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Supplementary Materials

Phase Chemistry and Spatial Organization Govern Chloride Corrosion Behavior of CrFeNi-Based Multi-Principal Element Alloys

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Complex nonlinear least-squares fitting was applied simultaneously to real and imaginary impedance components using inverse-impedance-modulus weighting (1/|Z| for each component). The one-time-constant resistor–constant-phase-element (1RQ) circuit R_s-(R_{ct} || CPE_{dl}). The alternative two-time-constant resistor–constant-phase-element (2RQ) circuit was R_s-(R_1 || CPE_1)-(R_2 || CPE_2), with $R_p = R_1 + R_2$. Multistart fits and changes in the fitting frequency window were examined. The weighted sum of squared errors (SSE) and corrected Akaike information criterion (AICc) quantify numerical fit quality for this specific objective.

Table S1. Full measured-frequency-range circuit fitting and model-sensitivity comparison.

Parameter	1RQ, Measured Frequency Range	2RQ, Measured Frequency Range	2RQ, Constrained (Full Measured Range)
Rs (Ω cm^2)	1.195	≈ 0 (bound)	0.744
R1 (Ω cm^2)	—	1.503	0.579
R2 / Rct (kΩ cm^2)	75.414	73.310	73.926
Rp (kΩ cm^2)	75.414	73.312	73.927
Additional-branch n1	—	0.269	0.600 (bound)
Dominant-branch n	0.902	0.908	0.906
Additional-branch fchar	—	2.43 MHz	123 kHz
Weighted SSE	0.25164	0.02510	0.03570
AICc	−1099.4	−1484.8	−1424.9

The constrained 2RQ fit limits Rs to 0.70–2.0 Ω cm^2 and CPE exponents to 0.60–1.00. In the unconstrained fit, the added branch has R1 ≈ 1.50 Ω cm^2 and fchar ≈ 2.43 MHz, beyond the 99,950 Hz upper measurement limit; Rs approaches the lower numerical bound. In the constrained fit, the additional exponent n1 reaches the imposed 0.60 bound.

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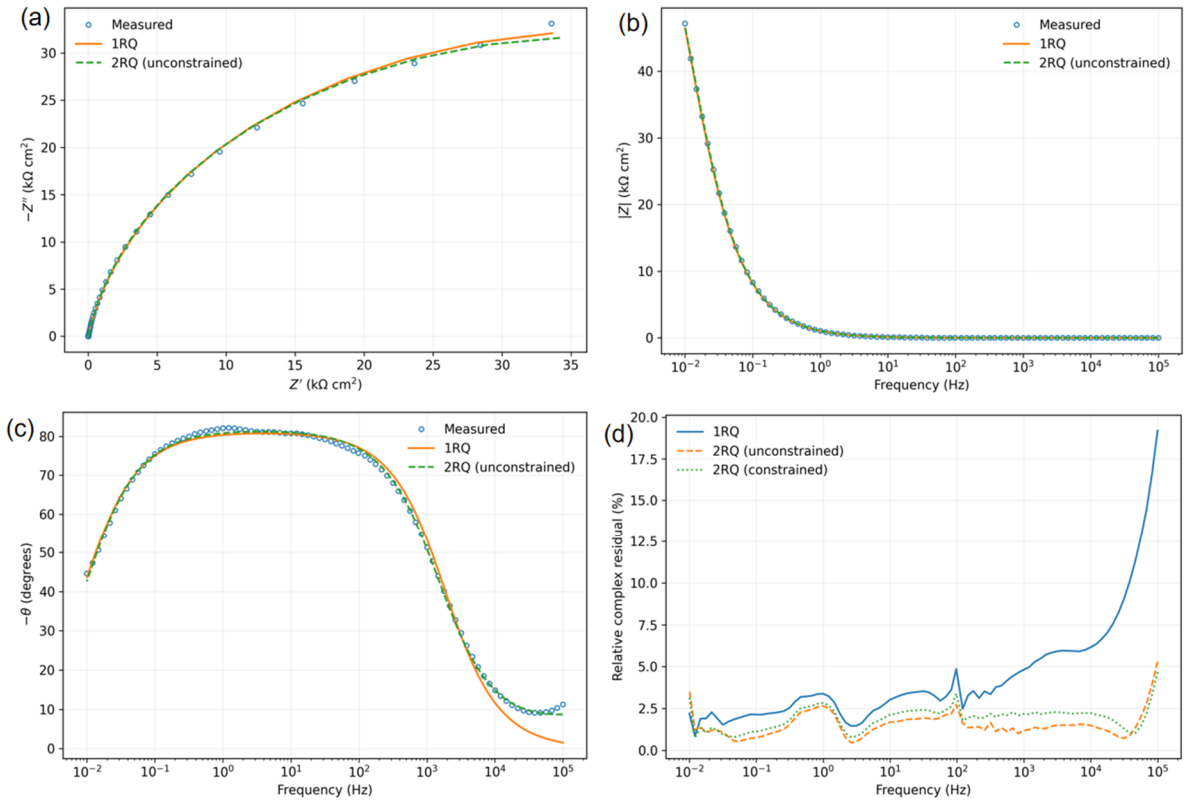


Figure S1. Experimental and fitted HR-CrFeNiAlTi impedance spectra: (**a**) Nyquist spectra: raw measurements compared with the 1RQ and unconstrained 2RQ fits; (**b**) Bode impedance magnitude; (**c**) Bode phase angle; (**d**) Relative complex residuals, including constrained 2RQ sensitivity test.