

Converting Molecular Layer Deposited alucone films into Al₂O₃/alucone hybrid multilayers by plasma densification.

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Alucones are one of the best-known films in the Molecular Layer Deposition (MLD) field, mainly due to their flexibility, but their stability in air is a concern. On the other hand, Al₂O₃ offers very low water vapour transmission rates (WVTRs), at the expense of strain sensitivity even for thin layers. As a consequence, alucone/Al₂O₃ nanolaminates withhold potential for synergistic behaviour. The impermeability of Al₂O₃ will impede moisture from entering the stack and, when it enters, it will encounter an alucone structure where it will absorb. Therefore, these nanolaminates can create a very tortuous path for water molecules and may lower the effective diffusion constant to the substrate.

In this work, we prove that alucone/Al₂O₃ nanolaminate synthesis can be successfully performed by alternating alucone MLD growth with static O₂ plasma exposures. Upon plasma treatment, only the top part of the alucone is densified into Al₂O₃, while the rest of the film remains relatively unaltered. X-ray reflectivity (XRR) and x-ray photoelectron spectroscopy (XPS) depth profiling show that the process yields a bilayer structure, which remains stable in air. Fourier-transform infrared spectroscopy (FTIR) measurements show that Al₂O₃ features are generated after plasma treatment, while the original alucone features remain, confirming that plasma treatment results in a bilayer structure. Also, an intermediate carboxylate is created near the interface. Calculations of Al atom density during plasma exposure point towards a partial loss of Al atoms during plasma treatment, in addition to the removal of the glycerol backbone.

The effect of different process parameters has been studied. Densification at the highest temperature possible (200°C) has the best alucone preservation without hindering its thermal stability. In addition, operating at the lowest plasma power is found the most beneficial for the film, but there is a threshold that must be surpassed to achieve successful densification. About 70% of the original alucone film thickness can be expected to remain after densification, but thicker films may result in more diffuse interfaces. Additionally, this process has also been successfully performed in multilayers, showing potential for encapsulation applications.

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