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Influence of Bi-Layer Structure and Sub-Layer Distance on the Hardness of Multilayer Thin Film TiAlN and CrN

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ABSTRACT

Mechanical properties are important in identifying the suitability of a material for a particular usage. One of the important properties is the hardness of the material, which can be defined as the resistance to the material to abrasion, deformation, scratching or to indentation by another hard body. Among others, this property is important for wear resistant applications. In order to obtain the mechanical characterization of thin films, apart from physical nanoindentation testing, researchers have also been using the finite-element modelling (FEM) method to simulate the nanoindentation test. In this study, a nanoindentation model of thin film CrN and TiAlN were developed and simulated to investigate the influence on the number of layers and thin film structure on the overall hardness of multilayer thin film CrN and TiAlN. A total of 10 sets of simulation was conducted with varying structural arrangement (i.e. CrN/TiAlN and TiAlN/CrN), bi-layer thickness (i.e. from 0.2 μm to 2 μm) and number of layers (i.e. 1, 2, 4, 8 and 10 layers). Based on the study, it was found that the optimum distance of sub base for multilayer TiAlN/CrN was 0.8 μm, while the optimum distance of sub-layer for CrN/TiAlN was 0 μm. It can be concluded that the type of material and the distance of sub base thin film

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layer to the maximum indenter depth will have significant influence on the overall hardness of the thin film system.

Keywords: *TiAlN/CrN, Nanoindentation, Finite Element Modelling*

Introduction

Knowledge of the mechanical properties of a thin film coating provides important information in the selection of suitable applications. This is because a thin film must have the required properties to function adequately and must be durable enough for the expected product lifetime. The most common properties considered are strength, ductility, hardness, impact resistance and fracture toughness. For a coating system, among others, hardness is one of the important criteria in mechanical properties, which is frequently used to assess the strength of a specific thin film coating.

The hardness of a coating system can be obtained through a number of test, including nanoindentation test. Apart from physically testing the thin films using nanoindentation technique, advancement in computer modelling have allowed scholars to model and simulate the hardness of various thin films. For example, Assimina and Huang [1] performed the finite element (FE) analysis simulations of nanoindentation on multilayer thin film coating by changing the ratio between the indenter's maximum displacement and thin-film thickness. The study had obtained the mechanical responses of soft film/hard substrate and hard film/soft substrate material systems, and the authors concluded that substrate properties have significant influence on the mechanical properties of the thin film. Lechinchi et al. [2] simulated Berkovich indentation experiments with the ABAQUS finite element software, and compared the simulation results with a physical nanoindentation test. The comparison between the experiment data and numerical results demonstrated that the finite element (FE) approach is capable of reproducing the loading- unloading behaviour of nanoindentation test.

Thin film coating is currently being widely used to provide extra surface protection for a number of dynamic loading tools, including bearing, cutting tools and piston rings. Nowadays, multilayer thin film coatings are usually preferred as compared to single layer thin film coating. This is because multilayer thin film coating has a number of advantages over single layers, as they combine the attractive properties of several materials, as well as exhibiting some completely new properties that are not observed with single layers. Each layer in such a multilayer structure contributes to the surface with some of its specific properties. Chromium nitride (CrN) is an example of hard coating with good tribological properties and excellent wear and corrosion resistance [3,4]. One of the main applications for CrN films is that it is used as a protective layer for tools and dies [5,6]. On the other hand, TiAlN is also an attractive thin film material due to its extreme hardness, high melting point, chemical inertness, superior oxidation resistance, and good thermodynamic stability [7]. Thus, the combination of TiAlN and CrN in a multilayer coating system should have better mechanical properties, including higher microhardness and fracture toughness, greater strength, elasticity and plasticity as compared to single layer coatings. However, there are still limited studies on the combination of thin film TiAlN and CrN in a multilayer coating system.

In this study, a nanoindentation model was developed using finite element modelling (FEM) method, involving the multilayer thin film combination of TiAlN and CrN coated on gray cast iron substrate. FEM was then further used to simulate the mechanical properties of TiAlN/CrN with various combinations of bi-layer thickness and structures to identify the mechanical behaviour within the structure. To our knowledge, there are still limited studies on the investigation of the effects of structural arrangement and bi-layer thickness on the overall mechanical properties of the coating. Thus, this current study will be able to contribute to the knowledge on the structural arrangement and bi-layer thickness of a coating system, particularly TiAlN and CrN coating, and how it will affect the overall mechanical properties of the coating system.

FEM Method

Ansys APDL Multiphysics software was used to simulate the mechanical properties of thin films for different coating system during the nanoindentation test. Figure 1 shows the flow chart of the study.

In this simulation, a 2D axial symmetry model was used to reduce computational time without compromising accuracy. For all simulations, it was assumed that the thin film is perfectly adhered to the substrate, and that the contact between the thin film and the indenter is frictionless. All the interfaces between different layers were assumed to be perfectly bonded. The indenter and the substrate were identical for all models. The indenter being used has a conical tip with the angle of 68^{o} from axial of rotation. The substrate was simulated using a rectangle of 6 μm (height) and 10 μm (witdh). The FE model used in this work is shown in Figure 2.

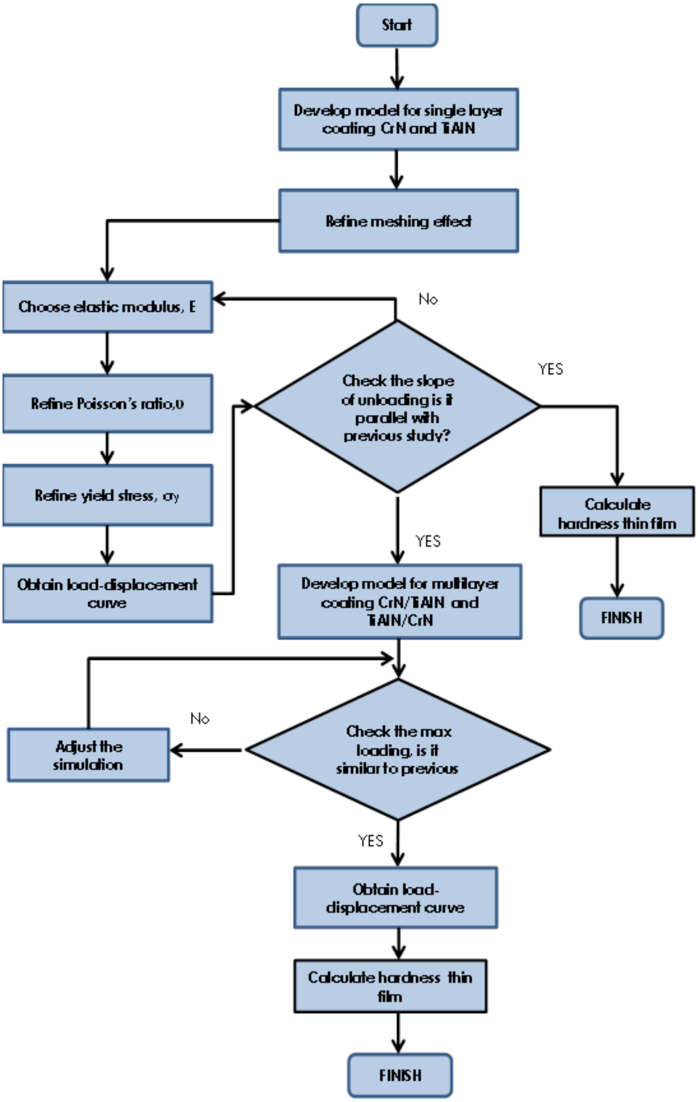

Figure 1: Flow chart of the development and simulation

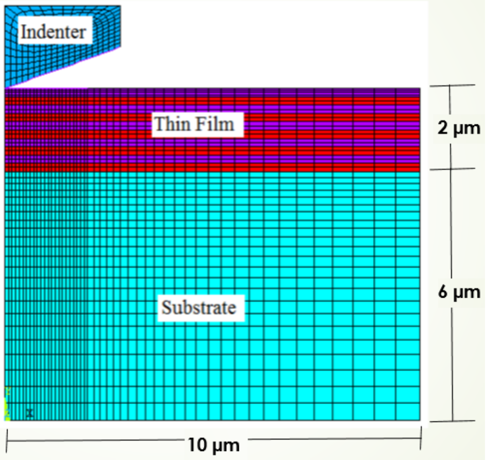

Figure 2: FE model configuration used in this study

Hardness Calculation from FEM Output

There have been a number of approaches developed to analyse nanoindentation data. By far, the approach that is most frequently used is the Oliver and Pharr method [8], which is based on the studies conducted by Doerner and Nix [9] and Sneddon [10]. In this approach, the hardness of the thin film, typically defined as the mean pressure under the indenter, is determined by using the maximum loading point of the resulting loading vs. depth of indenter curve. Equation 1 below shows the formula for calculating the hardness of the thin film based on the Oliver and Pharr method.

$$H = \frac{P_{\max}}{A} \tag{1}$$

In this equation, P_{max} is the maximum applied force and is obtained directly from the force-displacement curve, while A is the projected contact area of the indenter tip with the material. The expression for the contact area for a diamond indenter is approximated using Equation 2 below,

$$A = \pi\, h_c \tan^2 \alpha \tag{2}$$

where α is the conical angle, and h_c is the contact depth of the indenter. Equation 3 below illustrates the formula for calculating the value of h_c,

$$h_c = h_{max} - \varepsilon(h_{max} - h_f) \tag{3}$$

where h_{max} is the maximum displacement of the indenter, h_f is the final penetration depth and ε is the coefficient of the indenter, with the value of 0.72 for a conical indenter as provided by Sneddon [10]. In order to determine the mechanical response at the maximum indenter displacement, h_{max}, and also to assess the influence of substrate properties on the calculated mechanical properties of thin films, the ratio between the maximum indenter displacement and film thickness, h_{max}/t, was set to 0.05, 0.10,… up to 0.4 respectively. The resulting hardness values for the thin films will then be compared with previous literatures to ensure their validity.

Layer Structure and Arrangement

In this study, we developed the model for a 1, 2, 4, 8 and 10 number of layers with differing structure layers, which is shown in Table 1. In this case, differing structure layers means that the arrangement sub-layers are different. For example, for two number of layers, we have CrN as a sub-layer as one model and TiAlN as a sub-layer as another model. However, regardless of the number of layers and the structure of layers, the overall thickness of the thin film was preset to 2 μm. The mechanical properties of the materials used in the simulations are shown in Table 2. In order to ensure that the result from the simulation is accurate and can represent the individual thin film CrN, TiAlN and multilayer thin film CrN and TiAlN, the load vs displacement curve from the model were compared and verified with previous literatures.

Table 1: Configuration of thin film coating systems modelled in this work

Number of Layer	Structure of Layer	Thickness CrN (μm)	Thickness TiAlN (μm)
1	CrN	2	-
	TiAlN	2	-
2	CrN/TiAlN	1	1
	TiAlN/CrN	1	1
4	CrN/TiAlN/ CrN/TiAlN	0.5 x 2	0.5 x2
	TiAlN/CrN/ TiAlN/CrN	0.5 x 2	0.5 x 2
8	CrN/TiAlN/ CrN/TiAlN / CrN/TiAlN/ CrN/TiAlN	0.25 x 4	0.25 x 4
	TiAlN/CrN/ TiAlN /CrN/ TiAlN/ CrN/ TiAlN/CrN	0.25 x 4	0.25 x 4
10	CrN/TiAlN/ CrN/TiAlN / CrN/TiAlN/ CrN/TiAlN/ CrN/TiAlN/	0.2 x 5	0.2 x 5
	TiAlN/CrN/ TiAlN /CrN/ TiAlN/ CrN/ TiAlN/CrN/ TiAlN/CrN	0.2 x5	0.2 x5

Table 2: Material properties used in the finite element simulation

	Elastic Modulus (GPa)	**Poisson's Ratio**	**Yield Stress (GPa)**
Gray Cast Iron Substrate	130	0.21	0.827
CrN Thin Film	297	0.25	6.5
TiAlN Thin Film	420	0.177	14
Diamond Indenter	1141	0.07	35.7

Results and Discussion

The loading-unloading curve of each sample was simulated with a two-dimensional model. Figure 3 shows the resulting force vs displacement curve obtained from the nanoindentation simulation for single layer thin film CrN and TiAlN. Based on the figures, it can be seen that the maximum loading for TiAlN is higher as compared to the maximum loading of CrN. This means that, higher force is needed to penetrate TiAlN to the depth of 0.2 µm as compared to CrN. Due to this, the hardness for TiAlN was found to be higher as compared to CrN, as shown in Figure 4, where the value of hardness for TiAlN was found to be 31 GPa as compared to the value of 19 GPa recorded by CrN.

Table 3 and Table 4 show the difference of mechanical properties between the FEM simulation from this study to studies from the previous literatures. For the single layer, the value of hardness of TiAlN from previous literatures ranged from 28-32 GPa, and as such, the value of hardness of 31GPa from the TiAlN simulation is within the range found in previous literatures. As for CrN, the value of 19 GPa from the simulation is 10% higher than the value found in previous literature. The difference for the value of CrN may be due to the differing parameter used, such as indenter geometry, residual stresses, frictional contact between the surface and type of substrate [11]. Since the value obtained was nearly similar to the values recorded in previous literatures, it can be said that the model developed in this study can represent the material TiAlN and CrN, and can be used as basis for multilayer thin film model development.

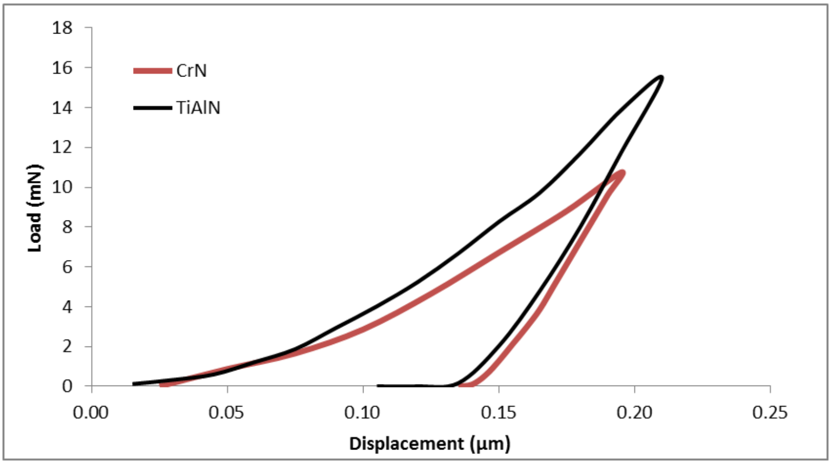

Figure 3: Loading- unloading curve for single layer thin film CrN and TiAlN

Table 3: Difference of mechanical properties between the FEM simulation from this study as compared to previous literatures single layer TiAlN thin film

Young Modulus (GPa)	Poisson's ratio	Yield stress (GPa)	Hardness (GPa)	References
420	0.177	14	31	Simulasi FE
440	0.227	NIL	29.4	[12]
315	NIL	NIL	31	[13]
385	NIL	NIL	28-32	[14]

Table 4: Difference of mechanical properties between the FEM simulation from this study as compared to previous literatures for single layer CrN thin film

Young Modulus (GPa)	Poisson's ratio	Yield stress (GPa)	Hardness (GPa)	References
297	0.25	6.5	19	Simulasi FE
380	0.22	4	NIL	[15]
341	NIL	NIL	29	[16]
250	NIL	NIL	20	[17]

Based on Figure 5, it was found that the resulting minimum hardness obtained for multilayer CrN/TiAlN and TiAlN/CrN were higher than the hardness of monolithic coating of CrN (i.e. 20 GPa) and TiAlN (i.e. 31 GPa). This shows that, the thin film at the surface of the multilayer coating system will be the dominant thin film, and the sub base will influence the hardness of the dominant thin film. In this case, the addition of TiAlN as a sub-base for CrN increased the original hardness of CrN, and the addition of CrN as a sub-layer increased the original hardness of TiAlN. The maximum hardness obtained for multilayer TiAlN/CrN and CrN/TiAlN were 35 GPa, respectively, which is similar to the value recorded by 26 GPa.

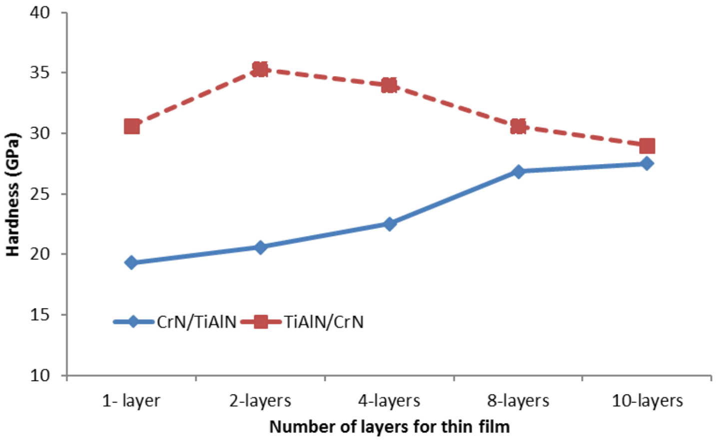

Figure 5: The value of hardness for CrN/TiAlN and TiAlN/CrN with different numbers of layers

However, while the addition of TiAlN sub layer to CrN was found to continuously increase the hardness of the multilayer thin film CrN/TiAlN, the addition of CrN as sub base only increased the hardness of multilayer thin film TiAlN/CrN up to two layers. One of the possible reasons for this is due to the significant influence exerted by TiAlN on the overall thin film system. This is because, the nearer the sub-layer is to the indenter tip, the more influence the sub-layer will exert to the coating system. In order to study this, we have analysed the thin film based on the distance of sub base to the maximum indenter depth.

Figure 6 shows the hardness of CrN/TiAlN and TiAlN/CrN with varying distances from the sub-layer to the maximum indenter depth. The curve obtained had clearly shown that, since TiAlN had higher hardness than

CrN, the nearer the distance of sub base TiAlN to the maximum indenter depth, the overall hardness of the thin film system will be higher. On the other hand, the nearer the sub base CrN to the maximum indenter depth, the lower the hardness of the multilayer thin film TiAlN/CrN.

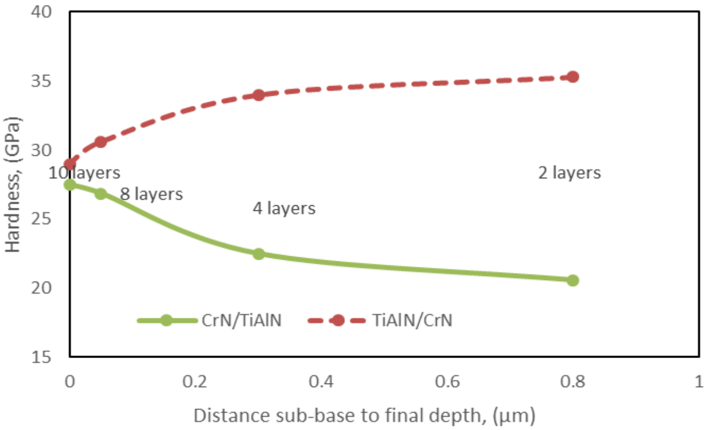

Figure 6: The value of hardness for CrN/TiAlN and TiAlN/CrN with different distance of sub-layer to the maximum indenter depth

Conclusions

In this study, we have systematically studied the influence of the number of layers and structure of layers on the overall hardness of thin film TiAlN/CrN and CrN/TiAlN using simulation of finite element (APDL). Based on the findings, it was found that the surface thin film layer will be the dominant material, and the addition of sub base will only influence the hardness of the dominant thin film layer. It has been found that, for multilayer soft/hard system, the overall hardness will increase as the sub base is nearer to the maximum indenter depth, while for hard/soft system, the overall hardness will decrease as the sub base is nearer to the maximum indenter depth. We have also found that the optimum distance of sub base for multilayer TiAlN/CrN was 0.8 μm, while the optimum distance of sub base for CrN/TiAlN was 0 μm. However, since the hardness of thin film also depends on other aspects such as stress, grain size, chemical composition, structure and deposition parameters, more work is needed to investigate the influence of these aspects on the mechanical properties of the thin film.

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