## **One-Pot** Synthesis of NiAl-CO<sub>3</sub> LDH Anti-Corrosion Coatings from CO<sub>2</sub>-Saturated Precursors

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## **Supporting Information**

## SI-1 XRD pattern of NiAl-CO<sub>3</sub> LDH powders



**Fig. S1** XRD pattern of NiAl-CO<sub>3</sub> LDH powders. The sample was synthesized according to the published recipe (M. Wei, X. Y. Xu, X. R. Wang, F. Li, H. Zhang, Y. L. Lu, M. Pu, D. G. Evans and X. Duan, *Eur. J. Inorg. Chem.*, 2006, 2831).

SI-2 Influence of concentration of CO<sub>2</sub> dissolved in the precursor solution on nucleation and growth of NiAl-CO<sub>3</sub> LDH crystallites



**Fig. S2** Magnified cross-sectional SEM images of NiAl-CO<sub>3</sub> LDH films prepared from (a) DI aged and (b) CO<sub>2</sub>-saturated precursor solutions, respectively.

Magnified cross-sectional SEM images of both the NiAl-CO<sub>3</sub> LDH (DI) and NiAl-CO<sub>3</sub> LDH (CO<sub>2</sub>-satur) films are shown in Fig. S2. It is clear that the cross-sectional morphology of two NiAl-CO<sub>3</sub> LDH films is distinct. The average size for LDH crystallites in Fig. S2a is ~1  $\mu$ m. In contrast, the crystal size of LDHs in Fig. S2b is much smaller (~0.1  $\mu$ m). Obviously, the number density of LDH crystallites per unit area in Fig. S2b is much higher than that in Fig. S2a, which strongly supports the fact that increasing the concentration of CO<sub>2</sub> in the precursor solution gives rise to drastic increase of the nucleation density of NiAl-CO<sub>3</sub> LDH crystallites on the substrate.

In addition, compared with the NiAl-CO<sub>3</sub> LDH (DI aged) film, the excellent anticorrosion capacity of the NiAl-CO<sub>3</sub> LDH (CO<sub>2</sub>-satur) film indicates that LDHs prepared from CO<sub>2</sub>-saturated precursor solution are well-intergrown and few intercrystalline defects are present in the film. These results convincingly demonstrate that the growth rate of NiAl-CO<sub>3</sub> LDH crystallites in the CO<sub>2</sub>-saturated precursor solution is considerably higher. SI-3 Illustration of the long-term durability of NiAl-CO<sub>3</sub> LDH (CO<sub>2</sub>-satur)-coated Al substrate



**Fig. S3** SEM images of the (a) NiAl-CO<sub>3</sub> LDH (CO<sub>2</sub>-satur) film and (b) bare Al substrate after immersion in 3.5 wt.% NaCl solution for 15 days.