

## SUPPORTING INFORMATION

### **Controllable synthesis of layered double hydroxide nanosheets to build organic inhibitor-loaded nanocontainers for enhanced corrosion protection of carbon steel**

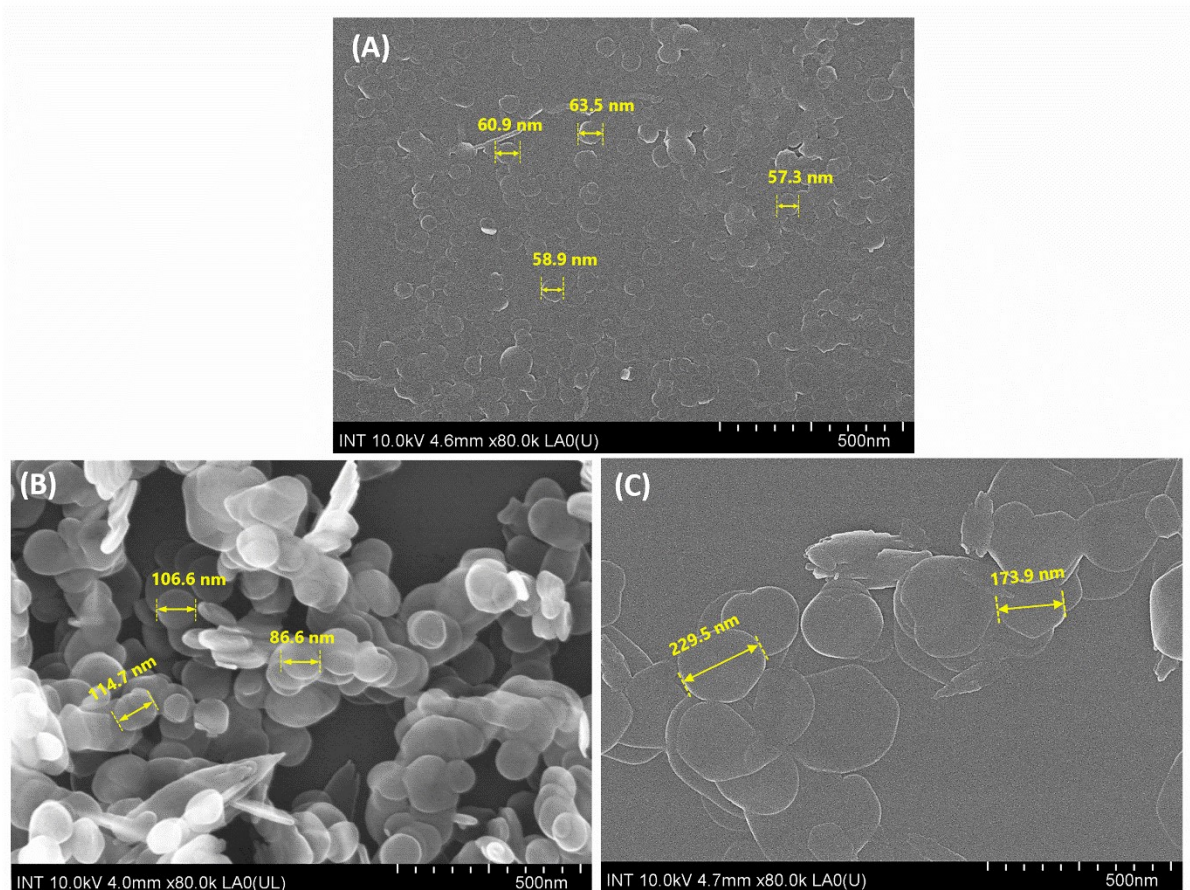
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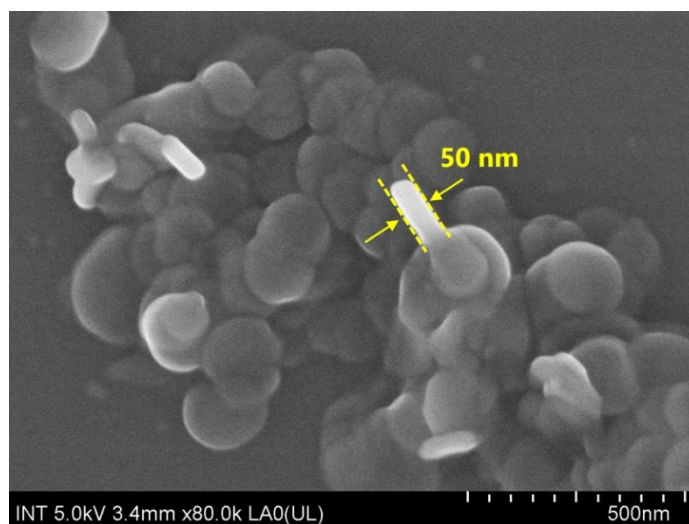
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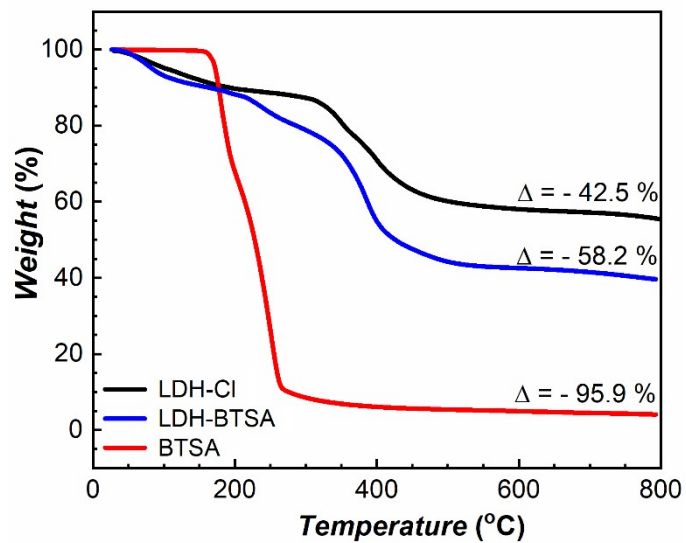
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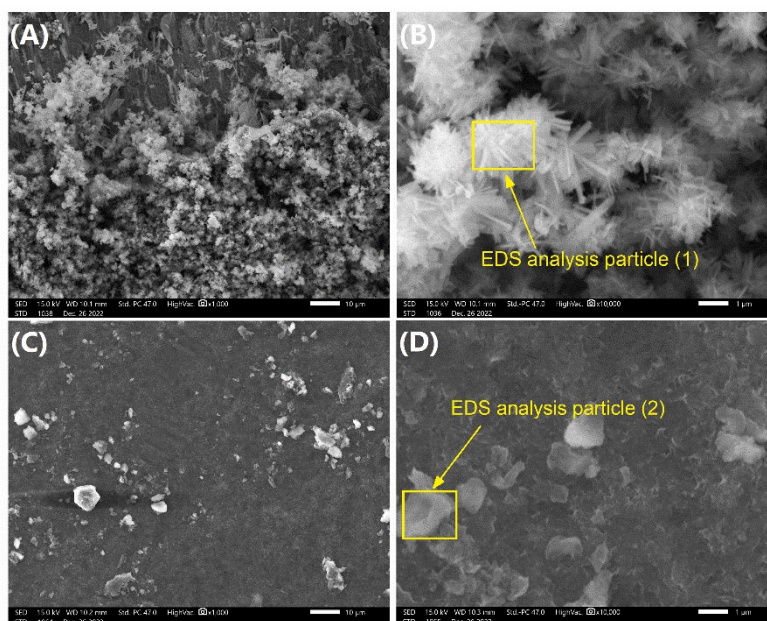
**Figure S1.** FE-SEM image of LDH-Cl samples hydrothermally treated at 80 °C (A), 100 °C (B) and 150 °C (C)



**Figure S2.** FE-SEM image of prepared LDH-Cl-F hydrothermal treatment at 125 °C



**Figure S3.** TGA variation curves of BTSA, LDH-Cl and LDH-BTSA



**Figure S4.** SEM image of steel electrode surface after 24 hours immersion in 0.1 M NaCl solution without inhibitor (A, B) and with 3 g/L of LDH-BTSA (C, D).

**Table S1.** The zeta potential and hydrodynamic size of the prepared LDH-Cl under various hydrothermal treatment temperatures.

Samples	Hydrothermal temperature (°C)	Zeta potential (mV)	Z-average (nm)	PI value
LDH-Cl-80	80	+61.7	77.4	0.171
LDH-Cl-100	100	+72.1	115.4	0.195
LDH-Cl-125	125	+73.9	168.4	0.180
LDH-Cl-150	150	+75.6	212.5	0.131

**Table S2.** Basal spacing and lattice parameters of LDH crystallites.

Samples	LDH-CI					LDH-BTSA				
	2 $\theta$ (°)	$\theta$ (radians)	d (nm)	Lattice parameters (nm)		2 $\theta$ (°)	$\theta$ (radians)	d (nm)	Lattice parameters (nm)	
				a	c				a	c
(003)	11.279	0.098	0.7839			5.372	0.047	1.6438		
(006)	22.643	0.198	0.3924	0.3058	2.3531	11.238	0.098	0.7867	0.3038	4.8258
(110)	60.509	0.528	0.1529			60.951	0.532	0.1519		

The  $2\theta$  positions of the peak in the XRD pattern are used to calculate the basal spacing ( $d_{003}$ ) and lattice parameters of LDH. The interlayer spacing was determined using Bragg's law:  $d = \lambda / 2\sin\theta$ . Here,  $\lambda$  is the X-ray wavelength ( $\lambda = 1.5418$  nm), and  $\theta$  is the (003) diffraction angle. Lattice parameters  $a$  and  $c$  represent the metal-metal cation distances in the lamellar layer and unit cell thickness of LDH, respectively, were calculated using the  $2\theta$  angle position of (003), (006) and (110) diffraction peaks:  $a = 2d_{(110)}$  and  $c = 3/2[d_{(003)} + 2d_{(006)}]$  [1,2].

[1] Vieira, D.; Sokol, D.; Smalenskaite, A.; Kareiva, A.; Ferreira, M.; Vieira, J.; Salak, A. Cast iron corrosion protection with chemically modified MgAl layered double hydroxides synthesized using a novel approach. *Surf. Coat. Technol.* **2019**, *375*, 158-163.

[2] Zhao, Y.; Li, F.; Zhang, R.; Evans, D. G.; Duan, X. Preparation of layered double-hydroxide nanomaterials with a uniform crystallite size using a new method involving separate nucleation and aging steps. *Chem. Mater.* **2002**, *14* (10), 4286-4291.

**Table S3.** The Tafel parameters from the polarization curves of steel electrodes in 0.1 M NaCl solution with and without inhibitors after 2 h of immersion.

Samples	$E_{\text{corr}}$ (mV)	$i_{\text{corr}}$ ( $\mu\text{A}\cdot\text{cm}^{-2}$ )	$\beta_a$ (mVdec $^{-1}$ )	$-\beta_c$ (mVdec $^{-1}$ )	$\eta_i$ (%)
Blank	-613.589	19.202	102.4	885.4	-
BTSA	-335.079	0.598	178.4	112.0	96.9
LDH-Cl	-555.169	12.987	114.4	820.3	32.4
LDH-BTSA	-439.462	1.866	202.2	143.1	90.3

Where  $E_{\text{corr}}$ ,  $i_{\text{corr}}$ ,  $\beta_a$ ,  $\beta_c$ ,  $\eta_i$  are corrosion potential, corrosion current density, Tafel slope of anodic, cathodic, and inhibition efficiency, respectively. The inhibitor efficiency was determined by the equation:  $\eta_i = 100\% \times (i^{\circ} - i^{\text{n}}) / i^{\circ}$ ; where  $i^{\text{n}}$  and  $i^{\circ}$  is the corrosion density of the electrode exposed in NaCl with and without the inhibitor.

**Table S4.** Fitting parameters from the EIS diagram of mild steel electrode immersed in NaCl 0.1 M solution with and without inhibitors at different immersion time

Samples	Time (h)	$R_s$ ( $\Omega.cm^2$ )	$R_p$ ( $\Omega.cm^2$ )	$Q_{dl}$ ( $mFs^{(n-1)}. cm^{-2}$ )	$n_{dl}$	$R_f$ ( $\Omega.cm^2$ )	$Q_f$ ( $mFs^{(n-1)}. cm^{-2}$ )	$n_f$
Blank	2	27.3	1300	1.287	0.68	-	-	-
	4	33.3	1224	2.066	0.65	-	-	-
	8	33.8	1078	2.119	0.65	-	-	-
	24	34.5	1059	2.506	0.62	-	-	-
LDH-Cl	2	26.8	1571	0.559	0.73	-	-	-
	4	32.6	1776	1.125	0.77	-	-	-
	8	30.4	1501	1.382	0.70	-	-	-
	24	30.7	1440	2.042	0.75	-	-	-
BTSA	2	29.2	14190	0.132	0.42	3457	0.184	0.77
	4	29.1	9979	0.281	0.54	2660	0.286	0.72
	8	28.9	4935	0.521	0.58	266.2	0.836	0.62
	24	28.2	2409	1.216	0.66	94.3	1.195	0.59
LDH-BTSA	2	29.1	6905	0.136	0.78	2746	2.778	0.88
	4	29.4	9703	0.111	0.79	1406	1.601	0.73
	8	29.8	11271	0.093	0.80	1261	0.776	0.81
	24	30.6	3270	0.221	0.86	135.8	0.331	0.73

Where  $R_s$ ,  $R_p$ , and  $R_f$  represented solution resistance, polarization resistance, and film resistance, respectively.  $CPE_{dl}$  and  $CPE_f$  are constant phase elements of the double layer and film,  $Q$ , and  $n$  values are parameters of CPE.