Abstract

The present study followed a comprehensive experimental approach to reveal the mechanisms of ion transport processes at polymer/oxide/metal interfaces, to evaluate determinant parameters for the progress of corrosive de-adhesion and to address them to the so called `electrolyte front position´. This could be achieved after probing of the interface stability by initiation of cathodic or combined cathodic/anodic delamination and monitoring its progress in particular with the height-regulated Scanning Kelvin Probe. The polymer/substrate interface design could be specifically adjusted by variation of the substrate material, in particular zinc and iron samples or steel substrates with distinct passive films. It was also focussed on the influence of the interfacial contact between adhesive layer and nano-rough oxide surface as well as the barrier properties of latex copolymer films of different monomer composition, glass transition temperature and polymer network density. Polymer/substrate interface potentials were compared to resulting ion distributions after interfacial ion transport processes applying X-ray Photoelectron Spectroscopy and Time-of-Flight Secondary Ion Mass Spectrometry. It could be shown that ions do not diffuse along the interfaces. In fact, ion migration in an electrostatic field is dominant even in extremely oxygen deficient humid atmosphere. Reactive electrolyte spreading along uncoated oxide surfaces can be applied to simulate corrosive delamination processes without the complex polymer/ substrate interface structure. It was proven that electrochemical interface degradation precedes a macroscopic polymer de-adhesion. The progress of delamination strongly responded to the available interfacial water activity. Increased hydrostatic pressure of the defect electrolyte accelerated the interfacial ion transport, but electrochemical stimulation turned out to be an even more effective tool to address corrosive de-adhesion kinetics. Nevertheless they were also found to end up with self-inhibition at reduced atmospheric moisture; the relevant lateral dimensions of the electrolyte front thereby could be confined to cover less than 250 µm. The polar surface energy component turned out to be a suitable parameter to estimate resulting polymer/ iron oxide adhesion forces and can be applied to predict rates of subsequently initiated delamination processes. Reactive electrolyte spreading along uncoated iron surfaces in contrast is not determined by the respective water contact angles and surface energies. Flexible and elastic polymers with a glass transition temperature below ambient temperature resulted in reduced pore densities, which could be evaluated by Electrochemical Impedance Spectroscopy. Such films obviously also sealed interfacial free volumes and inhibited corrosive delamination. An advantageous effect of reduced averaged polymer chain lengths could be explained with an increased macromolecular mobility to support latex particle interdiffusion and coalescence. It can be concluded that important results towards a distinctly increased knowledge of corrosion processes at polymer/oxide/metal interfaces and promising strategies for interface protection were received.