated for tailored residual stress. The residual stress in this laminate was studied by X-ray diffraction (XRD). There was considerable compressive residual stress in the face layer. A Finite Element Analysis (FEA) was performed to support the measured XRD results and to determine the stress field in the laminate. The residual stress measured by XRD had satisfactory agreement with the analytically calculated and FEA values. The measured value by XRD was −385 ± 20 MPa over most of the face layer. The calculated and FEA values were −386 MPa and −371 MPa, respectively. FEA also showed significantly modified stresses and the maximum tensile stress near the edge region which are possible crack generators in the presence of flaws or contact damage.

Key words : Residual stress, Ceramic laminate, X-ray diffraction, Finite element analysis

1. Introduction

Ceramic materials such as carbides, oxides or nitrides have been widely used in armor applications. The fracture and pulverization of ceramic material can dissipate the kinetic energy of projectiles. The flow motion of these hard ceramic fragments surrounding the projectile abrades the tip or even its entire body. In turn, the kinetic energy is further dissipated, and the impact area is increased.

As the new generation of vehicle armor systems, laminated ceramic materials with metal cladding deserve special attention. Laminated ceramics have an advantage over former ceramic armor systems. They can suppress the crack propagation into the laminate body if there is compressive residual stress in the face layer. Residual stresses linked with external factors such as surface flaws induced by surface treatments, contact damage or external loads can be destructive or beneficial. Tensile residual stresses can expedite crack growth.[1] They induce coating failure in thin films and delamination in ceramic laminates. However, compressive residual stresses can enhance mechanical strength, if they are placed in the regions of maximum tensile stress or at the strength controlling flaws. In other words, compressive pre-stress can prevent cracking from external forces. Thus, the main motivation for producing laminated ceramics is increased apparent mechanical strength of the surface layer. Therefore, it is necessary to generate compressive residual stress in the face layer; the thermal expansion coefficient should be greater in the core material than in the face material. In elastic theory, the residual stresses in this laminate design can be calculated by equations (1) and (2) assuming they are wholly from a thermal expansion mismatch.

$$\sigma_{face} = \frac{E_{face}\Delta T}{(1-\upsilon_{face})} \times (\sigma_{face} - Z) \quad (1)$$

$$\sigma_{core} = \frac{E_{core}\Delta T}{(1-\upsilon_{core})} \times (\sigma_{core} - Z) \quad (2)$$

where Z =

$$\frac{2(1-\upsilon_{core})E_{face}\alpha_{face}t_{face} + (1-\upsilon_{face})E_{core}\alpha_{core}t_{core}}{2(1-\upsilon_{core})E_{face}t_{face} + (1-\upsilon_{face})E_{core}t_{core}}$$

υ = Poisson's ratio; α = thermal expansion coefficient; t = layer thickness, E = Young's modulus and ΔT = Temperature difference between $T_{stressfree}$ and T_{core}

The stress state in hot-pressed 75% SiAlON+25% SiC/SiC/75% SiAlON+25% SiC 1-2" diameter laminate discs were studied by X-ray diffraction. Then Finite Element Analysis (FEA) for this laminate system was performed to complement the X-ray diffraction stress measurements.

2. Experimental

2.1. Specimen preparation

α-SiAlON and SiC starting powders were obtained from Kennametal and hot-pressed under a pressure of 28 MPa after holding one hour at 1900°C in a graphite die to prepare the specimens for the elastic properties and microstructural analysis.[2] SiAlON and SiC were co-sintered; liquid phase sintering aids(Yb_2O_3) were used to reduce the sintering temperature of SiC. The surfaces of the hot pressed samples

†Corresponding author : Young Ho Jin
E-mail : jyh03@kma.ac.kr
Tel : +82-2-2197-2966 Fax : +82-2-977-5317

Fig. 1. Laminated ceramic disc design.

were ground and polished to 0.5 m for the X-ray and microscopic analyses. For a stress-relieved reference sample, the hot-pressed specimens were crushed and ground to 30-40 m in a mortar. The laminated ceramic disc design is shown in Fig. 1. The co-sintered SiAlON/SiC/SiAlON discs were easily cracked due to a large thermal expansion mismatch between the face and core layer. To reduce the thermal expansion mismatch, a composite material (75 volume% of SiAlON + 25 volume% of SiC) was used as the face layer, resulting in uncracked specimens. Total thickness and diameter of the laminate disc were 12 mm and 25.4 mm respectively.

2.2. Characterization

Various mechanical properties of the specimens were measured for the X-ray stress analysis and were also used as input data in the FEA. The elastic properties vary with temperature but in the stress calculation and FEA modeling, the measured values at room temperature were used.

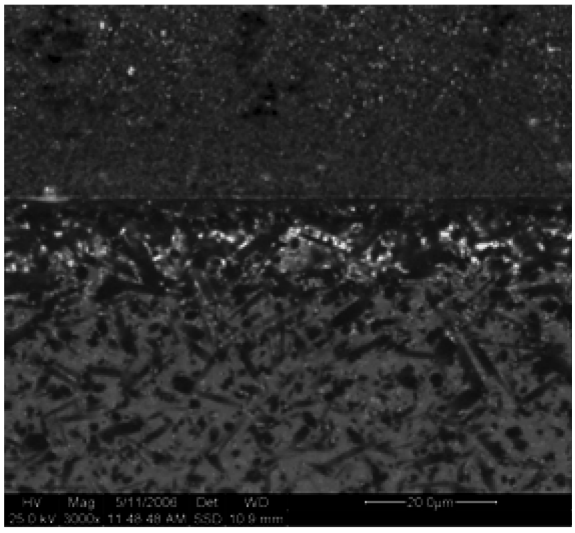
The residual stress analysis was based on structural information because it used crystallographic planes as strain gauges. The crystal and microstructures of SiAlON and SiC were characterized using X-ray diffraction (X'Pert Pro MRD, PANalytical, AlMELO, the Netherlands) and SEM (Quanta200 ESEM, FEI company, Hillsboro, OR).

3. Result and Discussion

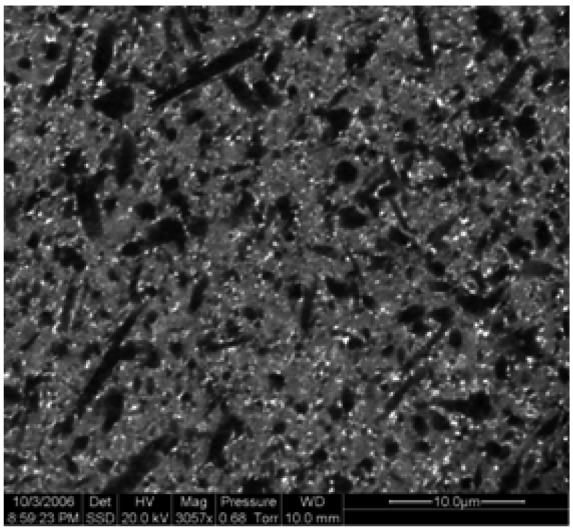
3.1. Microstructural analysis

Fig. 2(a) shows the microstructure of the sintered SiC (upper) and SiAlON (lower) laminate. The microstructure of SiAlON was composed of equiaxed α-SiAlON grains (grey) ranging in size from 0.1 μm to a few microns, and elongated β grains. Fig. 2(b) shows the microstructures of the 75% SiAlON/25% SiC composite. The SiC particle sizes in the composite are 0.1 μm to 2 μm. Elongated β-SiAlON grains (black) were detected. The SiC particles were dark and circular. It has been reported that some liquid phases assist the β transformation in samarium and ytterbium-stabilized α-SiAlON.[3,4] This suggests that a small amount of phase transformation (α → β) could have occurred during sintering.

A small amount of the glassy or Yb-rich crystalline phase was present (white). The occurrence and content of the residual glassy phases are related to the Yb content.[3] Although most of the α-SiAlON stabilizing additive in the starting powder was used for sintering of α-SiAlON, some remained as a part



(a)



(b)

Fig. 2. Back-scattering SEM micrographs of samples, (a) side and (b) surface view.

of a residual intergranular glassy phase and can degrade the mechanical and chemical properties of the material above the glass-softening temperature, normally 900-1100°C[6]. This affects the stress free temperature in laminates and limits the overall magnitude of residual stress.

3.2. XRD reflections

When a crystallite is compressively contained, the interplanar spacing (d-spacing) is decreased unless the plane is parallel to the stress axis. XRD can measure the lattice strain using d-spacing as internal strain gauge since the peak position shifts toward a higher 2θ direction in the presence of compressive stress.

X-ray diffraction patterns for the hot-pressed SiAlON and

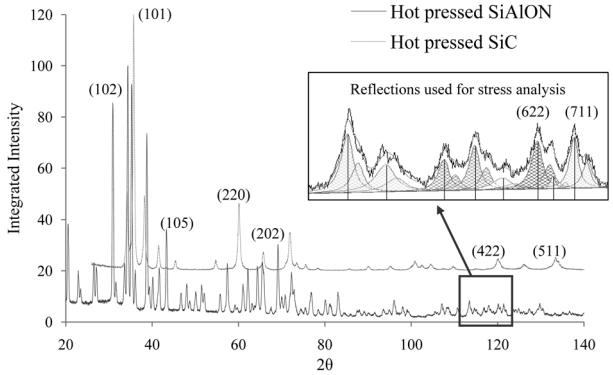


Fig. 3. X-ray whole pattern of Hot-Pressed SiAlON and SiC.

SiC are shown in Fig. 3. From an inspection of the X-ray diffraction patterns, two 2θ angle reflections, (622) and (711), were chosen for the stress analysis because of their high intensities and less overlap with other reflections. In general, high 2θ (above 120°) angle reflections are used in stress analysis, because peak shifts are more sensitive to applied stress at higher 2θ.[6)]

3.3. X-ray diffraction elastic constants

The equations used in stress tensor calculation are

$$\text{Lattice Strain } \left(\varepsilon_{\phi\,\psi}^{hkl}\right) = \frac{d_{\phi\,\psi}^{khl} - d_0}{d_0}$$

$$= \frac{1}{2} S_2 \sin^2\psi[\sigma_{11}\cos^2\phi + \sigma_{12}\sin 2\phi + \sigma_{22}\sin^2\phi - \sigma_{33}] \quad (3)$$

$$+ \frac{1}{2} S_2[\sigma_{13}\cos\phi 2\psi + \sigma_{23}\sin\phi\sin 2\psi] + \varepsilon_{\phi 0}^{\text{hkl}}$$

where

$$\varepsilon_{\phi 0}^{\text{hkl}} = \frac{d_{\phi 0}^{khl} - d_0}{d_0} = \frac{1}{2} S_2[\sigma_{33}] + S_1[\sigma_{11} + \sigma_{22} + \sigma_{33}]$$

$S_1 = -\upsilon/E$, $S_2 = 2(1+\upsilon)E$ (υ = Poisson's ratio, E = Young's modulus), σ_{ij} = residual stresses, ε = measured strain, ϕ, ψ = specimen tilting and rotation angles.

If the stress state is triaxial, there are 6 unknowns in the equation (3), so at least 6 different directions in the specimen had to be x-rayed. S_1 and S_2 are called Diffraction Elastic Constants (DEC). It was necessary to get absolute distinguished from Young's modulus (E) because it is dependent on crystallographic directions. In this study they were determined for SiAlON following ASTM Standard E1426-984 and calculated from the single crystal elastic constants for SiC[7)] based on the quasi-isotropic state.[8)]

A four point bending fixture which was machined to determine DEC is described in Fig. 4.[9)] In the bending fixture, a series of loads were applied to the SiAlON slab, then the resulting lattice strains were obtained by x-raying the strained surface. The hot-pressed specimens were 60 mm × 15 mm × 2 mm in size. A screw was used as a load input device. Strain gauges were installed where the maximum tensile stress was applied.

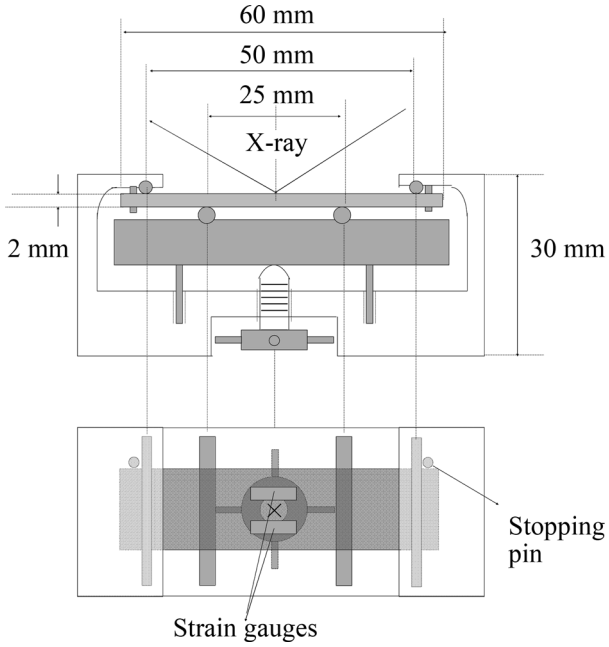


Fig. 4. Four point bending fixture.

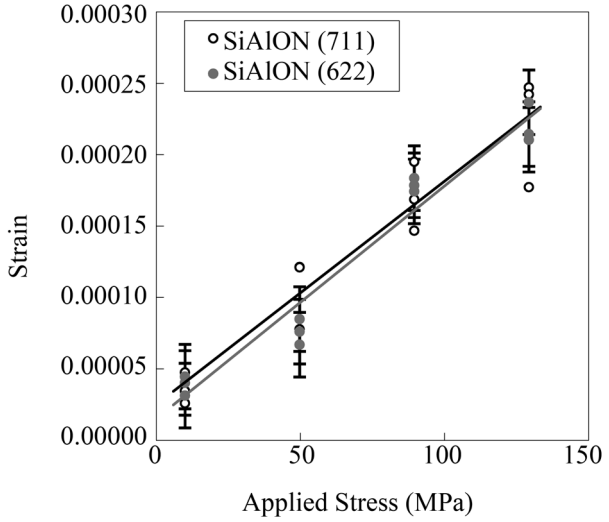


Fig. 5. Strain-stress curve for applied stress measured by XRD.

Two stopping pins held the sample at the center of the diffractometer. The measurement conditions were the same as those used for the stress measurement on the laminates. The measurements were repeated seven times. Each peak position was obtained by fitting the reflection profiles with a pseudo-Voigt function in the JADE8 computer program. The plots of strain versus applied stress are shown in Fig. 5. From the slopes of these plots, the DEC can be determined using equation (3).

3.4. Residual stress in the face layer

The conventional X-ray stress analysis was used because the surface layer was thick enough considering the X-ray depth of penetration (~62 m for SiAlON). All measurements

were carried out using a Philips X'Pert X-ray diffractometer equipped with a Cu target. The operating voltage and current were 45 kV and 40 mA, respectively. For the triaxial stress analysis, 6 different measurement directions were chosen because there are 6 unknowns in triaxial stress calculations.[10] The laminate was step-scanned at 0.02°/step intervals with a 12 s dwell time. To reduce the effect of height misalignment, surface roughness, or defocusing, a parallel beam geometry with an acceptance angle of 0.18° was used. To control the irradiated area, an X-ray lens with a point focus was chosen. The data were collected on the composite laminate over the 2 range 118.5°-123.5° for the SiAlON (622) reflection because the (622) reflection showed a better peak to noise ratio than the (711) reflection. To get sufficient data, 30 scans in 6 different ϕ directions were made, each repeated twice.

The residual stress on SiAlON can be obtained by measuring the peak shift with an increasing tilt angle. The peak positions of the SiAlON (622) reflection shifted toward higher 2θ regularly with increasing tilt angle (the specimen tilt angle was ψ). This represented the compressive residual stress in the face layer. Fig. 6 shows the residual lattice strain of the (622) reflection versus the $\sin^2\psi$ plot measured by X-ray diffraction. The residual stresses (σ_{ij}) were obtained using equation (3).

$$\sigma_{ij} = \begin{bmatrix} -402 & -7 & -35 \\ -7 & -347 & -42 \\ -35 & -42 & -40 \end{bmatrix} \pm 12 MPa$$

The stress state was biaxial which means there are only 3 unknowns in the stress equation.

The calculated stress in the face layer using equation (1) based on conventional elastic theory[2] was 386±30 MPa. There was reasonable agreement between the measured and calculated stresses.

Apparently a considerable compressive residual stress has been produced by laminating ceramic composite materials. The shear stress values can be ignored because they were small. The general stress state on the surface layer of the ceramic laminate can be described as a quasi-rotational symmetric biaxial stress state which means the stress measurement on the composite laminate requires measurements at only one ϕ rotation angle with a series of tilt angles.

The Rietveld method can be used in stress analysis later as a combination of structural refinement and residual stress analysis.

3.5. Finite element analysis (FEA)

The Finite Element Analysis (FEA) was carried out using a commercial finite element code, ANSYS to support the measured XRD results and to determine any stress gradients in the laminate. The physical properties of the materials in Table 1 were used for the modeling. The model was designed with the same geometry as the laminate to compare the modeling results with the measured stress values. Then the sintering schedule was applied to the cross-section of the laminate as the boundary conditions. Fig. 7 shows the modeled residual stress contour plot by FEA. The FEA modeled stress in the face layer (B in the Fig. 7) was 371 MPa in compression. The stresses measured by XRD, calculated from elastic theory and modeled by FEA agreed with each other.

Table 1. Properties of SiAlON and SiC

Properties	SiAlON	SiC
Density(g/cc) Room Temp.	3.405	3.255
Hardness(GPa) Room Temp.	24.8 ± 1	28.3 ± 1
Elastic Modulus(GPa) Room Temp.	302 ± 2	446 ± 3
Shear Modulus(GPa) Room Temp.	127 ± 1	189 ± 1
Poisson's Ratio Room Temp.	0.19	0.18
Thermal Expansion Room Temp. to 1900°C	4.53×10^{-6}	5.91×10^{-9}
Stress Free Temperature (°C)	1200	1300

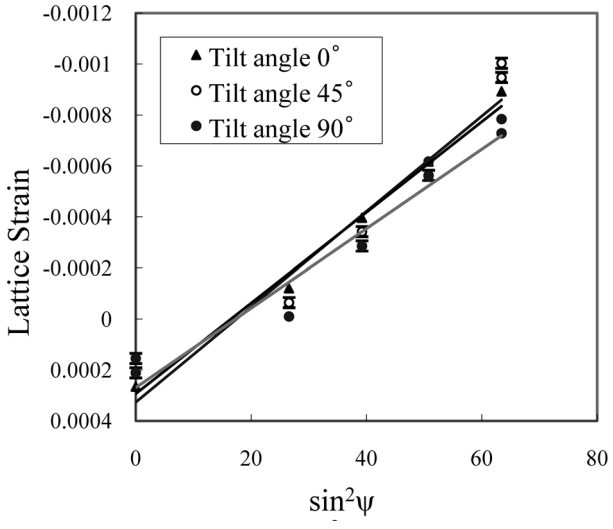


Fig. 6. Strain vs. $\sin\psi^2$ plots (Triaxial stress analysis for the SiAlON face).

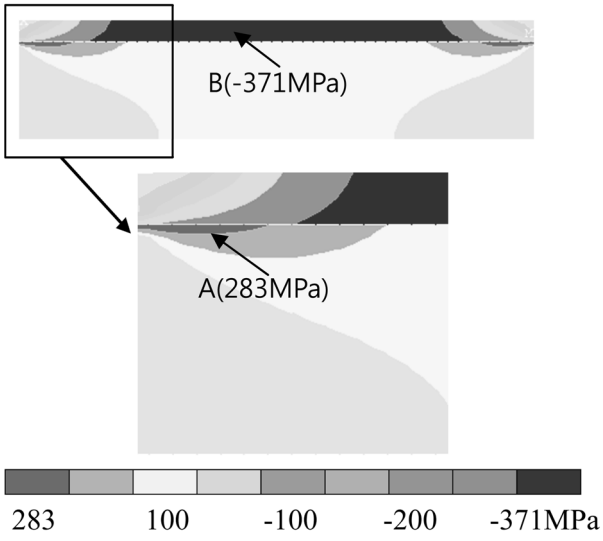


Fig. 7. Residual axial stress contour plot.

There were significant stress gradients near the edge area. The residual stress in the face layer decreased toward the edges and increased into the laminate near the free edges. Interestingly, the maximum tensile stress (283 MPa) appeared in this area (A in the Fig. 7). This stress concentration effect predisposes the laminate to delamination and makes it difficult to produce laminated materials. It is generally known that edge cracks or incidence of channeling is the "Achilles heel" of laminated materials.[10)]

The significantly modified residual stresses from their overall laminate stresses and tensile residual stress in the edge regions are the most important sources of crack propagation in the presence or possibility of cracks.

3.5. Possible applications

Ceramic laminates with compressive residual stress in the face layer can be used in ceramic composite armor systems. When a projectile hits ceramic composite armor, hydrodynamic flow and kinetic repulsion occur simultaneously[11)]. As the projectile continues to penetrate, the laminated ceramic layer breaks into hard and small ceramic fragments while abrading the projectile tip and dissipating its kinetic energy. The compressive residual stress in the laminated ceramic suppresses the cracks propagation. The backing layer which has high fracture toughness will further absorb the kinetic energy of the projectile and hold the fracture of the ceramic layer. However the tensile residual stress in the edge area might deteriorate the mechanical strength of the ceramic armor. Therefore it will be required to clad the whole laminate with metal to increase the compressive stress and to minimize the delamination occurring by edge tension.

The edge effects on the stress field and other possible factors that deteriorate the lamination effect will be verified later.

4. Conclusion

Ceramic laminates (SiAlON+SiC/SiC/SiAlON+SiC) were fabricated and the residual stresses were determined using X-ray diffraction. A considerable compressive residual stress was tailored in the face layer. Initial stress analysis was carried out for a triaxial stress state but the measurement revealed a rotational symmetric biaxial stress state in the laminated ceramics.

The measured stress values were in good agreement with the calculated and FEA values. The grain interactions could be ignored in the FEA because the measured stress values agreed with the FEA values without correcting for grain interaction effects.

REFERENCES

1. R. De Wit, "Effect of Residual Stress on Fatigue," pp. 229-244, Proceedings of the 10th Sagamore Army Materials Research Conference, New York, 1963.
2. Y. JIN, "Residual Stress Analysis in SiAlON -SiC-SiAlON Ceramic Laminate by X-ray Diffraction," pp. 31-37, MS Thesis, Pennsylvania State University, 2007.
3. Z. Shen, T. Ekström, and M Nygren, "Ytterbium-stabilized α-SiAlON Ceramics," *J. Appl. Phys*., **29** 893-904 (1996).
4. Y. J. Park and H. D. Kim, "Synthesis of SiAlON Ceramics with Novel Magnetic Properties," *J. Kor. Ceram. Soc*., **46** [5] 525-27 (2009).
5. V. A. Izhevskiy, L. A. Genova, J. C. Bressiani, and F. Aldinger, "Progress in SiAlON Ceramics," *J. Eur. Ceram. Soc*., **20** [13] 2275-95 (2000).
6. SAE international, Residual Stress Measure- ment by X-ray Diffraction; pp. 78-80, SAE International, Warrendale, PA, 2003.
7. S. Majumdar, D. Kupperman, and J. P. Singh, "Determinations of Residual Thermal Stresses in a SiC - Al_2O_3 Composite Using Neutron Diffraction," *J. Am. Ceram. Soc*., **71** [10] 858-63 (1988).
8. S. J. Skrzypek, A. Baczmanski, W. Ratuszek, and E. Kusior, "New Approach to Stress Analysis Based on Grazing-incidence X-ray Diffraction," *J. Appl. Cryst*., **34** [4] 427-43 (2001).
9. A. S. M. Y. Munsi, A. J. Waddell, and C. A. Walker, "A Method for Determining X-ray Elastic Constants for the Measurement of Residual Stress," *Strain*, **39** [1] 3-10 (2003).
10. V. Serge, D. M. Lipkin, G. D. Portu, and D. R. Clarke, "Edge stress in Alumina/Zirconia laminates," *J. Am. Ceram. Soc*., **80** [7] 1633-38 (1997).
11. S. H. Ahn, W. K. Jung, and H. J. Kim, "Development Trends of Ceramic Composite Armors for Combat Vehicles (*in Korean*)," *J. Kor. Soc. Prec. Eng.*, **22** [7] 7-18 (2005).