CHARACTERIZATION OF DAMAGE & OPTIMIZATION OF THIN FILM COATINGS ON DUCTILE SUBSTRATES

DISSERTATION

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* * * * *

The Ohio State University
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ABSTRACT

Coating systems provide protection to tribological components such as bearings that experience heavy contact loads from wear. In this study, hard coating-ductile substrate systems were characterized by experiments and computational models.

Tensile cracking experiments were carried out on tungsten carbide/diamond like carbon composite coatings on stainless steel substrates. Acoustic emission technique was used to monitor cracks in the coating. The fracture strain of the coating and the intercrack spacing were quantified in addition to strain corresponding to crack saturation. A 2-dimensional finite element model using cohesive zone elements was developed to predict cracking in thin film coating – interlayer – substrate systems that are subjected to tensile loading. The constitutive models were chosen to represent a metal carbide/diamond like carbon composite coating with a titanium interlayer and a steel substrate. Material properties of the coating and interlayer along with the cohesive finite element parameters were varied to study effects on stress distributions and coating cracking. Stress distributions were highly non-uniform through the coating thickness. Thus the initiation and arrest of tensile cracks differed from what is predicted by simple shear-lag theory. Inter-crack spacing distributions resulting from the variation of different parameters were quantified and compared with those from experiments.
Multilayer coatings with alternate hard and soft layers play increasingly important role because of the seemingly better performance in tribological and wear applications over monolithic coatings. However, the role of overall thickness, number of layers, and individual layer thickness cannot be overlooked and need to be optimized to minimize damage in the multilayer coatings. 2-dimensional finite element models using cohesive zone elements were developed to predict damage in multilayer coatings subject to spherical indentation. Damage in coatings was characterized as through thickness coating cracks and interfacial delamination. A design of computer experiments (DACE) approach was used to build metamodels in order to predict damage variables for a range of range design space consisting of 2, 4, 6, and 8 layers multilayer coating architecture.
DEDICATED TO MY PARENTS

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CHAPTER 1

INTRODUCTION

1.1. PROBLEM SIGNIFICANCE

Coating systems provide enabling technologies that have enhanced productivity for a wide variety of applications. One of the most effective ways to protect an engineering component operating in erosive, corrosive, or high temperature environment under heavy contact is to apply a thin, hard coating. Hard coatings are a class of coatings that have been developed as a surface engineering enhancement solution for cutting tools, dies, drills, and other tribological applications. All of these applications rely on the fact that the coatings are extremely hard, abrasion resistant, and/or provide low friction surfaces. Most hard coatings are ceramic compounds such as carbides, nitrides, ceramic alloys, cermets, and metastable materials such as diamond and cubic boron nitride. In general, hard coating properties and environmental resistance depend on composition, stoichiometry, impurities, microstructure, and texture. To effectively design coating systems for specific applications, it is necessary to know the chemical, mechanical, and tribological properties of the coatings. Thickness and layering are additional design variables for coating systems. The thickness of the coatings can vary from a few nm to 100 μm, and coatings can be single-layered or multi-layered. Figure 1.1 gives an
illustration of a typical coating-substrate system. Since hard coatings offer the “first line of defense” to the underlying substrate, it is not only important to design a coating architecture to minimize damage to the substrate but also to understand the failure mechanisms within the coating-substrate system. Figure 1.2 shows the damaged surface of a WC/DLC coating when it was subject to tensile loading.

Figure 1.1: Typial coating-interlayer-substrate system (balzers.com)

1.2. OBJECTIVE, APPROACH & SCOPE OF INVESTIGATION

The primary objective of this thesis is to investigate damage mechanisms for thin hard coatings on thick ductile substrates subject to tribological contact loading conditions. The central theme of the stated research is to predict the damage in coating given the material and geometric parameters of the coating-substrate system. Several research
studies have characterized damage via experiments. Mechanics based finite element models have also been developed to predict stresses responsible for damage initiation. However, very little research has been carried out to model damage progression in coating substrates systems. Hence, cohesive zone element finite element approach was used to model damage progression in coatings subject to loading conditions such as, tensile loading (such as seen in Figure 1.2) and spherical indentation. Also, the approach consisted of a parametric study to investigate the influence of material and process parameters in coating-substrate systems.

Figure 1.2: Parallel cracks in WC/DLC coatings subject to tensile loading. Notice the presence of maroparticles and pits on the surface of the coating.
The investigation also focused on multilayer coating architecture where the number of layers and thickness of each layer influence the performance of coatings. Various research groups have characterized multilayer coating architecture through experiments and suggested empirical optimum designs to minimize damage. Some computational models have also been developed over the years to investigate the response of multilayer coatings subject to typical loading conditions. However, there is a strong need to propose optimum designs based on the computational models to minimize damage in multilayer coatings. However, even with modern computational resources, computer modeling can be time consuming. Therefore, metamodeling techniques were applied to propose optimized multilayer architecture to minimize damage in coatings.

Defects in the form of macroparticles and pits are commonly observed in most coatings. Such defects can also be seen in Figure 1.2. The defects can alter the strength of the coating. Since the occurrence of these defects is a random phenomenon, a deterministic-probabilistic approach is needed to characterize the influence of defects. To this effect, finite element models containing defects were developed and a Monte-Carlo type approach was used to investigate the role of defects in the damage of coating systems.

The type and role of substrate though is crucial to the investigation, the substrate was mainly modeled as homogenous and “fracture-resistant”.

1.3. THESIS OVERVIEW

This dissertation has been organized into 5 chapters. Chapter 2 presents a literature review on the state of the art of coatings technology and techniques to characterize them
via experiments and numerical models. Chapter 3 and 4 are written as papers each containing its own abstract, introduction and conclusion. In Chapter 3, tensile cracking in coatings has been discussed. The chapter discusses how 2-dimensional finite element models using cohesive zone elements were used to model tensile cracking.

Chapter 4 discusses the role of multilayer coating architecture. It presents the use of 2-dimensional finite models to model 2, 4, 6, and 8 multilayer coating architecture subject to spherical indentation. The chapter also discusses the use of design of computer experiments and Kriging model to predict damage in multilayer coating architecture and obtain optimum layer thickness.

Chapter 5 summarizes the results and discusses the future direction to characterize hard coatings.
CHAPTER 2

LITERATURE REVIEW

2.1. PROCESSING AND MICROSTRUCTURE OF COATINGS

Over the years, thin film deposition techniques have been constantly evolving to produce new materials and materials structures. Thin films may be deposited by various physical vapor deposition techniques (PVD) and chemical vapor deposition (CVD) techniques. Techniques that have been successfully used to prepare high quality thin film coatings include pulsed laser ablation (PLA), filtered cathodic vacuum arc (FCVA) deposition, magnetron sputtering, RF plasma assisted chemical vapor deposition (PACVD) and mass selected ion beam (MSIB) deposition. Figure 2.1 shows a schematic of sputtering deposition chamber. Sputtering process is carried out in temperatures in the range of 200-250°C. In this deposition technique, the target coating material is subject to high negative voltages. Argon gas is introduced in the deposition chamber and the electrical gas discharge leads to the formation of positive argon ions that are accelerated in the direction of the coating material, which is atomised by the bombardment. A reactive gas such as nitrogen or acetylene is introduced and the evaporated particles of atomized metal react with the gas to form compounds and get deposited on the substrate. Typical
material systems include Titanium Nitride (TiN), chromium nitride, (CrN), and metal carbides, and diamond like carbon. Diamond like carbon (DLC), also known as tetrahedral amorphous carbon (ta−C), is an amorphous state of carbon consisting mainly of sp$^3$ carbon atoms. In recent years DLCs have attracted significant attention because they are produced in coating form at relatively low temperatures and have been found to possess high elastic modulus, hardness, atomic smoothness, and chemical inertness. These noteworthy properties originate from the fact that DLCs form a network of sp$^3$ carbon atoms that is continuous, highly oriented, and very rigid. Furthermore, properties can be tailored within a wide spectrum by controlling the sp$^3$/sp$^2$ ratio. With their diamond like properties, DLCs have tremendous potential for application in wear coatings, microelectronics, microtribology, and biomedical technology.

Figure 2.1: Deposition of coating by sputtering technique. Coutesy: Oerlikon Balzers
Vercammen et al. have investigated the mechanical and tribological properties of various state-of-the-art DLC coatings [1]. Results from Vercammen et al.’s work are summarized in Table 1. The hardness and Young’s modulus of the coatings were measured by depth sensing indentation using a NanoTest 500 with a Berkovich indenter. The microstructure, chemistry and the properties of the coatings also depend on the process parameters. For instance, the deposition of boron carbide by DC mode sputtering is influenced by parameters such as processing gas pressure, heating during deposition, bias voltage during deposition, and substrate. Various studies have been carried out on the effects of the above-mentioned parameters. Hu et al. [2], for instance, have discussed the effects of bias voltage on the microstructure and the coating properties. Knotek et al. [3] have concluded that both the heating and the applied negative bias voltage level at the substrate increase the hardness. The heating had only a slight influence, whereas the negative substrate bias voltage was established as an important factor to increase film hardness. Knotek et al. [3] concluded that the decrease of argon processing gas pressure yields higher hardness values for the coatings. The results of Knotek et al. have been summarized in Tables 2.2-2.4. The results of the influence of variation of the applied negative substrate bias voltage on the coating observed by Knotek et al. [3] are given in Figure 2.2
<table>
<thead>
<tr>
<th>No.</th>
<th>Deposition Method</th>
<th>Layer Structure</th>
<th>Thickness ($\mu$m)</th>
<th>Roughness Ra ($\mu$m)</th>
<th>Hardness (GPa)</th>
<th>Elasticity (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>RF PACVD (13.56 MHz)</td>
<td>a–C:H single layer</td>
<td>1.26±0.07</td>
<td>0.009</td>
<td>17.4±0.5</td>
<td>153±2</td>
</tr>
<tr>
<td>2</td>
<td>RF PACVD (13.56 MHz)</td>
<td>a–C:H/a–C:H Si multilayer</td>
<td>8.40±0.37</td>
<td>0.012</td>
<td>17.7±0.8</td>
<td>144±7</td>
</tr>
<tr>
<td>3</td>
<td>RF PACVD (low freq)</td>
<td>a–C:H single layer</td>
<td>1.85±0.07</td>
<td>0.018</td>
<td>19.1±1.1</td>
<td>142±4</td>
</tr>
<tr>
<td>4</td>
<td>Vacuum arc discharge</td>
<td>a–C single layer</td>
<td>0.91±0.04</td>
<td>0.075</td>
<td>28±5.8</td>
<td>238±30</td>
</tr>
<tr>
<td>5</td>
<td>Ion Beam</td>
<td>a–C:H–Si single layer</td>
<td>0.48±0.06</td>
<td>0.014</td>
<td>10.0±0.9</td>
<td>181±22</td>
</tr>
<tr>
<td>6</td>
<td>Magnetron Sputtering</td>
<td>a–C:H/a–C:H–W multilayer</td>
<td>3.36±0.21</td>
<td>0.017</td>
<td>11.5±0.5</td>
<td>139±5</td>
</tr>
<tr>
<td>7</td>
<td>Sputtering</td>
<td>C/C–Cr multilayer</td>
<td>3.23±0.03</td>
<td>0.010</td>
<td>9.8±0.3</td>
<td>119±3</td>
</tr>
</tbody>
</table>

**Table 2.1:** Mechanical and Tribological properties of various state of the art DLC coatings [1]
<table>
<thead>
<tr>
<th>Temperature(°C)</th>
<th>100</th>
<th>180</th>
<th>300</th>
<th>400</th>
<th>500</th>
</tr>
</thead>
<tbody>
<tr>
<td>Microhardness (HV0.01)</td>
<td>1950</td>
<td>1940</td>
<td>1900</td>
<td>1950</td>
<td>2260</td>
</tr>
</tbody>
</table>

**Table 2.2:** Vickers microhardness as a function of substrate temperature without substrate bias [3]

<table>
<thead>
<tr>
<th>Pressure (Pa)</th>
<th>1</th>
<th>2</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Microhardness (HV0.01)</td>
<td>2410</td>
<td>2200</td>
<td>1900</td>
</tr>
</tbody>
</table>

**Table 2.3:** Influence of argon pressure level on microhardness [3]

<table>
<thead>
<tr>
<th>Substrate Bias Voltage (V)</th>
<th>0</th>
<th>-50</th>
<th>-100</th>
</tr>
</thead>
<tbody>
<tr>
<td>Microhardness (HV0.01)</td>
<td>3080</td>
<td>3830</td>
<td>2860</td>
</tr>
</tbody>
</table>

**Table 2.4:** Influence of substrate bias voltage on microhardness at an Ar-pressure of 1 Pa [3]
In recent times nanocomposites have generated enormous interest. Nanocomposites, by definition, consist of two or more phases with at least one phase having nanometer-scale dimensions. The two phases may be any combination of ceramics, metals, and/or polymers. Several properties like modulus, tensile strength, \textit{etc.}, have been found to improve to the addition of nanometer sized phases. Nanocomposite \textit{coating} materials have recently attracted increasing interest due to the possibility of the synthesis of materials with unique properties, e.g. super hardness. Recently, a reactive

\textbf{Figure 2.2:} Substrate Bias influence on B-C coating hardness at argon pressure of 4Pa during DC deposition [3]
sputtering process, which injects small amounts of hydrocarbon gas, has been used to produce novel, thin film coatings comprised of carbide phases embedded in DLC matrices. Much of the previous work with these types of coatings has focused on titanium carbide (TiC)-DLC and tungsten carbide (WC)-DLC coatings [4-7]. These metal carbide DLC systems have shown good tribological properties and high hardness. Eckardt et al. [8], recently studied (BC)-DLC coatings on ball bearing steel (100Cr6) and high speed steel (DIN 1.3207). Coating adhesion was enhanced by a titanium interlayer. BC/DLC coating systems produced by DC magnetron sputtering with 4 different flow rates of acetylene gas have been investigated by the present author [9].

2.2. MULTILAYER COATINGS

The mismatch between the elastic properties of the substrate and the coating (characterized by Dundur’s parameters) is considered to have a role in the fracture mechanisms. Leyland, Matthews and coworkers have often emphasized the importance of high hardness to Young’s modulus ratio (H/E) of the coating to accommodate deformation [10, 11]. This has led to nanocomposite coatings with low E value combined with high hardness value.

Multilayer coatings, with alternating soft and hard layers, provide an alternative approach to accommodate deformation. Subramanian and Strafford [12] have provided a good review for multilayer coatings in tribological applications. Bouzakis et al. [13] have investigated the improvement of cutting performance of coated tools through PVD multilayer films over monolithic coatings deposited on K35 cemented carbide inserts.
The monolithic coating considered was TiAlN and the multilayer coating consisted of 10 alternating TiAlN and TiN layers. Both systems were subjected to impact testing and FEM simulations were carried out. The multilayer coatings were found to have better adhesion properties in both experimental and FEM studies. Matthews et al. [14] have investigated the role of multilayer coatings in bending deformation. They employed physical and FEM simulations for their investigation. The physical simulation consisted of bending a beam of 6 alternate layers of low and high elastic modulus metals (lead and brass, respectively). FEM was also developed for the bending experiment. The experimental and FEM results were then compared with those comprising of a beam of brass layer only. Although the simulations were highly simplified and idealized representations of multilayer coatings, it was demonstrated that due to shear deformation occurring primarily in low elastic modulus layers, higher elastic modulus layers do not experience as high bending stresses as a single layer of high elastic modulus would encounter. Djabella and Arnell [15] have presented an analysis of stresses in multilayered systems though FEM. Loading conditions of Hertzian contact in an elastic half-space by a static spherical indenter and a sliding cylindrical indenter were considered in this analysis. Four and six layer multilayer coating architectures were considered by Djabella and Arnell. It was found that stress distributions were less severe in the outer layer and at the coating-substrate interface for a six layered system compared to a four layered system. Bull and Jones [16] investigated the effect of increase in number of layers (Ti/TiN) on coating adhesion and tribological performance via scratch testing. It was concluded that multilayer coatings, in general, have improved adhesion and toughness,
leading to better wear performance over single layer TiN coatings. Voevodin et al. [17] investigated the multilayer Ti-TiN coatings on steel substrate subject to transverse bending loading conditions induced by a rigid cylindrical indenter. Models were developed and it was concluded that maximum shear and normal stress moved to the coating volume for 10 pairs of Ti/TiN layers compared to the maximum stresses under the coating-substrate interface for a single pair of Ti/TiN. Gorishnyy et al. [18] investigated the stresses in single layer, bilayer, and multilayer Cr/CrN coatings through FEM of Hertzian contacts and pin on disk experiments. It was concluded that highest adhesion and lowest wear rates were observed for multilayer Cr/CrN coatings but the performance degraded with increase in thickness of Cr layers. While Matthews and coworkers have emphasized the importance of hardness to modulus ratio (H/E) of the coatings, Suresha et al [19] argued that enhanced toughness of TiN/TiAlN multilayer coatings is not due to increase in strain capacity (H/E) of the film, but because multilayers display additional modes of plasticity leading to permanent bending and compression of the film.

It is clear from the above mentioned investigations that while multilayer coatings offer promising solutions for wear applications, demands of different applications can be met by adjusting relative thickness of different layers. For instance, Ma et al. [20] emphasized the need for optimization of Ti layer thickness for greater coating adhesion strength in TiN/Ti multilayer coatings.
2.3. DEFECTS IN THIN FILM COATING

Defects in coating in the form of macroparticles, and craters are common phenomena, seen in most PVD coatings. Persson et al. [21] classified these defects formally as: (a) Pin holes or craters, (b) Droplets, and (c) partly covered droplets. Example of such defects in PVD CrN coating as observed by Persson et al. is shown in Figure 2.3.

![Figure 2.3](image)

**Figure 2.3:** Types of defects observed in PVD CrN coatings (a) Pin hole defects, (b) Droplets, and (c) Partly covered droplets. (Observed by Persson et al. [21].)
Defects in the form of macroparticles are due to droplets incorporated during film growth and the pinholes are generated as a result of debonding of macroparticles from the coating [22-26]. For instance, Figure 2.4 illustrates the formation of pin holes or craters due to the expulsion of macroparticles during unbalanced magnetron sputter deposition of TiN and TiAIN coatings onto high speed steel.

Figure 2.4: Illustration of macroparticle expulsion causing crater formation [24]

Macroparticles can also be present in multilayer coatings and cause serious drawbacks in the tribological performance of coatings.
Figure 2.5 shows a cross sectional view of TiN/TiAlN multilayer architecture affected by droplets. Carvalho et al. [27] reported significant increase of roughness in TiN/TiAlN multilayer coatings due to the presence of macroparticles. The increase in roughness changes the real contact area leading to more friction and wear.

Matthews and Lefkow [28] reported that while macroparticles may not cause a problem in single element coatings, they cause degradation in coatings such as TiN. They reported that, when droplet-sized particles of Ti formed in the coating, they were softer than the surrounding matrix of TiN causing a detriment to the coating performance. Macroparticles are also responsible for local loss of adhesion in coatings, for instance, as reported by Munz et al. [29]. Pin holes also have a deleterious effect on the performance
of coatings as they cause exposure to the multilayer structure and substrate leading to loss in erosion and corrosion resistance. Persson et al. [21] substantiated the corrosive damage in PVD CrN coatings due to pin holes by observing macroscopic corrosion pits in the coating surface when the coating material was exposed to liquid aluminum. Their investigations concluded that liquid aluminum seeped through the pin holes which acted as channels through the coating to the substrate and in the process damaged the substrate.

There are quite a few investigations quantifying the number and size of macroparticles in PVD coatings. Keutel et al. [30] quantified the number of macroparticles of different sizes in different pulse arc deposited nitride coatings and proposed a modified deposition process. The size of droplets and number of droplets also depend on the process parameters of coating deposition. For instance, Munz et al. [25] observed that the size and number of droplets depend on the melting point of metals used during ion etching, a precleaning procedure for cathodic arc evaporation. They compared the droplet generation by different target materials, Al, Cu, TiAl, Ti, Zr, Cr, Nb and Mo and observed a general decrease of surface contamination by droplets with target materials of higher melting points. The maximum droplet size was reported to be around 20 μm and the maximum number of droplets was counted to be approximately 100,000 per square mm. There are also reports of investigations of reducing the macroparticle content in coatings. Some methods that have been proposed to overcome the droplet problem include distributed discharge arc, steered arc or arc with magnetic field filter [31]. Taki et al. [32-34] have prescribed using a shield between the cathode and the
substrate to prevent droplet adhesion. Vetter et al [35] have also found a strong correlation between the defect densities of the magnetron sputtered a-C coatings and the growth conditions during their deposition. Four main factors are believed to be influencing the defect density: (a) cleaning state of the chamber, (b) different starting temperatures before coating, (c) sputtering power, correlated directly with the growth rate and the substrate temperature, and (d) shielding of the substrates resulting in a decrease of the impingement rates of small particles at the growing surface generated during growth. The last factor is very similar to the finding of Taki et al [32], albeit the latter is concerned with coatings produced from vacuum arc deposition technique. The presence of macroparticles and pin holes result in different local coating properties, e.g., inhomogeneous stress state. Anders [36] presented a review of different filtering techniques to guide the plasma efficiently to the substrate and in the process reduce the macroparticle content. Harris et al. [37] investigated the effect of poisoning the cathode to reduce the macroparticle content and size in PVD TiN coatings. Cathode poisoning was done by increasing the partial pressures of reactive gas during operation. Zhao et al. [38] investigated the effect of negative pulsed bias on the reduced count of macroparticle count. Baouchi and Perry [39] studied the distribution of size and number of macroparticles in cathodic arc evaporated TiN films. They found that while number of macroparticles was independent of process conditions, size and shape of the macroparticles changed with process conditions.
2.4. FRACTURE IN COATINGS

Fracture in hard thin film coatings is complex and controlled by the coating material, substrate and the interface which bonds the system together. To understand the performance of the coated systems, it is critical to understand the fracture mechanisms and the sequence of fracture events during loading and unloading cycles. Hutchinson and Suo [49], and Evans and Hutchinson [50] have provided extensive discussions of the application of fracture mechanics to film-substrate composites. Hutchinson and Suo [49] have discussed the sources and effects of mode mixity in fracture of layered systems, concept of steady state cracking, and classification of crack patterns. They have described the ERR of a straight and kinked cracks, singular crack tip fields for the interface cracks, crack kinking out of the interface. They have also shown that the crack driving force is independent of crack size in multilayers when the crack grows long compared to the layer thickness. Such a crack gets arrested only when met by other crack. Evans and Hutchinson [50] formulated the condition of a crack advancement along the interface by equating the ERR of the interface crack with the interfacial fracture energy. The interface fracture energy was determined as the product of decohesion energy, a dissipative function which allows for plastic deformation, and friction along the crack faces.

A plane strain problem of an elastic thin film containing a crack oriented perpendicular to the interface was studied by Beuth [47]. The substrate was semi infinite and under residual tension. The stress intensity factor (SIF) and the steady state energy release rate (ERR) due to channeling were determined. The change in the curvature of
the coating-substrate system due to film cracking was also determined. A finite element model was developed to generalize the aforementioned results [48]. The dependence of SIF on plasticity of the substrate was studied and was concluded that yielding of the substrate increases SIF and, therefore, the likelihood of film cracking.

Thin films and multilayers fail from stresses which are too large and some typical failure modes are illustrated in Figure 2.6 [51]. Interfaces between dissimilar materials are susceptible to debonding and delamination. To a large extent, adhesion at the coating-substrate interface and toughness of the coating itself determines the durability of hard coating systems. Loss of adhesion at the coating-substrate interface leads to premature failure of otherwise wear resistant and tough coatings. The damage mechanisms are generally activated by residual stresses, both thermal and intrinsic. The deposition processes which include sputtering, spraying, spin coating, vapor deposition, etc. give rise to intrinsic stresses. Depending on the process, the deposition temperature can be low or high. The intrinsic stresses may be generated due to mechanisms like grain growth, defect annihilation, phase transition, and evaporation of a solvent. The difference in the coefficient of thermal expansion between the film and the substrate (or the different layers of the multilayer) can give rise to thermal stresses due to changes in temperature. These stresses can be of the order of several GPa for some coated systems [52].
Figure 2.6: Failure modes of coating under residual stresses: (a) delamination of coating, (b) perpendicular microcracking, and (c) bucking of coating. Source: [51]

The coating failure modes by cracking and by spalling are dependent both on the sign or magnitude of residual stresses and on the relative strengths of coating-substrate interfaces. Under compressive stresses, spallation may result either from interface cracks or by bucking and cracking of the coating [52-54]. When the film is under tension, isolated surface cracks develop from pre-existing defects in the coating and can propagate and channel across the film. These cracks generate shear stresses along the interface which may result in loss of adhesion.
The coating adhesion to the surface of the substrate is one of the most important properties in mechanical applications. The adhesion of the coating on the substrate is also influenced by the substrate roughness. The influence of roughness of substrate on TiN adhesion to steel was studied by Valli [82]. Takadoum and Benani [83] have studied the influence of substrate roughness and coating thickness on adhesion, friction, and wear of TiN films. Scratch tests were performed and it was concluded that the adhesion of TiN decreased with increase in roughness of the substrate. Majority of the work done to investigate the influence of substrate roughness on coating wear/delamination is based on scratch testing and empirical models. Approaches based on numerical modeling techniques are very few in this research field. Diao and Kandori [84], for instance, have used finite element method to study the effect of substrate roughness on delamination of hard coatings under sliding loading. Delamination maps showing the local delamination of hard coating under sliding contact were produced that show the effects of shear strength ratio of coating to substrate, the ratio of coating thickness to half contact width, and the interfacial roughness and adhesion strength.

There have been some investigations with regards to “favorable damage mechanisms” in coatings. It is believed that since strain energy stored in the coating is the driving force behind surface cracks and delamination, life of coating can be enhanced by reducing the strain energy. Although such investigations have been very limited in the area of thin film wear coatings [40], altering strain energy in thermal barrier coatings by way of introducing porosity, or periodic segmentation of coating (surface cracks normal to coating) have been reported [41-44]. Zhou and Kokini investigated the effect of the
length and density of surface precracks on delamination [45, 46]. They concluded that shorter precracks and larger pre-crack density can delay the interfacial fracture.

2.5. TENSILE CRACKING APPROACH

Scratch testing is a widely used technique for evaluating the adhesion of thin, hard coatings (e.g., [55]). Scratch testing is useful to compare the adherence of similar coating-substrate systems, however, it fails to quantify parameters like interfacial strength between the coating and the substrate and cohesive strength of the coating. A novel method for evaluating adhesion strength was proposed by Agrawal and Raj [56]. This “tensile cracking” approach evaluates both the cohesive strength of the coating and the interfacial adhesion strength between the coating and the substrate. The approach is based on subjecting a brittle coating on a ductile substrate to tension and thereby propagating coating cracks that are oriented transverse to the tensile direction. A micrograph showing transverse cracks in a tungsten carbide/DLC (WC-DLC) coating on a stainless steel tensile specimen is shown in Figure 2.7.

The crack density increases with substrate tension until the crack spacing reaches a saturation value when it is no longer influenced by the increase in substrate tension. As given in the following equation, the model relates \( \hat{\tau} \), the interfacial shear strength between the coating and the substrate to \( \hat{\sigma} \), the tensile fracture strength of the film, \( \hat{\lambda} \), the characteristic saturation crack spacing, and \( \delta \), the coating thickness [56]:

\[
\hat{\tau} = \frac{\pi \delta \hat{\sigma}}{\hat{\lambda}}. \tag{2.1}
\]

For a highly elastic coating, the tensile strength of the coating, \( \hat{\sigma} \) can be written as
\[ \hat{\sigma} = E \hat{\varepsilon}_f , \]  

(2.2)

where, \( E \) is the modulus of elasticity of the coating and \( \varepsilon_f \) is the strain corresponding to the onset of cracking in the coating. Agrawal and Raj proposed the full sine wave function shown in Figure 2.8a for approximating the interfacial shear stress. This approximation results in zero interfacial shear stress values at the midpoint and at both ends of the inter-crack spacing in the coating. Other research groups have asserted that, as shown in Figure 4b, the interfacial shear stress should have its maximum value located at each end of the inter-crack spacing and be zero at the midpoint of the inter-crack spacing in the coating [57-60]. The analytical work of Yanaka et al. [60] and Wojciechowski and Mendolia [59] and the experiments and FEA of Chen et al. [57, 58] indicate that the interfacial shear stress distribution is best approximated by a distribution like the one shown in Figure 2.8b. The previous work with finite element analysis of tensile cracking assumes preexisting cracks by introducing fine notches in the model [57-61]. Few researchers have developed finite element models which can simulate the formation and propagation of tensile cracks and thus predict coating performance.
**Figure 2.7:** A WC-DLC coating pulled to 4% strain resulting in longitudinal cracks. Tensile loading direction is perpendicular (horizontal in the figure) to the longitudinal

**Figure 2.8:** Two different theories predicting the distribution of interfacial shear stresses along the coating-interlayer interface between two cracks when the coating-interlayer-substrate system is subject to tensile loads (a) sinusoidal distribution with zero stress at the endpoints [56] and (b) maximum stress at the endpoints [57, 59, 60]
2.6. INDENTATION APPROACH

Instrumented indentation is continuous measurement of the load versus displacement. Since instrumented indentation needs only small volumes of material to probe mechanical properties, this technique is suitable for determining localized properties and is especially useful in probing material properties of thin coating. Instrumented indentation has been used to determine elastic properties, yield stress, and strain hardening [64, 65], and residual stresses [66]. General elasto-plastic properties extraction procedures from instrumented indentation experiments have been suggested [67, 68]. The instrumented indentation technique has the attributes of being quite simple and amenable to quantification.

There have been many investigations of failure properties of coatings using indentation (e.g., [69, 70]). Investigations have measured the adhesion of brittle films on a ductile substrate [71] and have observed preferred pathways for local cracking and separation in thermal spray coatings. Li and Bhushan [71] have used indentation techniques to measure the fracture toughness of thin, amorphous carbon films. Previous work by the author presented the results of Vickers indentation experiments carried out on four different boron carbide/DLC composition coatings that were sputtered deposited onto 52100 steel disks [9]. Although the indentation depths for these experiments ranged from less than 10% of the coating thickness to many times more than the coating thickness, qualitative comparisons of coating toughness were made. There have been numerous investigations to explore indentation coating failure using numerical methods.
Begley et al. analyzed wedge impression test for measuring interface toughness between films and substrates using numerical methods [72]. Chen et al. recently developed numerical methods to explore the mechanics of indentation induced lateral cracking [73]. Theoretical issues surrounding the extraction of elastic and plastic properties from indentation load versus displacement data have also advanced; however, there are still a number of ambiguities surrounding actual physical processes involving indentation of such small volumes (e.g., [74, 75]). Equipment calibration also becomes critical at such small displacements and loads.

The spherical indentation technique involves pushing a hard sphere into the surface of the coating-substrate system while the load and the depth of indentation are continuously measured. After the removal of the sphere, the impression is examined under an optical or scanning electron microscope. The impression usually reveals patterns of cracks in the film caused by tensile stretching of the film. The cracks are typically circumferential in nature, located near the periphery of the impression as indicated in Figure 2.9. In some cases, smaller radial cracks occur both inside and outside the impression. Regions of film within the cracked zone can also spall from the substrate.
Figure 2.9: An illustration of circumferential cracks produced on the surface of the coating by spherical indentation
Begley et al. [76] and Wang et al. [77] have investigated the spherical indentation of thin films and focusing on very large indentation depths (relative to the film thickness). The argument given by them is that for very low ratios of film thickness to ball (indenter) diameter and large indentation depths, the film does not play a significant role in the indentation process. The surface strain of the substrate governs the strain in the films and consequently, the experiment can be modeled by ignoring the influence of the film on the indentation process, except through its effects on the friction coefficient. Begley et al. have used flow theory to determine the near surface plastic strains beneath the spherical indenter impressed into a surface [ref]. The finite element model [76] predicted that the strains are strongly influenced by the coefficient of static friction as the coefficient changes from 0 to 0.3. However, for friction coefficients greater than 0.3 (i.e., in the sticking friction regime), there are no further changes in the strain field with the increase in friction coefficient values. The radial strains which are responsible for the circumferential cracks on the surface of the films were presented as functions of impression depth, yield strain of the substrate, and hardening exponent of the substrate.

Weppelmann and Swain [78] used finite element analysis to simulate spherical indentation and to determine the stresses responsible for the first fracture event of the film. The surface radial stresses were determined for various ratios of indenter radius and film thickness. A fracture mechanics analysis using the weight function method was carried out to calculate the stress intensity factors $K_I$ and $K_{II}$ within the near surface region of the film.
The crack growth in the film due to indentation may produce discontinuities or steps in the load displacement characteristics. The occurrence of these steps in the load displacement curves has been shown to be associated with the formation of circumferential cracks around the indented zone [79-81]. According to Li and Bhushan’s [71] theoretical analysis, fracture toughness of thin films can be defined as:

$$K_{IC} = \left[ \frac{E}{(1-\nu^2)2\pi C_R} \left( \frac{U}{t} \right) \right]^{1/2},$$

(2.3)

where, $E$ is the elastic modulus of the film, $\nu$ is the Poisson’s ratio of the film, $2\pi C_R$ is the crack length in the film plane, $U$ is the strain energy difference before and after the cracking, and $t$ is the film thickness. The strain energy difference, $U$ can be calculated from the steps or the discontinuities in the loading curve.

Sriram et al. have carried out studies to understand the mechanics of film fracture due to formation of circumferential cracks based on finite element analysis (FEA) of spherical indentation of a thin hard film deposited on a soft substrate, [81] The stress fields in the film, as well as the energy release rates associated with the circumferential cracks at different radial locations and indentation depths were studied using FEA. Using a domain integral formulation, the energy release rate was computed from the finite element results throughout the indentation analysis of films with preexisting circumferential cracks. The experimental data containing the indentation load (P) versus displacement (h) curve which shows steps or discontinuities were superposed on a computationally generated nomogram consisting of P versus h curves for different crack lengths and constant energy release rate, $J$ trajectories.
2.7. **COHESIVE ZONE FINITE ELEMENT MODELING**

Cohesive zone modeling is based on the consideration that infinite stresses at the crack tip are not realistic. The damage in the structure is essentially described by the cohesive elements but the continuum elements are not damaged.

**Figure 2.10:** Different forms of traction separation laws to define cohesive elements: (a) Needleman exponential law, (b) Bilinear law, and (c) Quadrilateral (box) law
Cohesive elements specifying only the damage of the material are inserted between the continuum elements following an arbitrary material law. The cohesive elements open when damage occurs and lose their stiffness at failure so that the continuum elements are essentially disconnected. As a result, crack propagation occurs only along continuum element boundaries. The separation of the cohesive elements is calculated from the displacement jump which is the difference of the displacements of the adjacent continuum elements. The classical investigation of Needleman [85, 86] and later on by Tvergaard and Hutchinson [87] has made the use of cohesive zone models very popular. The damage in the cohesive elements is governed by traction separation laws which specify traction across cohesive element for given displacement jumps. There are several forms of traction separation laws available in literature. Some of the most commonly used laws are shown in Figure 2.10. The area under the curve described by the traction separation law gives a measure of the work of separation to create two crack surfaces. The area under the curve is equivalent to the critical energy release rate, $G_C$. For instance, for the bilinear law in Figure 2.10b, the response of the cohesive zone element is determined by specifying $K$, the loading slope or cohesive stiffness, $\sigma_C$, the critical stress, and $G_C$, the area under the traction versus displacement curve (or $\delta_C$, the critical separation, instead of $G_C$). Cohesive finite element simulations are currently used [88] for strong films on a ductile substrate, but have also been used for composites investigations [89] and to study fatigue [90].

For most cases, as indicated in the bilinear traction separation law of Figure 2.11a, the reloading which occurs after unloading phase follows the same path as that of the
unloading rather than the previous loading path and, therefore, preserves the previous damage level. However, there is a limitation which prevents the crack growth under cyclic loading due to the progressive degradation of the cohesive properties in the cohesive failure zone. To overcome this limitation and to simulate fatigue cracking, several research groups, e.g., [91-96] have implemented cohesive zone elements with progressively degrading cohesive stiffness as shown in Figure 2.11b. For instance, if a cohesive surface is cycled between opening displacements of 0 and $\delta_0 (\delta_0 < \delta_c$ of Figure 2.11b), then Nguyen et al [94] have prescribed the following bilinear traction separation law:

$$T = K\delta_f \left(1 - e^{-\delta_c/\delta_f}\right), \quad (2.4a)$$

where, $K$ is the stiffness after ‘$n$’ cycles, given by:

$$K = \lambda^n K^0, \quad (2.4b)$$

with $K^0$ being the initial loading stiffness and $\lambda$ is the stiffness degradation parameter given by

$$\lambda = \frac{\delta_f}{\delta_0} \left(1 - e^{-\delta_c/\delta_f}\right)^2 + e^{-2\delta_c/\delta_f} \quad (2.4c)$$

where, $\delta_f$ is a characteristic opening displacement.
Figure 2.11: (a) Reversible and (b) Irreversible bilinear cohesive law[93]
2.7. OPTIMIZATION OF COATING ARCHITECTURE

As mentioned in Sec. 2.2, there exist numerous experimental and numerical investigations in the literature which offer qualitative and comparative performances of multilayer versus single layer coatings. However, there seems to be a gap in the literature when it comes to investigating an optimized coating architecture to minimize coating damage for a given set of loading conditions. It is of great importance and interest for a coating designer to know how to adjust the number of layers, layer thicknesses, and the overall coating thickness for an optimized coating performance. Lakkaraju et al. [97] have made an effort to bridge the aforementioned gap concerning optimization of multilayer coating architecture. FEM was used consisting of 8 layered multilayer coating on a thick substrate subject to Hertzian contact. The constitutive models were chosen to represent multilayer coating consisting of Cr/CrN deposited on A2 steel and 2024 Al substrates. The overall coating thickness was fixed to 2μm. A multi-objective optimization formulation was used to minimize von Mises stress in the topmost CrN layer and strain discontinuity in the coating thickness direction along the Cr/substrate interface. The thicknesses of Cr and CrN layers were chosen as design variables. The optimization was solved via sequential quadratic programming (Non Linear Programming by Quadratic Lagrangian, NLPQL) approach. Multiobjective Genetic Algorithm (MOGA) approach was also used as a comparative alternative to solve the optimization problem. Although this study is a right step with regards to the optimization of coating architecture, as mentioned below, there are certain limitations with the approach:
1) The choice of functions to be minimized, von Mises stress and strain discontinuity, may not be appropriate for representing damage in the coating. The brittle nature of thin CrN layer perhaps warrants a maximum principal stress of objective function. Also, strain discontinuity seems to be a vague parameter to quantify delamination of the coating. A stress-based approach better reflects damage in interface.

2) The choice of overall thickness of 2μm and fixed number of layers (8 in the study) are not justified. From a coating design standpoint it is important to know whether these indeed are optimum parameters for given load conditions. For instance, the cost of production of coatings is dependent on the number of layers. A designer would like to know the tradeoff in terms of cost with increase or decrease in number of layers.

3) The authors have not mentioned clearly what criteria were used to extract a solution from the Pareto set. It is perhaps better suited from a design perspective to prescribe a Pareto frontier first and then use engineering judgment to pick an optimal design point. Pareto frontiers can help deciding in terms of tradeoffs.

4) Although sequential quadratic programming can be an efficient way of solving non linear constrained optimization problems, from a design standpoint it may sometimes be well suited to obtain response surface like models so that the role of various design parameters can be ‘visualized.’ This is where metamodeling techniques can help.
The subsequent chapters discuss the finite element models developed to characterize damage in coating systems, and how models can be used for developing optimized multilayer coating architecture.

REFERENCES


CHAPTER 3

A COHESIVE ZONE FINITE ELEMENT APPROACH TO MODEL TENSILE CRACKS IN THIN FILM COATINGS

Abstract

A 2-dimensional finite element model using cohesive zone elements was developed to predict cracking in thin film coating – interlayer – substrate systems that are subjected to tensile loading. The constitutive models were chosen to represent a metal carbide/diamond like carbon composite coating with a titanium interlayer and a steel substrate. Material properties of the coating and interlayer along with the cohesive finite element parameters were varied to study effects on stress distributions and coating cracking. Stress distributions were highly non-uniform through the coating thickness. Thus the initiation and arrest of tensile cracks differed from what is predicted by simple shear-lag theory. Inter-crack spacing distributions resulting from the variation of different parameters were quantified and compared with those from experiments.

Keywords:

Thin film coatings, cohesive zone finite element modeling, tensile cracking, inter-crack spacing
3.1. INTRODUCTION

Coating systems provide enabling technologies that have enhanced productivity for a wide variety of applications. Hard coatings are a class of coatings that have been developed as a surface engineering enhancement solution for cutting tools, dies, drills, and other tribological applications. All of these applications rely on the fact that the coatings are extremely hard, abrasion resistant, and/or provide low friction surfaces. Most hard coatings are ceramic compounds such as carbides, nitrides, ceramic alloys, cermets, and metastable materials such as diamond and cubic boron nitride. Their properties and environmental resistance depend on composition, stoichiometry, impurities, microstructure, and texture. To effectively design coating systems for specific applications, it is necessary to know the chemical, mechanical, and tribological properties of the coatings.

To a large extent, adhesion at the coating-substrate interface and toughness of the coating itself determines the durability of hard coating systems. Loss of adhesion at the coating-substrate interface leads to premature failure of otherwise wear resistant and tough coatings. There have been many investigations of failure properties of coatings using indentation (e.g. [1, 2]). Investigations have measured the adhesion of brittle films on a ductile substrate [3] and have observed preferred pathways for local cracking and separation in thermal spray coatings [4]. Li and Bhushan [3] have used indentation techniques to measure the fracture toughness of thin, amorphous carbon films. Previous work by the authors presented the results of Vickers indentation experiments carried out
on four different boron carbide/DLC (Diamond-Like Carbon) composition coatings that were sputtered deposited onto 52100 steel disks [4]. Although the indentation depths for these experiments ranged from less than 10% of the coating thickness to many times more than the coating thickness, qualitative comparisons of coating toughness were made. There have been numerous investigations to explore indentation coating failure using numerical methods. Begley et al. analyzed wedge impression test for measuring interface toughness between films and substrates using numerical methods [5]. Chen et al. recently developed numerical methods to explore the mechanics of indentation induced lateral cracking [6]. Based on finite element analysis (FEA) of spherical indentation of a thin hard film deposited on a soft substrate, Sriram et al. have carried out studies to understand the mechanics of film fracture [7].

Theoretical issues surrounding the extraction of elastic and plastic properties from indentation load versus displacement data have also advanced; however, there are still a number of ambiguities surrounding actual physical processes involving indentation of such small volumes (e.g., [8, 9]). Equipment calibration also becomes critical at such small displacements and loads.

Scratch testing is a widely used technique for evaluating the adhesion of thin, hard coatings. Scratch testing is useful to compare the adherence of similar coating-substrate systems; however, it fails to quantify parameters like interfacial strength between the coating and the substrate and cohesive strength of the coating. A novel method for evaluating adhesion strength was proposed by Agrawal and Raj [10]. This “tensile cracking” approach evaluates both the cohesive strength of the coating and the interfacial adhesion strength between the coating and the substrate. The approach is based on
subjecting a brittle coating on a ductile substrate to tension and thereby propagating coating cracks that are oriented transverse to the tensile direction. A micrograph showing transverse cracks in a tungsten carbide/DLC (WC-DLC) coating on a stainless steel tensile specimen is shown in Figure 3.1. The crack density increases with substrate tension until the crack spacing reaches a saturation value when it is no longer influenced by the increase in substrate tension. The model relates \( \hat{\tau} \), the interfacial shear strength between the coating and the substrate to \( \hat{\sigma} \), the tensile fracture strength of the film, \( \hat{\lambda} \), the characteristic saturation crack spacing, and \( \delta \), the coating thickness [10]:

\[
\hat{\tau} = \frac{\pi \delta \hat{\sigma}}{\hat{\lambda}}. \tag{3.1}
\]

For a highly elastic coating, the tensile strength of the coating, \( \hat{\sigma} \) can be written as

\[
\hat{\sigma} = E \varepsilon_f, \tag{3.2}
\]

where, \( E \) is the modulus of elasticity of the coating and \( \varepsilon_f \) is the strain corresponding to the onset of cracking in the coating.
Agrawal and Raj proposed the full sine wave function shown in Figure 3.2a for approximating the interfacial shear stress. This approximation results in zero interfacial shear stress values at the midpoint and at both ends of the inter crack spacing in the coating. Other research groups have asserted that, as shown in Figure 2b, the interfacial shear stress should have its maximum value located at each end of the inter-crack spacing and be zero at the midpoint of the inter-crack spacing in the coating [11-14]. The recent analytical work of Yanaka et al. [13] and Wojciechowski and Mendolia [14] and the experiments and FEA of Chen et al. [11, 12] indicate that the interfacial shear stress distribution is best approximated by a distribution like the one shown in Figure 3.2b. The applicability of these two different shear stress distributions may be related to how the crack spacing compares to the coating thickness.
Figure 3.2: Two different theories predicting the distribution of interfacial shear stresses along the coating-interlayer interface between two cracks when the coating-interlayer-substrate system is subject to tensile loads (a) sinusoidal distribution with zero stress at the endpoints [10] and (b) maximum stress at the endpoints [11, 13, 14].

The previous work with finite element analysis of tensile cracking assumes preexisting cracks by introducing fine notches in the model [11-15]. Few researchers have developed finite element models which can simulate the formation and propagation of tensile cracks and thus predict coating performance. The present work uses cohesive zone elements to model crack development in the coating. After calibration with simple tensile cracking experiments, the finite element model could be used to predict the behavior of the coating-substrate system for the more complex loadings associated with actual applications. The next section of this chapter describes the finite element model and the various physical and numerical parameters. The model is based on tensile cracking experiments that were carried out with WC-DLC coatings on 310 stainless steel...
substrates. The third section presents results for varying model parameters and the final section then discusses the resulting simulated tensile cracking behavior.

3.2. FINITE ELEMENT MODEL DESCRIPTION

An idealization of the coating system that was modeled is shown in Figure 3.3a. Figure 3.3(b) shows the magnified view of the coating with details on where cohesive elements were placed. The ABAQUS 6.5.1 commercial FEA package was used for all the modeling. As indicated above, the model was loosely based on tensile cracking experiments with WC-DLC coatings. The coating was 1.3μm thick and was assumed to be homogeneous, isotropic, and perfectly elastic. The 310 stainless steel substrate followed an elastic-plastic material model with linear strain hardening.

The substrate thickness was 230 times the coating thickness. A 1.3μm thick elastic-plastic titanium interlayer was present between the coating and substrate. Base values for the substrate properties were obtained from in-house experiments. Base values for the coating modulus and interlayer properties, henceforth referred to as the “reference case,” were obtained from [16] and www.matweb.com [17], respectively. The coating is assumed to be homogeneous along the thickness. The material properties for the substrate, coating, and interlayer are provided in Table 3.1. Four node (QUAD4) plane strain elements were used for the analysis. As shown in Figure 3.3, symmetry boundary conditions were applied along the bottom of the model, and displacements were applied at the right and left edges. Displacements were specified at the edge nodes of the substrate and interlayer but not on the coating. The maximum displacement values were
such that the strain in the direction of the loading was 4%. The final 4% strain value was chosen to be consistent with experiments.

Figure 3.3: (a) Schematic of the FE model (not to scale); (b) illustration of cohesive zones in the coating (magnified view of (a)).
### Table 3.1: Coating, interlayer, and substrate properties used for the different simulations.

<table>
<thead>
<tr>
<th></th>
<th>Reference Case</th>
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<th>Mod. #2</th>
<th>Mod. #3</th>
<th>Mod. #4</th>
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<td>Properties</td>
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<td>properties</td>
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<tr>
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<td>2500-2650</td>
<td>2500-2650</td>
<td>2500-2650</td>
<td>1500-1650</td>
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<tr>
<td>$G_C$ (J/m²)</td>
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<td>32-37</td>
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Figure 3.4: Bilinear Law followed by the cohesive elements used in the study. Cohesive elements have a stiffness $K$ and fail when the critical stresses in the elements reach beyond the critical stress $\sigma_C$.

Cohesive zone finite elements were placed through the thickness of the coating. These bi-linear elements are available in the standard ABAQUS element library. The cohesive behavior assumes linear elastic traction-separation law prior to damage and a linear damage evolution based on energy dissipated due to failure, $G_C$. The input parameters for the cohesive elements are further described below. The layout of these elements is shown in Figure 3.3b. In the middle of the coating the distance between each row of cohesive zone elements is 1μm. Away from the middle of the model, the spacing was increased to 10μm. In this way, computational expense was reduced while focusing on a region that is far away from any edge effects. The interface is assumed to be perfectly bonded, and therefore there are no cohesive elements along the interface. This assumption is based on industry experience with state-of-the-art PVD thin film coatings. In addition, Wang et al. [18] have predicted that the interface toughness is greater than
150 J/m$^2$, while the film toughness is approximately 30 J/m$^2$ for DLC films on steel substrates.

As shown in Figure 3.4, the response of the cohesive zone element is determined by specifying $K$, the loading slope or cohesive stiffness, $\sigma_C$, the critical normal stress, and $G_C$, the area under the traction versus displacement curve. The area under the traction versus displacement curve is named $G_C$ because it is a measure of the critical energy release rate. Values for the cohesive zone element parameters are provided in Table 1. The critical normal stress values were estimated based on experiments with acoustic emission [19]. From experiments, the strain corresponding to the onset of cracking was estimated to be 1.2%. The critical normal stress, $\sigma_C$, can then be obtained from equation (3.2). Similar strain values were also observed by Wang et al for DLC films on steel substrates. [18]. The base-line $G_C$ values used in this study were consistent with those predicted by Wang et al. [18]. The cohesive stiffness was held constant for all cohesive zone elements.

Although at a given location in the coating the cohesive parameters were kept the same for all the coating thru-thickness lines of cohesive zone elements, the $\sigma_C$ and $G_C$ parameters were allowed to vary along x-direction. The different $\sigma_C$ values were obtained by using MATLAB’s random number generator to generate uniformly distributed values in a prescribed range. The range of variation for $\sigma_C$ was $\pm 75$ MPa. The critical separation ($\delta_C$, in Figure 3.4) was held constant, and therefore the range of variation for $G_C$ was 32-37 J/m$^2$. The random assignment of different parameter values to different cohesive zone elements served the following two purposes: (a) it simulated the natural, statistical
variation of the coating’s cohesive strength, and (b) the different values of critical stresses in different cohesive zones ensured that not all the cohesive zone elements failed together when the critical tensile load was reached.

The modeling of progressive damage involves softening of the material response, which leads to convergence difficulties in an implicit solution procedure. To overcome such convergence problems, ABAQUS/Standard implements a “viscous regularization parameter.” This parameter regularizes the traction-separation laws by permitting stresses to be just outside the limits set by the traction-separation law. Using a small value for the viscosity regularization parameter improves the rate of convergence of the model in the softening regime and does not alter the sequence of cracking. Detailed discussion of this parameter can be found in [20, 21]. The parameter was kept the same for all the cohesive zone elements. By trial-and-error, the smallest value that still produced converged solutions was used. The solution was carried out using implicit analysis and converged in approximately 550 total iterations. Up to the point where cohesive elements would start failing, the stresses in the coating were found to be the same in the models without and with cohesive elements. This indicates that the inclusion of the cohesive elements was not altering the stress state before damage initiation.

The model was also checked for mesh convergence. The “original” mesh scheme described above (5 elements between two neighboring cohesive elements in the middle of the coating and 9 elements along the thickness of the coating) was compared with two other mesh schemes: a “coarser” mesh containing 3 elements between two neighboring cohesive elements in the middle of the coating and 5 elements along the thickness of the
coating and a “refined” mesh containing 8 elements between two neighboring cohesive elements in the middle of the coating and 13 elements along the thickness of the coating. Figure 3.5 shows the horizontal-direction normal stress along the surface of the coating and plotted for a region with higher cohesive element density. The three different mesh schemes are shown at 2% strain. From Figure 3.5, since there is little difference between the “original” and “refined” meshes, it is assumed that mesh convergence was achieved with the “original mesh”.

![Graph](image-url)

**Figure 3.5:** Horizontal-direction normal stress profiles at 2% strain along the surface of the coating for 3 different mesh schemes.
3.3. RESULTS

In order to explore the effect of various parameters on the model response, one set of parameters was designated as the “reference case” (see Table 3.1). The subsequent studies were variations on the reference case. The next two subsections discuss the stress distributions in the reference case and the effect of variation of different parameters with respect to the reference case.

3.3.1 Stress Distributions

For the reference case, when the normal stress in the tensile direction ($\sigma_{xx}$) in the coating exceeds the smallest random $\sigma_C$ value, the first crack(s) appear. The tractions on the newly formed crack faces go to zero, and therefore the coating stresses are redistributed. An example of two $\sigma_{xx}$ surface stress contours between two cracks which are 20μm apart is shown in Figure 3.6. Since the $\sigma_{xx}$ surface stress is zero at the cracks, both edges of the plot show zero stress. For the “$i$th” load increment, the stress is still uniform in the middle region between the two cracks. In the “$i+1$” increment the stress in the middle of the region of interest has exceeded the critical value for one of the cohesive zone elements, a new crack is generated, and the $\sigma_{xx}$ surface stresses go towards zero. This crack progression is consistent with the shear lag models for tensile cracking [10-14].
Figure 3.6: Evolution of tensile stresses between two cracks 20 microns apart with the increase in load.

From shear lag assumptions and from the above description of the contours in Figure 3.6, one would expect the cracks to always form at the mid-point between two adjacent cracks. However, this expectation is based on the assumption that the stress profile in Figure 3.6 is unchanged through the thickness of the coating. Uniform stresses along the thickness of the coating are also an underlying assumption in the analytical models referred to above [10-14]. It was found that the stresses are not uniform through the coating thickness. Since one side of the coating is free and the other side is bound to the interlayer, stresses should vary through the thickness of the coating. The crack formed at the mid-point in Figure 3.6 because of the particular combination of geometric and materials parameters.
Figure 3.7: Normal stress distribution along paths in the coating between two cracks 10 microns apart.

In Figure 3.7, for the region between two cracks that are 10μm apart (there are no cohesive elements between these cracks) the $\sigma_{xx}$ stress profiles are plotted for paths at
different coating depths. The stress profiles are not at all self-similar, and the location of the maximum $\sigma_{xx}$ is not always at the mid-point between two adjacent cracks. In particular, the highest $\sigma_{xx}$ stresses are found close to the interface and close to the existing cracks. Figure 3.7 also serves to highlight that near-crack stresses along the surface of the coating are compressive, and near-crack stresses near the interface are tensile. All stresses become tensile as one approaches the mid-point of the region. A similar stress distribution was observed by Krishnamurthy and Reimanis.[15]

**Figure 3.8:** Crack in the coating originating from the interface. The inset shows experimental observation by Krishnamurthy and Reimanis [15]

As shown in Figure 3.8, the internal stresses and boundary conditions on an unbroken coating segment are such that the coating undergoes significant bending. The
bending of the coating results in compressive near-crack stresses close to the surface of the coating. This stress distribution has important implications for crack propagation, with the high tensile near-crack stresses close to the interface suggesting that the cracks will originate from the interface. Indeed, the tensile cracking experiments performed by Krishnamurthy and Reimanis indicated that cracks may be originating from the interface [15] as shown in the inset in Figure 3.8.

![Stress distribution](image)

**Figure 3.9:** Cracks in the coating for reference model (a) 2.5% strain, (b) 4% strain: Notice the crack arrest.
The near-crack compressive stresses that exist closer to the surface of the coating will cause cracks originating from the interface to be arrested. Figure 3.9a and 3.9b show a longer portion of the cracked coating for 2.5% and 4% overall strain, respectively. Figure 3.9 shows that, although multiple cracks originating from the interface were present at 2.5% strain, there was little or no continued crack propagation towards the surface even after 4% strain. If the coating is examined from the top (as most SEM micrographs do), it would appear that there are no more cracks on the surface of the coating and that the crack spacing has saturated.

3.3.2 Parameter Study

As shown in Table 1, the first modification of the reference case was increasing the modulus of the coating. All other material and cohesive parameters were kept the same. The higher coating modulus resulted in higher stresses at lower overall strains and, as expected, the fracture strain (strain at which the first crack appears) decreased. Furthermore, as a result of the higher stresses in the coating, it can be seen from Figure 3.10a that more cracks have progressed from the interface to the surface of the coating.

The second modification to the reference case model was made by increasing the slope of the tangent modulus for the plastic region of the interlayer. This essentially increased the strain hardening of the interlayer. It can be seen in Figure 3.10b that, as compared to the reference case, more cracks have progressed from the interface to the surface of the coating. This results from the higher stresses in the interlayer for the same amount of strain as compared to the reference case. With a less deformable interlayer, the unbroken coating segment does not bend as much and therefore the near-crack
compressive stresses close to the surface of the coating are diminished. As a result, cracks can more readily propagate to the surface.

**Figure 3.10**: Change in cracks from reference model due to (a) increase in coating modulus, (b) increase in tangent modulus slope in interlayer, and (c) decrease in tangent modulus slope in interlayer. All images correspond to 2.5% strain.
A third modification to the reference model was made by decreasing the slope of the tangent modulus for the plastic region of the interlayer. This change has the overall effect of making the interlayer more deformable. The contour plot in Figure 3.10c shows that there is more curvature of the segments and the cracks have larger openings. The curvature of the coating increases due to the increase in the softening of the interlayer. The increase in the curvature results in an increase of the compressive stresses near the surface of the coating. Consequently, there are fewer cracks progressing to the surface. Also, there is an increase in the curvature of the interface as well. This leads to the higher tensile stresses near the interface, and as a result, more cracks originate from the interface compared to the reference case. This can be seen in Figure 3.11a and 3.11b where $\sigma_{xx}$ stress profiles between two cracks are plotted at the surface of the coating and near the interface (paths 3 and 1 of Figure 3.7) for the two different interlayer cases. For the last modification of the reference case, the $\sigma_C$ values of all the cohesive zone elements were decreased by 1000 MPa compared to the reference case while keeping $G_C$ the same. Decreasing $\sigma_C$ while keeping $G_C$ constant essentially decreases the softening slope of the cohesive zone element and, hence, the critical separation ($\delta_C$, in Figure 3.4) increases. These changes result in a decrease in the fracture strain compared to the reference case. When comparing to the reference case, at 2% strain, Figure 3.12a and 3.12.), show that the decreased $\sigma_C$ results in more cracks generated. It could be argued that if the reference case coating-substrate system were pulled to higher strains, then more cracks would propagate to the surface. However, as will be discussed in the next section, there are
assumptions with the 2D model that need to be taken into account before making such an argument.

**Figure 3.11**: Comparison of $\sigma_{xx}$ stresses between two cracks 10$\mu$m apart for two interlayer cases (a) stresses at the surface of the coating (b) stresses near the coating-interlayer interface.
**Figure 3.12**: Differences in the formation of cracks for 2% strain: (a) reference case, (b) decreased $\sigma_C$.

### 3.4. DISCUSSION

The above parameter study indicates that cracking in the coating is sensitive to the material properties of the coating and interlayer and also to the cohesive zone element parameters. The model also demonstrates “surface level” crack saturation which means
that no new cracks will appear on the surface with increasing tensile strain. Shear lag analysis of tensile cracking that includes the assumption of uniform thru-thickness stresses predicts that the crack spacing will saturate [10-14]. The current simulations indicate that although the crack saturation can exist at the coating surface level, there may be sub-surface cracks that have not propagated all the way to the surface.

In the current model, further straining of the coating substrate system would eventually cause all cracks to propagate to the surface. However, this situation is the result of having a 2-d model. In reality, the stress state is no longer uniaxial at very high strains. The triaxial nature of the stress state in the plastic region causes slant cracks at approximately 45° to the existing transverse cracks appear in the coating [12]. The existence of these slant cracks is a fair indication that strains are beyond the surface-saturation strains and that the stress state is no longer uniaxial. Without a 3-d model, slant cracks cannot be modeled. Experimental results for the WC-DLC system indicated that slant crack began at approximately 4.5% strain. For this reason, the current simulations were run to 4% strain.

The amount of curvature or bending of coating due to asymmetric boundary conditions and compliance of the substrate is also worth noting. The through thickness cracks can deflect, depending on the fracture resistance of the interface, parallel to the coating-substrate interface. The deflection of cracks parallel to the interface and the bending of coating can ultimately result in spalling of the coating. This phenomenon is illustrated in Figure 3.13. The magnitude of bending moment is a function of the geometry and material parameters of coating, interlayer, and substrate.
Figure 3.13: Illustration of spalling of coating due to through thickness coating cracks and bending of coating

With increase in coating modulus, coating stress in the tensile direction increases for the same amount of strain. Hence, bending moment increases causing the coating to have more curvature. Figure shows the variation of curvature along the surface of coating between two cracks 10 μm apart for different elastic modulus of coating. Decreasing the interlayer strain hardening slope also increases the bending of coating as the compliance
below the coating decreases. Figure shows the variation of curvature along the surface of coating between two cracks 10 μm apart for different interlayer strain hardening slope.

Figure 3.14: Variation of curvature of coating with change in elastic modulus of coating
Figure 3.15: Variation in curvature of coating with change in interlayer hardening slope

The distribution of the crack spacing for the cracks in Figure 3.1 is given in Figure 3.16. The mean value of the crack spacing is 6.0 μm with a standard deviation of 2.4 μm. A normal distribution with the given mean and standard deviation values is provided in the same plot. The distributions of crack spacing from the numerical simulations described in Sec. 3.2 are shown in Figure 3.17a-d. Only cracks which have propagated all the way to the surface of the coating are included in the distribution plots. As seen from Figure
3.17a, the mean crack spacing for the reference case is less than that from the experiments. Also, as seen in Figure 3.17b and Figure 3.17d, with the increase in coating modulus and the decrease in \( \sigma_c \), respectively, the distribution of crack spacing is unrealistically small since almost every cohesive element has failed. As expected, Figure 3.17c shows that the mean crack spacing for the case of increase in tangent modulus slope of the interlayer is less than that of the reference case. It must be pointed that the distributions in Figure 3.17 are based on a coating span of only 40\( \mu \)m as compared to the coating span of 130\( \mu \)m used for the experimental results shown in Figure 3.16.

![Graph showing distribution of inter-crack spacing for WC/DLC coating pulled to 4\% strain.](image)

**Figure 3.16:** Distribution of inter-crack spacing for WC/DLC coating pulled to 4\% strain.
In the numerical simulations discussed above, the coatings were assumed to be isotropic, homogeneous, and free of defects. However, as seen in Figure 3.1, “pits” of various sizes are present on the surface of the coating. Although many of the cracks go through pits, there are also pits without any cracks. More detailed investigation is required to confirm whether or not pits are assisting crack propagation. Microstructural observations would need to be performed in order to determine if there are any additional microstructural inhomogeneities that affect crack spacing.

**Figure 3.17:** Distribution of inter-crack spacing observed in numerical simulation (a) reference case, (b) increase in coating modulus, (c) increase in tangent modulus slope in interlayer, and (d) decrease in σC of cohesive elements
Investigation of residual stresses was undertaken with compressive residual stresses of 1 GPa as uniform “pre” stresses in the coating. It was observed that inclusion of compressive residual stresses caused the fracture strain (strain at which the first crack appears) to increase from 1.2% to 1.7%. At lower strain values (e.g., 2%), there are fewer cracks compared to same strain value in the reference case. However at large strain values (>4%), results look very similar to the reference case results.”

3.5. SUMMARY

A 2D finite element model was created to simulate the response of a coating-interlayer-substrate system to in-plane uniaxial tension. Coating cracking was simulated with cohesive zone elements that followed a bilinear cohesive law. Some degree of randomness was introduced into the model through the assignment of random critical cohesive stresses. Due to the unsymmetrical boundary conditions at the top surface and coating-interlayer interface, bending occurred and introduced non-uniform stresses through the coating thickness. Thus coating cracks propagated from the interface and were often arrested near the surface of the coating by the presence of high compressive stresses. The effects of different coating modulus, tangent modulus for the interlayer hardening, and critical stress values ($\sigma_C$) of the cohesive zone elements were studied. The distribution of crack spacing for different parameter changes were quantified and compared to an experimental crack spacing distribution.

Due to the limitations of using a 2D model, crack formation and propagation at high plastic strain values was not predicted accurately. Incorporating measured residual
stresses into the model is necessary for more realistic simulations. The model has been shown to be sensitive to material and cohesive parameters and could thus be used to optimize coatings. The described tensile cracking experiment could be used to calibrate cohesive parameters for subsequent modeling of more complicating loading schemes.

REFERENCES


CHAPTER 4

OPTIMIZATION OF MULTILAYER COATING ARCHITECTURE

Abstract
As compared to monolithic coatings, multilayer coatings with alternating hard and soft layers are finding increased applications because of the seemingly better performance in tribological and wear applications. However, the roles of overall thickness, number of layers, and individual layer thickness cannot be overlooked and need to be optimized to minimize damage in the multilayer coatings. 2-dimensional finite element models using cohesive zone elements were developed to predict damage in multilayer coatings subject to spherical indentation. Damage in coatings was characterized as through thickness coating cracks and interfacial delamination. A design of computer experiments (DACE) approach was used to build metamodels in order to predict damage variables for a design space consisting of 2, 4, 6, and 8 layers multilayer coating architecture.

Keywords:
Multilayer coatings, cohesive zone finite element modeling, spherical indentation, DACE, Kriging
4.1. INTRODUCTION

4.1.1. Multilayer Coatings

Monolithic hard protective coatings are quite often used to increase the longevity of tools and tribological components in heavy duty service environments. However, there are limitations associated with monolithic coatings such as lack of multifunctional character, high residual stresses, problems associated with adhesion to substrate, etc. This has led to increasing use of multilayer coatings. Subramanian and Strafford [1] presented a good review of multilayer coatings for tribological applications. Multilayer coatings not only offer the combination of attractive properties from different materials, but also have observably increased tribological performance over monolithic coatings. Holleck and Schier [2] investigated the wear performance of multilayer PVD coatings. They compared single layer TiN, TiC, and multilayer TiN/TiC/B₄C coatings for hardness, friction coefficients, and life of coated tools and concluded that for each category, multilayer coatings had superior performance. Bull and Jones [3] investigated the performance of two types of multilayer coatings produced in Ti-N system: Structural multilayers in which the amount of ion bombardment that the coating receives during deposition was changed in a cyclic fashion to produce alternating layers of low and high residual stresses, and compositional multilayers in which nitrogen flow was interrupted periodically to produce alternating layers of titanium and titanium nitride. They concluded that both types of multilayers exhibited high hardness, good toughness, and improved adhesion leading to increase in wear resistance compared to single layer TiN coating. However, these properties were found to be dependent on periodic spacing of
layers. Dependence of tribological properties of multilayer coatings on periodic spacing of layers was also confirmed by investigations of Flores et al [4] and Romero et al [5]. Voevodin et al. [6] came up with a multilayer coating architecture design involving nanocomposite DLC layers for increased adhesion strength and improved load support capability under high contact loads. Bouzakis et al. [7] investigated the cutting performance of multilayer PVD TiN/TiAlN coatings in milling operations. They concluded that because of deceleration of cracks in multilayer coatings due to the layering structure, multilayer coatings exhibited improved wear performance over single layer TiAlN coatings.

Elastic modulus of the coating constitutes Dundur’s parameters which play an important role in the fracture mechanisms of coating substrate systems as discussed by Hutchinson and Suo [8]. It is believed that apart from hardness (H), modulus of elasticity (E) of coating also plays an important role in the tribological behavior of coatings. Leyland and Matthews [9] presented an article discussing the importance of H/E ratio for optimized tribological performance. They argued that high H/E ratio leads to better wear resistance of coatings as wear characterized by long elastic strain to failure can be described in terms of H/E ratio. The idea of increasing H/E of coatings is to increase deformation accommodation. Multilayers with alternating soft and hard layers also increase deformation accommodation as thin hard layers slide relative to each other by virtue of the shear deformation of low modulus layers. This theory was tested successfully by Matthews et al. [10] by simulating stack of multilayers with a beam comprising of alternate high and low elastic modulus layers. Shear deformation of Ti
layers in multilayer TiN/Ti coatings leading to fewer cracks during indentation loads was also confirmed by Ma et al. [11]. Xie et al. [12] also observed coating deformation primarily being accommodated by shear sliding and plastic flow of Ti interlayers in TiN/Ti multilayer coating subject to indentation loads. They also observed that radial cracks were arrested due to multilayer structure. Suresha et al. [13] argued that enhanced toughness of TiN/TiAlN multilayer coatings is not due to increase in strain capacity (H/E) of the film, but because multilayers display additional modes of plasticity leading to permanent bending and compression of the film.

The behavior of multilayer coatings has also been characterized by use of analytical and finite element models. For instance, Bull et al. [14] developed models based on work of indentation and energy approach to predict hardness and modulus response of multilayer coatings. They also considered effect of fracture on the model predictions. There have been many studies investigating contact stresses and fracture in layered solids subject to sliding contact. For instance, solutions for 2-D line contact were developed by Gupta and Wallowit [15], and King and O'Sullivan [16]. Oliveira and Bower [17] presented a fracture analysis of layered solids subject to sliding contact loading to calculate loads needed to initiate fracture in coatings and coating delamination. Djabella and Arnell [18] developed finite element models to predict stresses in multilayer coatings subject to Hertzian type contact loads. They compared the response of coatings comprising of 3 and 5 layers with elastic modulus decreasing from outer layer to substrate. They concluded that multilayered systems with the highest number of layers have the least severe stress distribution for a given total thickness. Chi and Chung
[19] evaluated the stress intensity factors for cracked multilayer coating substrate systems using finite element model. Their evaluation led to the conclusion that cracks in one layer coating propagate into the substrate easily compared to cracks in two or four layered coating when coating material is stiffer compared to the substrate. Gong and Komvopoulos [20] used finite element models and linear elastic fracture mechanics to analyze surface cracking behavior under rigid sliding contact loading condition. Fracture modes responsible for surface crack propagation were identified and influence of crack length and crack path were determined using the model. Monaghan and Murphy [21] used finite element models to study the influence of substrate on stresses in multilayer coatings in forming and cutting tools. Finite element models were also used by Bouzakis et al. [22] to predict load displacement behavior of multilayer TiN/TiAlN coating during nano indentation.

The above mentioned investigations clearly lead to the conclusions that the behavior of the multilayer coating substrate systems depend not only on the constitutive properties of individual layers, but also on individual layer thickness and number of layers. Hence, there is a need for optimized coating architecture to minimize coating damage for a given set of loading conditions. For instance, after studying the erosive and abrasive wear properties of TiN/Ti multilayer coatings, Bromark et al. [23] concluded that while multilayer coatings offer promising solutions for wear applications, demands of different applications can be met by adjusting relative thickness of Ti. Ma et al. [24] also emphasized the need for optimization of Ti layer thickness for greater coating adhesion strength in TiN/Ti multilayer coatings. Lyubimov et al. [25] presented a
mathematical model for stress analysis and failure assessment of multilayer PVD coatings. Thermal and intrinsic stresses were accounted for in the model and the failure mechanisms that were assessed were failure of each layer due to stresses in it, interfacial crack propagation, and buckling resulting in peeling of layers. The model was used to solve the optimal choice of thickness of layers to prevent failure. Gorishnyy et al. [26] investigated the stresses in single layer, bilayer, and multilayer Cr/CrN coatings through finite element models for hertzian contacts and pin on disk experiments. It was concluded that highest adhesion and lowest wear rates were observed for multilayer Cr/CrN coatings but the performance degraded with increase in thickness of Cr layers. They concluded their work by stating that a formal methodology is needed to optimize the overall coating design and individual layer thickness.

As indicated above, although there is a need to develop formal methodology for optimization of coating architecture design, there is a dearth of investigations formalizing this approach. Lakkaraju et al. [27] attempted to bridge this gap by formulating an approach to propose optimized coating architecture for minimizing damage in Hertzian contact loading conditions. The approach was based on setting up a finite element model with constitutive models representing multilayer coating consisting of eight alternating layers of Cr/CrN deposited on A2 steel and 2024 Al substrates. The optimization problem consisted of minimizing Von mises stress in the topmost CrN layer and strain discontinuity in the thickness direction along the Cr/substrate interface with individual layer thickness as design variables. The overall coating thickness was fixed to 2 μm. Sequential quadratic programming (Non Linear Programming by Quadratic Lagrangian,
NLPQL) approach was used to solve the optimization problem. While the approach is a rightful first step towards designing optimum coating architecture and deserves merit, there are some limitations: (a) The choice of functions to be minimized, von Mises stress and strain discontinuity, may not be appropriate for representing damage in the coating. The brittle nature of thin CrN layer perhaps warrants a maximum principal stress kind of objective function. Also, strain discontinuity seems to be a vague parameter to quantify delamination of the coating. A stress based function better reflects the damage in interface, (b) The choice of overall thickness of 2μm and fixed number of layers (8 in the study) are not justified in the study. From a coating design standpoint it is important to know whether these indeed are optimum parameters for given load conditions. For instance, the cost of production of coatings is dependent on the number of layers. A designer would like to know the tradeoff in terms of cost with increase or decrease in number of layers, (c) The authors have not mentioned clearly what criteria were used to extract a solution from the Pareto set. It is perhaps better suited from a design perspective to prescribe a Pareto frontier first and then use engineering judgment to pick an optimal design point. Pareto frontiers can help deciding in terms of tradeoffs and (d) Although sequential quadratic programming can be an efficient way of solving non linear constrained optimization problems, from a design standpoint it may sometimes be well suited to obtain response surface like models so that the role of various design parameters can be ‘visualized’. This is where metamodeling techniques can help.
4.1.2. Indentation to Characterize Fracture in Coatings

Coating systems are prone to failure due to fracture in coatings and/or substrate deformation. Fracture in coating systems primarily consist of cohesive and interfacial failure (delamination). Cohesive failure in the coating occurs when the energy release rate for flaws in the coating exceeds the fracture toughness of the coating. Similarly, fracture in the interface (of coating and substrate) occurs when the energy release rate for flaws in the interface exceeds the interfacial fracture toughness. Fracture in thin hard film coated systems is complex and controlled by the coating material, substrate and the interface which bonds the system together. There have been many investigations of failure properties of coatings using indentation (e.g. [28, 29]). Investigations have measured the adhesion of brittle films on a ductile substrate [30] and have observed preferred pathways for local cracking and separation in thermal spray coatings. Li and Bhushan [30] have used indentation techniques to measure the fracture toughness of thin, amorphous carbon films. Begley et al. analyzed wedge impression test for measuring interface toughness between films and substrates using numerical methods [31]. Chen et al. recently developed numerical methods to explore the mechanics of indentation induced lateral cracking [32].

The spherical indentation technique involves pushing a hard sphere into the surface of the coating-substrate system while the load and the depth of indentation are continuously measured. After the removal of the sphere, the impression is examined under an optical or scanning electron microscope. The impression usually reveals patterns of cracks in the film caused by tensile stretching of the film. The cracks are
typically circumferential in nature, located near the periphery of the impression. High radial normal stresses at the surface of the coating near the edge of indenter are responsible for circumferential cracks at the surface of coating. Cracks in the coating can also occur directly under the center of indenter near the interface because of high radial normal stresses. Figure 4.1 illustrates the formation of cracks in the coating during indentation. In addition, interfacial failure can also occur because of high shear stresses. Begley et al. [33] and Wang et al [34] have investigated the spherical indentation of thin films and focusing on very large indentation depths (relative to the film thickness). The argument given by them is that for very low ratios of film thickness to ball (indenter) diameter and large indentation depths, the film does not play a significant role in the indentation process.

**Figure 4.1:** Formation of coating cohesive cracks during indentation
Cohesive zone modeling is a popular method of characterizing damage in coatings. For instance, Abdul-Baqi and Van der Giessen [35] studied indentation induced delamination of the film from the substrate by employing cohesive elements along the coating-substrate interface. In another study [36], both circumferential cracking in the coating and delamination were investigated by employing through thickness and interfacial cohesive elements. The influence of material and cohesive parameters on the spacing of circumferential cracks was discussed.

The current research study aims to bridge the gap further in the area of optimization of coating architecture while addressing some of the limitations mentioned earlier. The subsequent sections in the chapter describe the finite element model used, optimization formulation, approach using design of computer experiments, results and discussion and finally the expected contribution.

### 4.2. Finite Element Model Description

In order to gain a better understanding of the performance of multilayer coating architecture, a benchmark finite element model consisting of monolithic coating on substrate subject to contact loading by a spherical indenter (axysymmetric conditions) was first considered. ABAQUS Standard was used for finite element simulations. Mesh containing 4-noded quadrilateral axysymmetric elements was employed. The smallest element size in the coating thickness direction was 0.25 μm. The nodes of the bottom and left boundaries of the mesh were constrained against displacement in the vertical and horizontal directions respectively. An illustration of the model is shown in Figure 4.2.
The indenter was modeled as rigid with a radius of 250 μm. Contact was established between the indenter and the coating using contact algorithms in ABAQUS with friction coefficient equal to 0.1 between the indenter and the coating. Load control option was considered where the normal load applied to the indenter increased linearly to the maximum prescribed load. A coating thickness of 2μm (= t_B) was considered and the coating was assumed to be homogeneous, isotropic, and perfectly elastic. Deformation plasticity model of ABAQUS was used for substrate. Yield strength of 2000 MPa and hardening exponent of 10 was used. The constitutive model for substrate was chosen to roughly represent 52100 steel and for coating to represent TiN. There is quite a lot of variation in the reported elastic modulus values of TiN in literature. The values range from 300 GPa to 600 GPa [37, 38]. For the current study, elastic modulus of TiN was assumed to be 400 GPa, as reported by Ma et al. [11]. Cohesive elements following bilinear traction separation law (shown in Figure 4.2) were employed at the coating-substrate interface just to monitor damage initiation at the interface. For the interface, shear strength (τ_C in Fig. 2) of 1500 MPa was assumed for cohesive elements at the interface. Although no cohesive elements were employed along the thickness of the coating, peak normal stress in the radial direction (σ_rr) in the coating were monitored. Fracture strength of 3000 MPa for TiN was assumed, i.e., σ_rr=3000 MPa implies that damage initiation takes place in coating. The fracture strength values used in this study were reported by Oettel et al [39]. Material and cohesive properties used in this study are summarized in Table 1. The following damage initiation criterion, Q, for cohesive zone elements following bilinear law at the interface, was used:
where, symbols $\sigma$ and $\tau$ represent normal and shear stresses respectively in cohesive element and $\sigma_C$ and $\tau_C$ represent critical normal and shear stresses respectively in the cohesive elements at interface at which damage initiates (as indicated in Figure 4.3). Obviously, a value that is less than 1.0 for $Q$ indicates that damage criterion has not been satisfied, while a value of 1.0 indicates that the damage criterion has been satisfied. It has to be noted here that during indentation, normal stresses perpendicular to the interface are negligible compared to shear stresses and therefore, the first term on the right side of equation 4.1 drops out.

$$Q = \left( \frac{\langle \sigma \rangle}{\sigma_c} \right)^2 + \left( \frac{\tau}{\tau_c} \right)^2$$  (4.1)
Figure 4.3: Bilinear Law followed by the cohesive elements under shear stress used at the coating-substrate interface. $G_{CI}$ is the energy dissipated due to failure at the interface.

The purpose of having a benchmark model was to establish baseline loading conditions for which damage *just* initiates in a monolithic coating-substrate system. Damage in the coating-substrate system subject to spherical indentation constitutes of through thickness circumferential cracks because of radial tensile stresses and delamination because of interfacial shear stresses. Delamination resulting from tensile stresses in the thickness direction along the interface during unloading of indenter and as pointed out by Abdul-Baqi and Van der Giessen [35] and Xia *et al.* [40] are not considered in this study. Figure 4.4a and 4.3b show the distribution of normal radial stresses along the surface of the coating and shear stresses along the coating-substrate interface respectively plotted against the radial distance from the center of the indenter when the coating substrate system is subject to spherical indentation. The peak stresses for both distributions occur near the edge of the indenter. The loading conditions were chosen such that the maximum normal radial stress in the coating and maximum shear
stress along the coating-substrate interface reach their respective critical fracture values (3000 MPa and 1500 MPa respectively) at end of the loading cycle. To reach the state of when damage just initiates, the maximum indentation load was 5N. Damage initiation criterion, for through thickness crack, $Q_{CB}$, (letter C in the subscript refers to circumferential crack and B refers to benchmark model) was defined as:

$$Q_{CB} = \left( \frac{\sigma}{\sigma_C} \right)^2$$  \hspace{1cm} (4.2)

where $\sigma_C = 3000$ MPa, the fracture strength of coating, and $\sigma$ is the maximum normal radial stress in the coating. Obviously, for the state when the indenter was pushed to the maximum load of 5N, damage initiation criterion for through thickness crack, $Q_{CB} = 1$. Damage initiation criterion, $Q_{IB}$ (letter I in the subscript refers to interface and B refers to benchmark model) for interface crack was monitored via the cohesive elements at the interface. For the state when the indenter was pushed to the maximum load of 5N interface, $Q_{IB} = 1$.

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<tr>
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<tr>
<td></td>
<td>Poisson Ratio</td>
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<tr>
<td>(used for multilayer designs only)</td>
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<td></td>
<td>$G_{CI}(J/m^2)$</td>
<td>100</td>
</tr>
</tbody>
</table>

Table 4.1: Material and cohesive zone properties used in FEM
Figure 4.4: Distribution of stresses during indentation in benchmark model (a) Radial normal stress along coating surface (b) Shear stress along the interface

Critical $\sigma_{rr} = 3000$ MPa

Critical $\tau_{rz} = 1500$ MPa
Once the benchmark model was established and the damage criteria recorded, the model was extended for multilayer coatings keeping loading conditions the same (i.e., peak indentation load = 5N). The constitutive models were chosen to roughly represent coating layers of TiN/Ti deposited on 52100 steel. The multilayer designs to be considered had even number of layers between 2 and 8 (i.e., 2, 4, 6, and 8). In all the multilayer designs, TiN layer was always the topmost layer and Ti layer was always the bottommost layer, just above the substrate. A schematic representation of the multilayer structure is shown in Figure 4.5.

![Schematic of multilayer coating architecture consisting of alternating TiN and Ti layers](image)

**Figure 4.5:** Schematic of multilayer coating architecture consisting of alternating TiN and Ti layers

The criterion for damage initiation in brittle TiN layers of multilayer architecture, \( Q^*_c \) was defined as,

\[
Q^*_{c\text{ multilayer}} = \left( \frac{\sigma_{\text{TiN}}}{\sigma_c} \right)^2
\]

(4.3)
where $\sigma_{\text{TiN}}$ is the maximum normal radial stress in any of the TiN layers. The damage initiation criterion at the interface for multilayer coating architecture, $Q_I^*$, was monitored via the cohesive elements at the interface. It has to be noted here that the maximum shear stress at the interface is limited by the plastic flow in the bottom most Ti layer. For instance, if the Ti layer would have been modeled as elastic-perfect plastic, then the maximum shear stress at the interface would be $0.577\sigma_Y$ where $\sigma_Y$ is the yield stress of Ti. Hence, $Q_I^*$ values will be much smaller than $Q_{IB}$. The initial FEM experiments consisted of fixing the total thickness to 6μm and equal layer thicknesses (e.g., all the layers in the 6 layer architecture would each be 1μm thick) and obtaining the values of $Q_C^*$ and $Q_I^*$. Figure 4.6 and Figure 4.7 show the contour plots of normal stresses in radial direction and shear stresses. It can be seen from Figure 4.6 that normal stresses are highest for 2 layer design and lowest for 8 layers design. Also, normal stresses in Ti layers for all the designs are compressive. Figure 4.8 show the variation of peak normal stress and shear stress with change in number of layers. Next, the top TiN layer thickness was reduced by 0.25 μm and the thickness of Ti layer above substrate was increased by the same amount for all the designs and $Q_C^*$ and $Q_I^*$ were recorded. This process of decreasing the top layer TiN thickness and increasing the bottom Ti layer was repeated again and $Q_C^*$ and $Q_I^*$ were recorded. Figure 4.9 shows the variation of $Q_C^*$ and $Q_I^*$ for above mentioned FEM experiments. It can be seen that $Q_C^*$ and $Q_I^*$ are exhibiting some form of inverse trend and is common to all the multilayer designs. Decreasing the thickness of top layer decreased $Q_C^*$ and increase in Ti layer thickness increased $Q_I^*$ values. Furthermore, both $Q_C^*$ and $Q_I^*$ values decrease with increase in number of layers.
for the same overall thickness. This trend points to adopting a procedure to search for such “Pareto fronts” in an exhaustive space of multilayer architecture designs where there is variation in number of layers, overall thickness and individual layer thickness.

**Figure 4.6:** Contour plots of normal radial stresses for (a) 2 layer, (b) 4 layer, (c) 6 layer and (d) 8 layers
Figure 4.7: Contour plots of shear stresses for (a) 2 layer, (b) 4 layer, (c) 6 layer and (d) 8 layers
Figure 4.8: Variation of maximum radial normal stresses and shear stresses with change in number of layers for the same overall thickness
The following sections state the optimization problem and give details of the experimental designs and the approach to solve the optimization problem.

### 4.3. OPTIMIZATION FORMULATION

A formal statement can be written as:

Minimize \( Q_I^* \) and \( Q_C^* \)

with \( t_1^*, t_2^*, t_3^*, \ldots, t_n^* ; \ n \in \{2,4,6,8\} \) as design variables.
subject to $\Sigma_{i=1}^{n} t_i^* = \beta; \beta \in [1,3]$,

where, $Q_I^*$ and $Q_C^*$ are ‘non dimensionalized’ damage criteria, $t_i^*$ is the non
dimensionalized thickness of the $i^{th}$ layer, and $\beta$ is the non dimensional overall thickness.

Non-dimensionalization is done with respect to the benchmark model, i.e.,

$$Q_I^* = \frac{Q_{I\text{multilayer}}}{Q_{IB}}, \quad Q_C^* = \frac{Q_{C\text{multilayer}}}{Q_{CB}}.$$  

$$t_i^* = \frac{t_i}{t_B},$$

$$\beta = \frac{\Sigma_{i=1}^{n} t_i}{t_B} \quad (4.4)$$

Simply put, the above formulation is to minimize the damage in the multilayer coating
architecture where the total number of layers can be 2, 4, 6, and 8 and the overall
thickness can be in the range 2-6μm.

The optimization formulation defined above clearly is non-trivial as explicit
formulations of the objective functions in terms of design variables are not available.
Hence, design and analysis of computer experiments (DACE) metamodeling techniques
can be used. This approach is described briefly in the following section.

4.4. DESIGN AND ANALYSIS OF COMPUTER EXPERIMENTS (DACE)

APPROACH

Computer models such as FEM are being increasingly used to model actual physical
processes. However, even with modern computational resources, computer modeling can
be time consuming. Also, lack of explicit information of the models with respect to
design variables can limit the predictive nature of the computer models with regards to optimization. Hence, metamodeling (which is essentially modeling of computer models) techniques are gaining popularity. Approaches based on fitting response surfaces to data collected by evaluating objective functions at a few data points are discussed extensively in the literature [42, 43, 44]. The response surfaces models (RSM) are then used to visualize input-output relationships and then estimate the location of the optimum. RSM based on modeling of objective functions with stochastic processes is usually termed ‘Kriging’. Appendix A briefly discusses the structure of Kriging models and how they can be used for prediction of responses.

To employ Kriging models, an experimental design matrix needed to be developed. For efficient Kriging models, the experimental design space needs to be (a) space-filling, and (b) non-collapsive. [45]. Space-fillingness essentially means that it important to get information from the entire design space, i.e., experimental design points should be well spread-out over the design space. There are several methods to created space filling designs such as Latin hypercube designs, orthogonal arrays, mean squared error designs, minimax and maximin designs. Maximin, follows on maximizing the minimum distance between the design points in the Euclidean space. Non–collapsive designs essentially ensure that design points will not take same value in any of the dimensions. This is because computer experiments are deterministic.

Initial computer experimental designs for 2, 4, 6, and 8 layers multilayer coating architecture were created as follows:

(a) Random assignment of total thickness for a design point.
(b) Given the total thickness, a set of “L” randomly selected layer thicknesses adding to total thickness are selected, i.e., coordinates of a design point are $(x_1, x_2, \ldots, x_L)$.

(c) Repeat steps a and b to give NL design points.

(d) Repeat a, b, and c 10,000 times and select the “best design” based on the criteria to minimize Euclidean distance in the "L" dimension space, and minimize geometric average Euclidean distance in all the projected 2-dimensional space.

(e) The layer thickness values were not continuous but based on a grid of 0.25 μm, i.e., minimum thickness of layers was 0.25 μm and thickness values were a multiple of 0.25. This is because the smallest element size for the mesh of the finite element model was 0.25 μm.

The above methodology of creating experimental designs roughly follows the maximin criteria. Figure 4.10 shows the experimental designs for a 2 layer coating architecture. A total of 10, 14, 27, and 40 initial experimental designs were created. $Q_i^*$ and $\sigma_{TiN}$ were evaluated for the multilayer experimental designs using the finite element model. The evaluated $Q_i^*$ and $\sigma_{TiN}$ values were used as input values to the kriging model. The next section discusses the results from the kriging model.
4.4. RESULTS & DISCUSSION

The parametric kriging model using a cubic correlation function, as described above, was used to predict damage initiation parameter for the coating-substrate interface, $Q_{i*}$ and maximum radial stress in TiN layers, $\sigma_{TiN}$, for 2, 4, 6, and 8 layers multilayer coating architecture. This section discusses uses how the kriging model was used, validated by additional experiments and then updated based on the results from additional experiments.

**Figure 4.10:** Design coordinates for 2 layers
Figure 4.11: Comparison of FEM and Kriging model prediction (a) stress in TiN layers, (b) Damage initiation parameter at interface for 4 layers designs
Computer experiments for 14 initial designs were carried out using the finite element model. $Q_I^*$ and $\sigma_{TiN}$ were evaluated for each experiment and were used as inputs for the kriging models. Kriging models were then used to predict $Q_I^*$ and $\sigma_{TiN}$ and the associated standard errors were recorded for 10,615 designs consisting of various combination of layer thickness and total thicknesses. 10 additional designs were chosen from the design space and computer experiments using the finite element model were run for those designs and were compared with the predicted results from the kriging models. Figure 4.11a and Figure 4.11b illustrate the comparisons.
Figure 4.12: Comparison of FEM and updated Kriging model prediction (a) stress in TiN layers, (b) Damage initiation parameter at interface for 4 layers designs

It can be seen that the predictions from the kriging models do not compare that well with those predicted by FEM. The kriging models were then run again with 10 additional inputs along with 14 initial inputs from the computer experiments and the updated predictions and the associated standard errors were recorded. 14 additional designs were chosen from the design space and then computer experiments using FEM were run for those designs and were compared with the predicted results from the kriging models. Figure 4.12a and Figure 4.12b show the comparisons and can be seen that the updated predictions made by the kriging model are reasonably close to the FEM
predictions. Kriging models were re-run with a total of 38 input points. The parameters associated with the kriging models for prediction of $Q_I^*$ and $\sigma_{TIN}$ are listed in Table 4.2.

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<th>Layer No. (from top)</th>
<th>$\Theta_i$ (i=1,2, 3,4) For $\sigma_{TIN}$</th>
<th>$\Theta_i$ (i=1,2, 3,4) For $Q_I^*$</th>
<th>Log likelihood For $\sigma_{TIN}$</th>
<th>Log likelihood For $Q_I^*$</th>
<th>$\beta_0$ (Avg $\sigma_{TIN}$)</th>
<th>$\beta_0$ (Avg $Q_I^*$)</th>
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**Table 4.2: Kriging model parameters for 4 layer designs**

The initial input space for 6 layers designs consisted of 27 designs and the kriging models were run to predict $Q_I^*$ and $\sigma_{TIN}$ for 10,685 designs. Kriging models were compared with FEM predictions and updated with 10 additional designs and then later for 5 more designs. The respective comparisons are shown in Figure 4.13a, Figure 4.13b, Figure 4.14a and Figure 4.14b.
Figure 4.13: Comparison of FEM and Kriging model prediction (a) stress in TiN layers, (b) Damage initiation parameter at interface for 6 layers designs
Figure 4.14: Comparison of FEM and updated Kriging model prediction (a) stress in TiN layers, (b) Damage initiation parameter at interface for 6 layers designs
Kriging models were re-run with a total of 42 input points for 6 layers designs. The parameters associated with the kriging models for prediction of $Q_I^*$ and $\sigma_{TiN}$ for 6 layers designs are listed in Table 4.3. The procedure was repeated for 8 layers designs with initial input of 40 designs, comparison and update with 15 additional designs and then later for 10 additional designs. The comparisons are shown in Figure 4.15a, Figure 4.15b, Figure 4.16a, and Figure 4.16b. Kriging models were used to predict for a total of 8098 designs of 8 layers.

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<th>Log likelihood For $Q_I^*$</th>
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</tbody>
</table>

Table 4.3: Kriging model parameters for 6 layer designs
Figure 4.15: Comparison of FEM and Kriging model prediction (a) stress in TiN layers, (b) Damage initiation parameter at interface for 8 layers designs
Figure 4.16: Comparison of FEM and updated Kriging model prediction (a) stress in TiN layers, (b) Damage initiation parameter at interface for 8 layers designs
The parameters associated with the kriging models for prediction of $Q_I^*$ and $\sigma_{TiN}$ for 8 layers designs are listed in Table 4.4.

<table>
<thead>
<tr>
<th>Layer No. (from top)</th>
<th>$\Theta_i$ (i=1,2,..,8) For $\sigma_{TiN}$</th>
<th>$\Theta_i$ (i=1,2,..,8) For $Q_I^*$</th>
<th>Log likelihood For $\sigma_{TiN}$</th>
<th>Log likelihood For $Q_I^*$</th>
<th>$\beta_0$ (Avg $\sigma_{TiN}$)</th>
<th>$\beta_0$ (Avg $Q_I^*$)</th>
<th>$\sigma^2$ (variance for $\sigma_{TiN}$)</th>
<th>$\sigma^2$ (variance for $Q_I^*$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2.3198</td>
<td>8.0853</td>
<td>-398.72</td>
<td>342.19</td>
<td>2210.6</td>
<td>0.110</td>
<td>6.45e+00</td>
<td>1.27e-04</td>
</tr>
<tr>
<td>2</td>
<td>2.0995</td>
<td>3.4124</td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>3.0466</td>
<td>10.2191</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>2.8127</td>
<td>3.9017</td>
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<tr>
<td>5</td>
<td>3.1112</td>
<td>12.1745</td>
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<td></td>
<td></td>
<td></td>
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<td></td>
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<tr>
<td>6</td>
<td>4.2910</td>
<td>2.9394</td>
<td></td>
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<td></td>
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<tr>
<td>7</td>
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<td>4.9058</td>
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<tr>
<td>8</td>
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<td>1.7476</td>
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</tbody>
</table>

**Table 4.4:** Kriging model parameters for 8 layer designs

For 2 layer designs, Kriging model was used with initial input of 10 designs. The fitted response was then compared for 2 additional designs, as shown in Figure 4.17a and Figure 4.17b. The Kriging model was run again with 12 input designs. The model was compared for 3 additional designs as shown in Figure 4.18a and Figure 4.18b. Kriging models were used to predict for a total of 265 designs of 2 layers. The parameters associated with the kriging models for prediction of $Q_I^*$ and $\sigma_{TiN}$ for 2 layers designs are listed in Table 4.5.
<table>
<thead>
<tr>
<th>Layer No. (from top)</th>
<th>$\Theta_i$ (i=1, 2) For $\sigma_{TN}$</th>
<th>$\Theta_i$ (i=1, 2) For $Q_i^*$</th>
<th>Log likelihood For $\sigma_{TN}$</th>
<th>Log likelihood For $Q_i^*$</th>
<th>$\beta_0$ (Avg $\sigma_{TN}$)</th>
<th>$\beta_0$ (Avg $Q_i^*$)</th>
<th>$\sigma^2$ (variance for $\sigma_{TN}$)</th>
<th>$\sigma^2$ (variance for $Q_i^*$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>6.977</td>
<td>22.256</td>
<td>-71.51</td>
<td>42.91</td>
<td>2699.8</td>
<td>0.157</td>
<td>2.65e+06</td>
<td>9.67e-04</td>
</tr>
<tr>
<td>2</td>
<td>5.744</td>
<td>1.4406</td>
<td></td>
<td></td>
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</table>

**Table 4.5:** Kriging model parameters for 2 layer designs

![Diagram](attachment:image.png)
Figure 4.17: Comparison of FEM and Kriging model prediction (a) stress in TiN layers, (b) Damage initiation parameter at interface for 2 layers designs
Figure 4.18: Comparison of FEM and updated Kriging model prediction (a) stress in TiN layers, (b) Damage initiation parameter at interface for 2 layers designs
The predicted $\sigma_{TiN}$ were then used to calculate $Q_c^*$ using equations 2 and 3 and were plotted against $Q_i^*$ as for all the layers designs. Pareto optimum points were identified for 2, 4, 6, and 8 layers designs and are shown in Figure 4.19, Figure 4.20, Figure 4.21, Figure 4.22. Figure 4.23 shows the Pareto frontiers of all the multilayer designs with the scale of $Q_c^*$ reduced to 0.1 for effective comparisons. The Pareto optimal frontier with respect to interfacial and cohesive failure of coatings presents a map for coating architecture design. It can be seen from Figure 4.23 that 2 layer designs exhibit the worst performance in terms of interface delamination initiation parameter, $Q_i^*$. Also, it can be seen from Tables 4.2-4.5, that the average value of $Q_i^*$, is decreasing with increase in number of layers, suggesting that coating architecture with more number of layers are less susceptible for delamination. The trend is similar with respect to the average maximum stress in TiN layers as seen in tables 4.2-4.5. The Pareto frontiers for cases of 2, 4, 6, and 8 layers can also given an idea of the tradeoffs associated from one frontier to another frontier, if the cost of production additional layers is known.

Some of the Pareto design points indicated by Kriging model were analyzed. All the designs exhibiting low $Q_c^*$ (less than 0.1) had the thinnest top TiN layer (0.25 microns). For instance, some design points in 6 layers and 8 layers architecture were picked for which predicted $Q_i^*$ was less than 0.11. Two such candidate points in 6 layers design had layer thickness values starting from top TiN layer as 0.25, 1.5, 1, 0.5, 0.75, 0.5, and 0.25, 0.25, 0.25, 1.75, 0.25, 0.25 respectively. $Q_i^*$ and $Q_c^*$ for the first design,
as predicted by Kriging model were 0.1003 and 0.0065 respectively. FEM results for the same design resulted in $Q_l^*$ and $Q_c^*$ values of 0.1014 and 0.0062 respectively. $Q_l^*$ and $Q_c^*$ for the first design, as predicted by Kriging model were 0.095 and 0.045 respectively while the predicted FEM values were 0.098 and 0.062. For instance, just based on the mean value of predictions and looking at Figure 4.23, it would appear that selecting a design with 8 layers may be the safest option. However, the selection process may also consider the difference in cost of production between 8 layers and 4 layers. The role of defects in coating and their influence on damage variables also play a role in the decision process. The investigation of role of defects remains topic for future research.

![Figure 4.19](image.png)

**Figure 4.19:** Predicted damage initiation variables and pareto frontier for 2 layers. The encircled points correspond to Figure 4.9
Figure 4.20: Predicted damage initiation variables and pareto frontier for 4 layers. The encircled points correspond to Figure 4.9
Figure 4.21: Predicted damage initiation variables and pareto frontier for 6 layers. The big red points correspond to Figure 4.9
Figure 4.22: Predicted damage initiation variables and pareto frontier for 8 layers. The big red points correspond to Figure 4.9
In this study, response of multilayer coating architecture to spherical indentation was considered by the use of finite element models. A benchmark model was first considered consisting of 2 μm thick monolithic coating (TiN) on ductile substrate (52100 steel). Loading condition were chosen such that it would initiate through thickness and interfacial fracture. Damage variables indicating these two fracture modes were
quantified. Finite element model was extended to multilayer coating architecture consisting of 2, 4, 6, and 8 alternating hard (TiN) and soft (Ti) layers on substrate. Loading conditions were kept same as that for the benchmark case. Design of computer experiments and a metamodel was established with the objective to minimize damage in multilayer coating architecture. Pareto frontiers were established between the variables responsible for though thickness and interface fracture were established. It was found that 2 layer designs were more susceptible for interfacial delamination. It is believed that pareto frontiers will help designers to choose the number of layers according to application area and the tradeoffs associated with respect to cost of additional layers.
REFERENCES


CHAPTER 5

SUMMARY & FUTURE WORK

5.1. SUMMARY

Hard coating-substrate systems were characterized through experiments, numerical modeling, and optimization. The experiments consisted of tensile cracking of the coated specimens. During the experiments, the acoustic emission technique was used to monitor formation of parallel cracks. A 2D finite element model was created to simulate the response of a coating-interlayer-substrate system to in-plane uniaxial tension. Coating cracking was simulated with cohesive zone elements that followed a bilinear cohesive law. Some degree of randomness was introduced into the model through the assignment of random critical cohesive stresses. Due to the unsymmetrical boundary conditions at the top surface and coating-interlayer interface, bending occurred and introduced non-uniform stresses through the coating thickness. Simulations showed coating cracks propagating from the interface and often being arrested near the surface of the coating by the presence of high compressive stresses. The effects of different coating modulus, tangent modulus for the interlayer hardening, and critical stress values \( \sigma_C \) of the
cohesive zone elements were studied. Crack spacing distributions for different parameter changes were quantified and compared to an experimental crack spacing distribution.

Due to the limitations of using a 2D model, crack formation and propagation at high plastic strain values was not predicted accurately. Incorporating measured residual stresses into the model is necessary for more realistic simulations. The model has been shown to be sensitive to material and cohesive parameters and could thus be used to optimize coatings. The described tensile cracking experiment could be used to calibrate cohesive parameters for subsequent modeling of more complicated loading schemes.

Finite element models were used to evaluate the response of multilayer coating architectures to spherical indentation. A benchmark model was first considered consisting of a 2 μm thick monolithic coating (TiN) on a ductile substrate (52100 steel). Loading conditions were chosen such that through thickness and interfacial fractures initiate. Damage variables indicating these two fracture modes were quantified. The finite element model was extended to multilayer coating architectures consisting of 2, 4, 6, and 8 alternating hard (TiN) and soft (Ti) layers on the substrate. Loading conditions were kept the same as that for the benchmark case. Design of computer experiments and a metamodel was established with the objective to minimize damage in the multilayer coating architecture. Pareto frontiers were established between the variables responsible for though thickness and interface fracture were established. It was found that 2 layer designs were more susceptible to interfacial delamination. It is believed that Pareto frontiers will help designers to choose the optimal number of layers according to application area and to account for tradeoffs associated with the cost of additional layers.
5.2. FUTURE WORK

Though it is believed that they play an important role in determining damage parameters, in this study residual stresses were not considered in the finite element models. For instance, hard coatings like TiN exhibit high intrinsic stresses due to deposition process and thermal mismatch conditions, and due to defect growth mechanisms. These intrinsic stresses are mostly compressive in nature. As a result of these stresses, it is difficult to attain good adhesion between the coating and substrate. Introduction of a Ti layer between coating and substrate seems to improve adhesion because of the accommodation of TiN internal stresses. However, as Bemporad et al. [1] point out, there is a need for optimum Ti layer thickness to accommodate both intrinsic stresses and stresses from service conditions to avoid fracture of the coating. To produce more realistic simulations, finite element models need to be developed to account for thickness dependent intrinsic stress distributions in the multilayer coating architecture.

In the models developed in this study, coatings were assumed to be defect free. However, as shown in Figure 2.7, earlier, defects in the form of macroparticles and pits are commonly observed phenomena in PVD coatings. The role of defects in coating also needs to be included in the computational model. For instance, to simulate the presence of defects by virtue of macroparticles and pinholes, cohesive elements along the thickness of the coating can be employed in the finite element model. The defects can be characterized by their size and their location. Several studies have shown experimentally observed macroparticle size distribution on the surface of coating [2, 3]. Figure 5.1
shows such a distribution. The distribution of defects can then be incorporated in the finite element model and a Monte Carlo approach can be used to study the coating damage evolution. It is believed that the success with these finite element simulations will enable more relevant simulations of actual wear processes. Only when the models required to form a strong basis for optimal design for coatings systems are developed will the prospect of achieving even greater progress in the understanding and use of tribological coatings be realized.

Figure 5.1: Macroparticle size distribution in cathodic–arc-evaporated TiN coatings for different nitrogen partial pressure. Increment in diameters is 0.25 microns [2]
REFERENCES


APPENDIX A

Stochastic process model

Consider a function of k variables evaluated at n points. A sample point i can be denoted by

\[ \mathbf{x}^{(i)} = (x_1^{(i)}, \ldots, x_k^{(i)}) \]

Let the corresponding function be denoted by

\[ y^{(i)} = y(\mathbf{x}^{(i)}); \quad i = 1, \ldots, n \]

A response surface can be fit by linear regression using the model:

\[ y(\mathbf{x}^{(i)}) = \sum_h \beta_h f_h (\mathbf{x}^{(i)}) + \epsilon^{(i)}; \quad i = 1, \ldots, n \]  \hspace{1cm} (A1)

where, \( \epsilon^{(i)} \)'s are normally distributed independent error terms mean zero and variance \( \sigma^2 \).

However, for deterministic computer experiments, \( \epsilon^{(i)} \)'s cannot be independent. The lack of fit is entirely due to modeling error. Hence, we may write \( \epsilon^{(i)} \) as \( \epsilon(\mathbf{x}^{(i)}) \). In a Gaussian stochastic process approach, instead of assuming that the errors are independent, they are assumed to be correlated and the correlation is related to the distance between the points,

\[ \text{Corr}[\epsilon(\mathbf{x}^{(i)}), \epsilon(\mathbf{x}^{(j)})] = \exp[-d(\mathbf{x}^{(i)}, \mathbf{x}^{(j)})] \]  \hspace{1cm} (A2)

where, \( d \) is a weighted distance between points given by:

\[ d(\mathbf{x}^{(i)}, \mathbf{x}^{(j)}) = \sum_{h=1}^{k} \theta_h |x_h^{(i)} - x_h^{(j)}|^2 \]  \hspace{1cm} (A3)

The stochastic process model can be re-written as:
\[ y(x^{(i)}) = \beta_0 + \epsilon(x^{(i)}); \ i = 1, \ldots, n \]  \hspace{1cm} (A4)

where, \( \beta_0 \) is the mean of the stochastic process model, and \( \epsilon(x^{(i)}) \) is \( \text{N}(0, \sigma^2) \) with correlations given by equations 2 and 3. For the cubic correlation function which has been used for the current investigation

\[
\text{Corr}[\epsilon(x^{(i)}), \epsilon(x^{(j)})] = \sigma^2 R(x^{(i)} - x^{(j)} | \xi)
\]

where,

\[
R(h|\xi) = \prod_{i=1}^{n} R(h_i | \theta_i)
\]

\[
R(h_i | \theta_i) = \begin{cases} 
1 - 6\left(\frac{h_i}{\theta_i}\right)^2 + \frac{6|h_i|^3}{\theta_i^3}, & \text{if } |h_i| \leq \theta_i/2 \\
2(1 - \frac{|h_i|^2}{\theta_i^2}), & \text{if } \theta_i/2 \leq |h_i| \leq \theta_i \\
0 & \text{if } \theta \leq |h_i| 
\end{cases}
\]  \hspace{1cm} (A5)

Let \( y = (y^{(1)}, \ldots, y^{(n)})' \) denote the vector of observed function values, and \( R \) denote the \( n \times n \) matrix whose \((i,j)\) element is \( \text{Corr}[\epsilon(x^{(i)}), \epsilon(x^{(j)})] \). The likelihood function is given by:

\[
\frac{1}{(2\pi)^{n/2}(\sigma^2)^{n/2}|R|^{1/2}} \exp \left[ -\frac{(y - \mu)^' R^{-1} (y - \mu)}{2\sigma^2} \right]
\]  \hspace{1cm} (A6)

The parameters \( \beta_0, \sigma^2, \theta_1, \ldots, \theta_2 \) are estimated by choosing them to maximize the likelihood of the sample (i.e., differentiate the likelihood with respect to the parameters and set it to zero).

**REFERENCE**

Chapter 2


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**Chapter 3**


**Chapter 4**


Chapter 5

