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预镀层金属薄板微细成形的膜层断裂机制
与多尺度建模研究

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**MECHANISM AND MULTISCALE MODELING OF
COATING FRACTURE IN MICRO-FORMING OF
COATED METALLIC SHEET**

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摘要

质子交换膜燃料电池是一种零污染、高效率的新能源装置，其中金属双极板通过微细成形的流道结构承担着多种重要功能，且需要通过表面真空镀膜保证其导电性和耐蚀性。预镀层技术通过对金属薄板基底进行高效镀膜后直接成形，可以显著地提高制造效率、降低成本。但是，预镀层成形后易现微裂纹、不锈钢薄板基底暴露腐蚀，严重影响其使用性能。由于膜层厚度小于 $1\ \mu\text{m}$ 、一般为纳米晶或非晶结构，与金属薄板基底存在几何尺度和力学性能差异，膜层断裂行为受到金属薄板基底塑性变形的影响，预镀层金属薄板中的膜层断裂机制与传统均质材料存在显著区别，其断裂行为复杂。对预镀层金属薄板进行微细成形时，膜层的断裂与金属薄板基底微观尺度的不均匀塑性变形关联，建立考虑基底微观结构影响的膜层断裂预测模型面临挑战，现有理论难以有效指导预镀层金属双极板微细成形工艺的设计，制约其性能提升和应用。

本文针对预镀层金属薄板塑性变形时的膜层断裂规律进行了实验表征和理论建模研究。在不锈钢薄板基底上制备了多种金属膜层，探究了膜层材料、厚度和组织影响下的膜层断裂行为；揭示了膜层断裂与基底变形之间的对应关系，阐明了基底组织、界面变形和膜层晶粒对膜层断裂的作用机制；建立了金属基底上膜层断裂的多尺度力学仿真模型，准确预测了基底微观塑性变形诱导的膜层断裂；基于多尺度模型计算的膜层断裂规律，发展了预镀层金属薄板微流道成形的全场仿真方法，为高性能预镀层双极板的微细成形工艺提供了设计指导。本文的研究成果主要包括以下几个方面：

1) 金属薄板基底上的膜层断裂机制

金属薄板基底上的膜层断裂同时受到基底微观结构和膜层力学特性的影响，为研究由此引发的复杂断裂机制转变，在 316L 不锈钢薄板上制备了 100~1000 nm 厚度的铌、铜、高熵合金膜层，探究了单轴拉伸时不同膜层的裂纹形貌及演化规律，设计了关联膜层裂纹与基底微观结构的表征方法，揭示了基底微观非均匀塑性变形诱导的膜层断裂机制。研究表明，膜层厚度 100 nm 时，其断裂均由金属基底的微观塑性变形诱导，膜层裂纹对应于基底的晶粒滑移线、晶界、孪晶界，其中基底晶粒滑移线对应的裂纹占 90% 以上；随着膜层厚度增加至 500 nm 以上，非晶膜层从基底塑性变形诱导的断裂机制转变为脆性通道开裂，而纳米晶膜层仍保持基底塑性变形诱导的断裂机制，且晶粒粗化导致断裂应变提升，由剪切断裂转变为颈缩断裂。

2) 金属基底塑性变形诱导膜层断裂的判断准则

金属基底的微观塑性变形诱导的膜层断裂难以采用宏观准则判断。因此，对膜层

裂纹对应的基底微观结构进行了晶体学统计分析,采用分子模拟探究了膜层与基底界面处的耦合变形和纳米晶膜层的裂纹扩展响应,提出了基于基底微观结构特征的膜层断裂判断准则。结果表明,膜层厚度 100 nm 时,基底晶粒的尺寸大于 5 μm 且滑移方向与表面夹角超过 30°时,易诱导膜层剪切断裂;基底晶界上的膜层断裂则通过晶界的变形相容性判断,相容因子低于 0.5 的基底晶界易产生应变集中进而诱导膜层断裂。纳米晶膜层的厚度增加时,晶粒粗化提升裂纹尖端的位错发射能力,断裂韧性增强;然而,由于膜层与基底界面附近显著的应力集中,316L 不锈钢基底中{111}和{110}取向的晶粒易形成变形孪晶,沿孪晶界产生更高的滑移台阶并诱导膜层断裂。

3) 预镀层金属薄板的多尺度断裂预测建模

传统膜层断裂模型缺乏对金属基底微观非均匀塑性变形的描述,无法准确预测基底变形诱导的膜层裂纹形成和演化。因此,建立了金属基底上膜层断裂的多尺度力学仿真模型:纳米尺度上,通过分子模拟计算了膜层内聚区模型的断裂能量,传递到微观尺度;微观尺度上,开发了结合膜层扩展有限元和基底亚晶粒滑移带的代表性体积单元,计算了膜层的剪切断裂演化;宏观尺度上,将膜层的微观裂纹演化转化为失效参数,并通过晶体塑性描述金属基底晶粒的非均匀变形,从而准确预测了实际样品中的膜层断裂,精度相比传统模型显著提升。分析发现,基底晶粒的滑移方向与表面夹角为 30 至 50° 时,膜层发生基底变形诱导的剪切断裂,与实验结果一致。

4) 预镀层金属双极板的微流道成形工艺优化

现有微细成形理论无法同时预测预镀层金属薄板的宏观成形质量和微观膜层开裂,难以有效指导其成形工艺的开发。因此,对基底采用局部晶体塑性建模,结合膜层裂纹演化的多尺度计算结果,建立了可以准确预测膜层断裂的预镀层金属薄板全场成形仿真方法,并结合实验结果论证了模型的有效性。基于数值计算的结果,分析了成形参数对膜层断裂和成形轮廓的影响规律,建立了相应的工艺窗口,为高性能预镀层双极板的微流道成形工艺设计提供了理论指导。通过优化成形参数,微流道圆角区域的膜层裂纹减少 50%,有效避免了膜层断裂引起的耐蚀性能劣化。

为准确预测并避免预镀层双极板成形时的膜层断裂,本文围绕金属薄板基底上的膜层断裂行为开展了详细研究,通过系统的实验表征阐明了膜层的断裂机制及其转变,建立了膜层、基底和界面的微观力学响应到宏观膜层断裂的多尺度仿真模型,为预镀层金属薄板中复杂的力学行为和高性能微细结构的制造工艺提供了实验和理论基础。

关键词: 微细成形, 预镀层金属薄板, 膜层断裂机制, 金属基底塑性, 多尺度建模

Abstract

Proton exchange membrane fuel cells are zero-pollution and high-efficiency new energy devices, in which metallic bipolar plates (BPPs) perform multiple critical functions with the formed micro-channels and require surface coating to ensure the conductivity and corrosion resistance. Pre-coating technology can significantly improve the manufacturing efficiency of metallic BPPs and reduce the costs, where stainless steel sheets are efficiently coated and then directly formed. However, the pre-coatings develop micro-cracks during forming that expose the stainless steel substrate, degrading the corrosion performance. In coated metallic sheets, nanocrystalline/amorphous coatings with thickness $<1\ \mu\text{m}$ exhibit notable geometric and mechanical mismatches with metallic sheets, and the coating fractures are influenced by plastic deformation of the substrate. Therefore, the coating fractures on metallic sheets differ fundamentally from those of homogeneous bulk, yet the underlying mechanism remains to be elucidated. During forming, coating cracks are correlated with microscopic deformation of the metallic sheet. Therefore, it is challenging to establish prediction models for coating cracks associated with substrate microstructure, and design guidance for the forming process of pre-coated BPPs remains lacking, limiting their performance and application.

In this study, the coating fracture behaviors of coated metallic sheets were characterized and modeled. Metallic coatings of different materials, thicknesses, and microstructures were deposited on stainless steel sheets, and their fracture mechanisms were thoroughly clarified. Coating cracks were correlated with substrate deformation, and governing factors in coating fracture were revealed, including substrate microstructures, interfacial deformation, and coating grains. A multiscale model was developed, accurately predicting the coating cracks induced by substrate deformation. Based on the calculated crack evolution, a full-field simulation approach was established to analyze coating cracking in the micro-channel forming of coated metallic sheets, and design guidance was provided for the forming process of high-performance pre-coated BPPs. The key research contents are summarized as follows:

1) Mechanism analysis of coating fracture on metallic sheet substrate

Coating fractures on metallic sheet substrates are influenced by both the substrate microstructure and the mechanical properties of the coating. To clarify the induced complex transitions of coating fracture mechanisms, niobium, copper, and high-entropy alloy coatings

with thicknesses of 100-1000 nm were deposited on 316L stainless steel (SS316L) sheets, and their crack morphologies and evolutions during uniaxial tension were investigated. An integrated characterization method was designed to correlate coating cracks with substrate microstructures, revealing coating fracture induced by microscopic substrate deformation. When the coating thickness is 100 nm, all the cracks are caused by inhomogeneous plastic deformation of different substrate microstructures. Coating cracks initiate on intragranular slip traces, grain boundaries, and twin boundaries of the substrate, with over 90% of cracks attributed to substrate slip traces. With increasing coating thickness to >500 nm, the fracture mechanisms of amorphous coatings transition from substrate-deformation-induced to brittle channeling. In contrast, the nanocrystalline coating fractures remain induced by substrate deformation, with an increased fracture strain due to the grain coarsening, and the dominant fracture mechanism changes from shear fracture to necking fracture.

2) Criteria for coating fracture induced by substrate deformation

The microscopic deformation of the metallic substrate induces coating fracture, which is difficult to determine using macroscopic criteria. Consequently, the substrate microstructure inducing coating fracture was analyzed statistically with crystallographic calculations, and molecular dynamics (MD) simulations were used to explore the deformation at the interface between coating and substrate and the crack propagation in nanocrystalline coatings. The coating fracture criteria based on substrate microstructures were proposed. When the coating thickness is 100 nm, substrate grains >5 μm with slip directions >30° to the surface lead to coating shear fracture, while coating cracks on substrate grain boundaries can be determined by deformation compatibility of the grain boundary, with those exhibiting a compatibility factor <0.5 being prone to strain localization and inducing coating fracture. As the coating thickness increases, the larger nano-grains promote the dislocation emission at the crack tip, thereby improving the fracture toughness. However, the severe stress concentration near the interface enhances deformation twinning in {111} and {110} orientated grains in the SS316L substrate, and the resultant higher slip steps along the twin boundary induce coating cracking.

3) Multiscale modeling for coating cracking prediction in coated metallic sheet

Due to the lack of description for microscopic inhomogeneous deformation in metallic substrates, conventional models fail to predict the formation and evolution of coating cracks induced by substrate deformation, and a multiscale mechanical model was thus established.

Derived from nanoscale MD simulations, the coating fracture energy of the cohesive zone model was migrated to a microscale representative volume element (RVE), where a substrate slip band model at the sub-grain scale was developed and combined with the extended finite element method to calculate the coating shear fracture. The obtained microscopic crack evolutions were migrated as a macroscopic failure parameter in the coating, while necking strain is derived using the bifurcation theory. Finally, combined with the crystal plasticity (CP) model of metallic substrates to describe the grain-scale deformation, we can accurately predict all coating cracks in practical samples, significantly improving over conventional models. The calculation results reveal that coating shear fractures are attributed to substrate slips oriented 30-50° relative to the surface, consistent with experimental analysis.

4) Optimization of the micro-forming process of pre-coated metallic BPPs

The existing micro-forming theory cannot simultaneously predict macroscopic forming quality and microscopic coating fracture, and lacks the forming guidance for coated metallic sheets. Therefore, the full-field forming simulation model with accurate prediction of coating cracks was established, utilizing local CP RVE in the substrate and the failure parameters of the coating. The applicability was validated with experimental results, and the influences of forming parameters on microscopic coating fracture and macroscopic forming profiles were analyzed based on the numerical simulations. The corresponding process windows were obtained to guide the micro-channel forming process of high-performance pre-coated BPPs. After optimization of the forming parameters, the coating crack density was decreased by 50%, and the deterioration of corrosion resistance was successfully minimized.

To accurately predict and avoid coating cracking during the forming of pre-coated BPPs, this work systematically investigated the coating fracture behaviors on metallic substrates. The transitions of coating fracture mechanisms were clarified, and a multiscale modeling framework was proposed, bridging microscopic mechanical responses of coating, substrate, and interface with macroscopic coating fractures. These findings provide experimental and theoretical insights into the complicated mechanical behaviors in coated metallic sheets and their engineered manufacturing for high-performance micro-components.

Key words: micro-forming process, coated metallic sheet, coating fracture mechanism, metallic substrate plasticity, multiscale modeling