The Microstructure and Protective Properties of Electroplating Zinc Films on NdFeB Magnet from a Chloride-free Nonaqueous Solution

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Figure S1 SEM surface morphology of Zn film at different distances from the proximal cathode electroplating from the 0.1M-EG bath: (a) 7-8 cm, (b) 6-7 cm, (c) 5-6 cm and (c) 4-5 cm.



Figure S2 SEM surface morphology of Zn film at different distances from the proximal cathode electroplating from the 0.3M-EG bath: (a) 7-8 cm, (b) 6-7 cm, (c) 5-6 cm and (c) 4-5 cm.



Figure S3 SEM surface morphology of Zn film at different distances from the proximal cathode electroplating from the 0.5M-EG bath: (a) 7-8 cm, (b) 6-7 cm, (c) 5-6 cm and (c) 4-5 cm.



Figure S4 SEM surface morphology of Zn film at different distances from the proximal cathode electroplating from the 0.7M-EG bath: (a) 7-8 cm, (b) 6-7 cm, (c) 5-6 cm and (c) 4-5 cm.



Figure S5 The (002) reflection of $Zn_{0.5M-3}$ and $Zn_{0.7M-7}$ film on brass with corresponding FWHM.



Figure S6 The full-scale XPS spectra of $Zn_{0.5M-3}$ and $Zn_{0.7M-6}$ film.



Figure S6

Figure S7 The variation of current density with deposition time at different deposition potentials (a) in the 0.1M-EG bath and (b) in the 0.3M-EG bath (60 °C, vs Ag/AgCl). The fitting results of $(I/I_{max})^2$ vs. t/t_{max} (c) the 0.1M-EG bath and (d) the 0.3M-EG bath.



Figure S8 The cross-section morphology of the deposited Zn film on NdFeB from an aqueous alkaline zinc plating bath



Figure S9 XRD of the deposited $Zn_{0.7M-6}$ film after immersing in 3.5 wt.% NaCl at different times.

From Figure S9, it can be observed that there are some $Zn(OH)_2 \cdot 0.5H_2O$ and $Zn(OH)_2$ in the initial stage of corrosion, and most of the Zn coating still exists

without losing its crystal structure and characteristics. Over time, a large amount of $Zn_5(OH)_8Cl_2\cdot H_2O$ and $Zn(OH)_2$, as well as some $Zn(OH)_2\cdot 0.5H_2O$, began to appear in the film. In the end, only $Zn_5(OH)_8Cl_2\cdot H_2O$ remained the corrosion product. It is speculated that the following reaction occurred:

$$Zn \rightarrow Zn^{2+} + 2e^{-} \tag{1}$$

$$Zn^{2+}+2H_2O \rightarrow Zn(OH)_2+2H^+$$
⁽²⁾

$$O_2 + 4e^- + 2H_2O \rightarrow 4OH^-$$

As the immersing time prolongs, the concentration of OH^- in the solution increases, resulting in the continuous generation of $Zn (OH)_2$ and $Zn(OH) \cdot 0.5H_2O$. Due to the continuous consumption of OH^- by Zn^{2+} , the reaction (2) continues to proceed. This is also why $Zn_5(OH)_8Cl_2 \cdot H_2O$ constantly increases.

(3)