© 2020 Wiley-VCH GmbH



Supporting Information

for Adv. Sci., DOI: 10.1002/advs.202002658

Growth of NiAl Layered Double Hydroxide on Graphene for Application as an Excellent Anticorrosive Microwave Absorber

Xuefei Xu, Shaohua Shi, Yulin Tang, Guizhen Wang*, Maofan Zhou, Guoqing Zhao, Xuechun Zhou, Shiwei Lin, Fanbin Meng*

Supporting Information

Growth of NiAl Layered Double Hydroxide on Graphene for Application as an Excellent Anticorrosive Microwave Absorber

Xuefei Xu, Shaohua Shi, Yulin Tang, Guizhen Wang*, Maofan Zhou, Guoqing Zhao, Xuechun Zhou, Shiwei Lin, Fanbin Meng*

X Xu, Dr. S Shi, Y Tang, M Zhou, G Zhao, X Zhou, Prof. G Wang, Prof. S Lin. State Key Laboratory of Advanced Materials of Tropical Island Resources (Ministry of Education), Hainan University, Haikou, Hainan, 570228, P.R. China.

Prof. F Meng, State Key Laboratory of Advanced Technologies of Materials (Ministry of Education), School of Materials Science and Engineering, Southwest Jiaotong University, Chengdu, Sichuan, 610031, P.R. China.

E-mail: wangguizhen@hainanu.edu wangguizhen0@hotmail.com (Guizhen Wang), mengfanbin_wing@126.com (Fanbin Meng) The reflection loss values of as-prepared samples were researched according to the transmission line theory:

$$Z_{\rm in} = Z_0 \left(\frac{\mu_{\rm r}}{\varepsilon_{\rm r}}\right) \tanh\left[j\left(\frac{2\pi f d}{c}\right)(\mu_{\rm r}\varepsilon_{\rm r})^{\frac{1}{2}}\right] \#(1)$$
$$RL = 20 \log\left|\frac{Z_{\rm in} - Z_0}{Z_{\rm in} + Z_0}\right| \#(2)$$

where Z_{in} is the input impedance of the absorber, Z_0 the impedance of free space, μ_r the relative complex permeability, ε_r the complex permittivity, *f* the frequency of microwaves, *c* the velocity of light, and *d* the thickness of the absorber.

The attenuation constant (α) was calculated by following equation:

$$\alpha = \frac{\sqrt{2}\pi f}{c} \times \sqrt{(\mu''\varepsilon'' - \mu'\varepsilon') + \sqrt{(\mu''\varepsilon'' - \mu'\varepsilon')^2 + (\mu'\varepsilon'' + \mu''\varepsilon')^2}} \#(3)$$

The impedance matching can be expressed as the equation:

$$Z = \frac{Z_{in}}{Z_0} = \sqrt{\left|\frac{\mu_r}{\varepsilon_r}\right|} \tanh\left[j\left(\frac{2\pi fd}{c}\right)\sqrt{\mu_r\varepsilon_r}\right] \#(4)$$

Where Z_{in} is the impedance of microwave absorber and Z_0 is the impedance of free space. When the *Z* value is close to 1, microwave can enter into the absorber easily and then be converted to thermal or other energy.

The effective permittivity (ε_{eff}^{MG}) of them is highly dependent on the, which can be measured as the following equation:

$$\varepsilon_{eff}^{MG} = \varepsilon_1 \frac{(\varepsilon_2 + 2\varepsilon_1) + 2f_v(\varepsilon_2 - \varepsilon_1)}{(\varepsilon_2 + 2\varepsilon_1) - f_v(\varepsilon_2 - \varepsilon_1)} \#(5)$$

Where ε_1 is permittivity of the solid, ε_2 permittivity of the void and f_v the pore volumes.



Figure S1. SEM images of (a) 50 NiAl-LDH/G, (b) 100 NiAl-LDH/G and (c) 150 NiAl-

LDH/G.



Figure S2. (a) XRD pattern of Al₂O₃/G. (b) XPS spectra of NiAl-LDH/G samples.



Figure S3. The frequency dependence of (a) real permittivity (ε') and imaginary permittivity (ε''), (b) real permeability (μ'') and imaginary permeability (μ''), (c) dielectric loss tangent (*tan* δ_e) and Magnetic loss tangent (*tan* δ_m) of graphene. The frequency dependence of (d) real permeability (μ'), (e) imaginary permeability (μ'') and (f) magnetic loss tangent (*tan* δ_m) of NiAl-LDH/G samples.

Epoxy	$R_{sol} (\Omega cm^2)$	CPE _{pro}		$R_{pro} (\Omega cm^2)$	CPE _{dl}		$R_{ct} (K\Omega cm^2)$	χ^2
		$\frac{Y_0}{(10^{-5} \Omega^{-1} \mathrm{cm}^{-2} \mathrm{s}^{\mathrm{n}})}$	n	-	$\frac{Y_0}{(10^{-5} \Omega^{-1} \mathrm{cm}^{-2} \mathrm{s}^n)}$	n	-	
1 day	49.1±0.4	12.0±1.1	0.6	25.9±3.3	3.7±0.6	0.8	5.3±0.1	1.2×10^{-3}
3 days	47.7±0.4	10.1 ± 0.4	0.7	23.2±2.6	4.7±0.2	0.8	5.2±0.1	1.3×10^{-3}
5 days	48.6±0.4	10.5 ± 1.2	0.7	33.4±4.0	3.8±0.7	0.8	4.5±0.1	9.3×10 ⁻⁴
7 days	50.5±0.4	10.1±0.9	0.6	40.4±6.4	3.9±0.9	0.8	4.6±0.1	7.7×10^{-4}
14 days	48.2±0.2	11.5±0.5	0.7	50.6±7.2	3.5±0.7	0.9	4.7±0.2	4.1×10^{-4}
21 days	50.2±0.3	8.9±0.8	0.7	80.1±10.6	1.6±0.4	0.9	2.9±0.2	4.1×10 ⁻⁴

Table S1. The electrochemical parameters fitted from EIS results of pure epoxy coated steel after immersion of different time.

LDH/G	$R_{sol} (\Omega cm^2)$	CPE _{pro}		$R_{pro} (\Omega cm^2)$	CPE _{LG}		$R_{LG} (\Omega \text{ cm}^2)$	CPE _{dl}		$R_{ct} (K\Omega cm^2)$	χ^2
		$\begin{array}{c} Y_0 \\ (10^{-8} \ \Omega^{-1} \\ cm^{-2} \ s^n) \end{array}$	n	-	$\begin{array}{c} Y_0 \\ (10^{-5} \ \Omega^{-1} \\ cm^{-2} \ s^n) \end{array}$	n	-	$\frac{Yc_0}{(10^{-5} \ \Omega^{-1}} cm^{-2} \ s^n)$	n	-	
1 day	71.0±2.1	13.2±2.3	0.6	26.1±1.7	10.1±1.0	0.6	303.9±26.9	6.9±0.9	0.8	10.7±0.2	1.2×10^{-3}
3 days	60.3±2.2	$7.4{\pm}1.1$	0.6	31.5±1.7	10.7 ± 0.8	0.6	319.9±24.6	6.7±0.6	0.8	12.4±0.2	7.5×10^{-4}
5 days	68.9±2.5	6.0 ± 0.6	0.7	60.0 ± 2.0	$14.0{\pm}1.0$	0.6	352.3±34.6	6.5±0.9	0.8	10.4±0.2	8.7×10^{-4}
7 days	54.7±1.5	11.2±1.3	0.7	28.3±1.2	14.9 ± 1.0	0.6	296.6±33.9	6.2±0.9	0.9	10.3±0.2	7.7×10^{-4}
14 days	37.2±0.8	16.2±1.7	0.8	18.1±0.7	16.3±0.7	0.7	425.3±48.1	4.2±0.6	0.9	8.7±0.1	5.4×10^{-4}
21 days	39.5±0.9	11.3±0.9	0.6	25.2±0.8	16.3±0.7	0.7	546.7±72.0	3.2±0.6	0.9	7.9±0.1	3.6×10 ⁻⁴

Table S2. The electrochemical parameters fitted from EIS results of 7 wt.% NiAl-LDH/G coated steel after immersion of different time.

WILEY-VCH



Figure S4. (a) Evolution of OCP values for bare steel, pure epoxy coated steel and 7 wt.% NiAl-ccepoxy coated steel and 7 wt.% NiAl-LDH/G coated steel with continuous immersion of 21 days.

Table S3. The parameters fitted from tafel plots results in Figure 8h.

samples	E_{corr} (mV)	i_{corr} (mA/cm ²)	b _a (mV/dec)	b _c (mV/dec)
Steel	- 926	1.2×10^{-2}	4225	- 1988
Epoxy	- 667	1.1×10^{-2}	4766	- 4852
LDH/G	- 381	1.0×10^{-2}	5854	- 5554

The corrosion process on the surface of bare carbon steel:

Fe → Fe²⁺ + 2e⁻#(6)
Fe²⁺ → Fe³⁺ + e⁻#(7)
H₂O +
$$\frac{1}{2}$$
O₂ + 2e⁻ → 2OH⁻#(8)

$$2Fe^{2+} + O_2 + 2H_2O \rightarrow 2FeOOH + 2H^+ \#(9)$$

WILEY-VCH

 $Fe^{2+} + 2Fe^{3+} + 80H^- \rightarrow Fe_3O_4 + 4H_2O\#(10)$