Cathodic electrophoretic deposition (EPD) of phenylenediamine-modified graphene oxide (GO) for anti-corrosion protection of metal surfaces

Min-Ju Hwang a, Myeong-Gi Kim b, Sanghoon Kim a, Ye Chan Kim b, Hee Won Seo b, Jung Keun Cho b, In-Kyung Park b, Jonghwan Suhr a,b, Hyungil Moon c, Ja Choon Koo c, Hyouk Ryeol Choi c, Kwang Jin Kim d, Yongsug Take e, Jae-Do Nam a,b,*

a Department of Energy Science, Sungkyunkwan University, Suwon, 16410, Republic of Korea
b School of Chemical Engineering, Department of Polymer Science and Engineering, Sungkyunkwan University, Suwon, 16410, Republic of Korea
c School of Mechanical Engineering, Sungkyunkwan University, Suwon, 16410, Republic of Korea
d Active Materials and Processing Laboratory, Department of Mechanical Engineering, University of Nevada, Las Vegas, NV, 89154-4027, USA
e Department of Chemical Engineering, Inha University, Incheon, 22212, Republic of Korea

Article info
Article history:
Received 8 July 2018
Received in revised form 1 October 2018
Accepted 4 October 2018
Available online 5 October 2018

Keywords:
Electrophoretic deposition
Graphene oxide
Anti-corrosion
p-Phenylenediamine
EPD kinetics
GO coating

Abstract
Anti-corrosion metal protection of the 2D structured graphene oxide (GO) was investigated in this study using its intrinsic impermeability against reactive molecules stemming from the small geometric pore size and π-orbital repelling fields of graphene. The electrophoretic deposition (EPD) of GO sheets on metals is an attractive coating method but the negatively-charged GO only allows the anodic EPD process, which makes it difficult to achieve high-quality coating layers due to gas bubbling and electrochemical oxidation of metals. Thus, we imposed the positive charge to GO sheets using p-phenylenediamine (PPD) and successfully carried out the cathodic EPD in the ethanol aqueous solution. The cathodic EPD of GO on copper provided a linear growth rate with both deposition time and voltage providing the thickness over 6.0 μm in a void-free and robust feature, which could hardly be achieved by other coating methods. Converting the GO coating to the reduced GO (RGO) by thermal reduction, we successfully increased the RGO adhesion strength up to the highest adhesion grade of 5B in the cross-cut adhesion test. An excellent anti-corrosion protection capability of the developed RGO coating was demonstrated by a decreased corrosion current density and increased corrosion potential in potentiodynamic polarization analysis.

1. Introduction
Corrosive oxidation of metal surfaces often raises serious reliability issues in long-term utilization in various environments. Particularly, copper is an essential engineering material widely used in hostile environments exposed to corrosion, e.g., electrical power lines, water utility pipelines, heat conductors, heat exchangers, etc [1]. Such reactive metal surfaces as copper have been protected against corrosion/oxidation by modification of their surface properties using passive oxide layers, organic layers and chromium based metal coatings, etc [2]. However, most of these techniques usually raise such inevitable issues as low coating thickness, deterioration of thermal and electrical conductivity, and poor adhesion strength often causing delamination in specific applications. Particularly when copper is exposed to corrosive chemicals of electrolytes under electrical potential in such applications as current collectors of electrochemical devices in batteries, fuel cells, solar cells, and desalinators [3], the protective coating of copper is one of the key issues in extending its applicability and durability.

Recently, protective capability of graphene has attracted a great attention due to its intrinsic impermeability to most molecules [4,5]. Graphene’s π-orbitals form a dense and delocalized electron cloud and subsequently blocks the gap within its aromatic rings,