Supplementary Materials: Ultra-Long-Term Reliable Encapsulation Using an Atomic Layer Deposited $HfO_2/Al_2O_3/HfO_2$ Triple-Interlayer for Biomedical Implants

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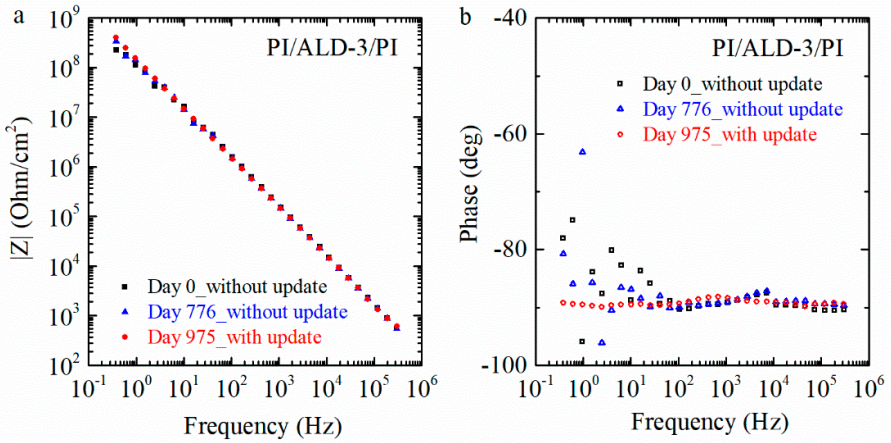

Figure S1. EIS measurements comparison for PI/ALD-3/PI coating samples when the potentiostat updated with a low current module: (**a**) bode plot of impedance spectra against frequency; (**b**) bode plot of phase against frequency. The current resolution extends from 760 pA down to 76 fA.

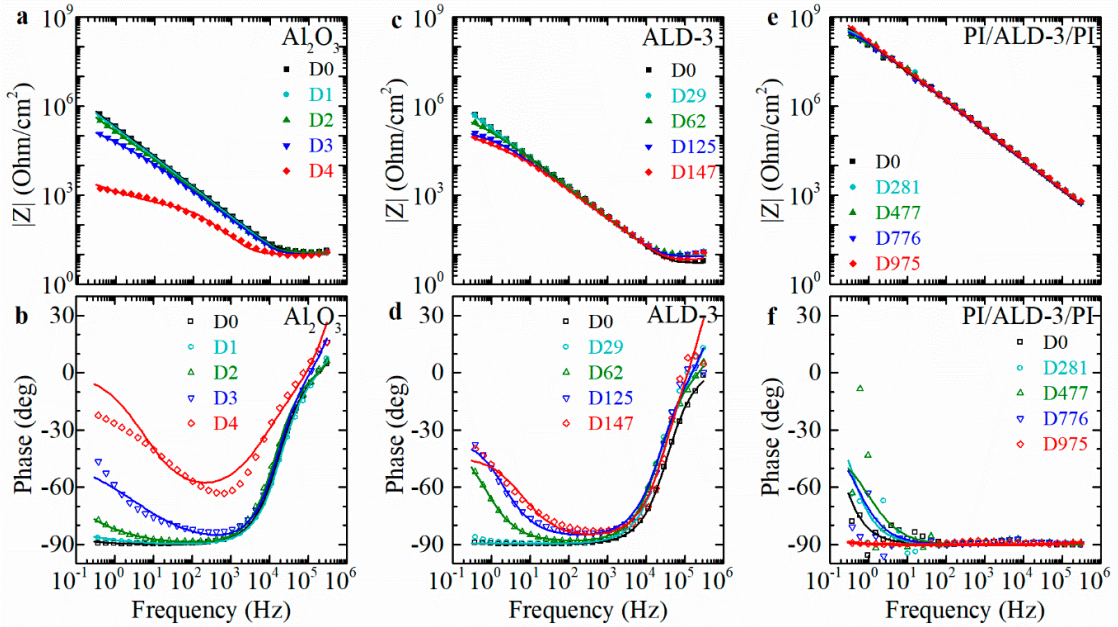

Figure S2. Fitting results in bode plot for the barriers based on equivalent circuit model in Figure 6a: (**a**,**b**) Al_2O_3; (**c**,**d**) ALD-3; (**e**,**f**) PI/ALD-3/PI.

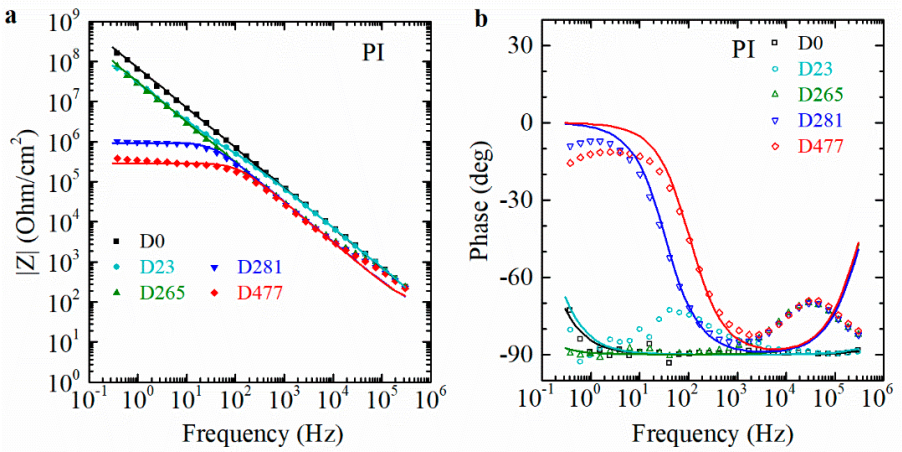

Figure S3. Fitting results for PI barrier based on equivalent circuit model in Figure 6a: (**a**) bode plot of impedance spectra against frequency; (**b**) bode plot of phase against frequency.

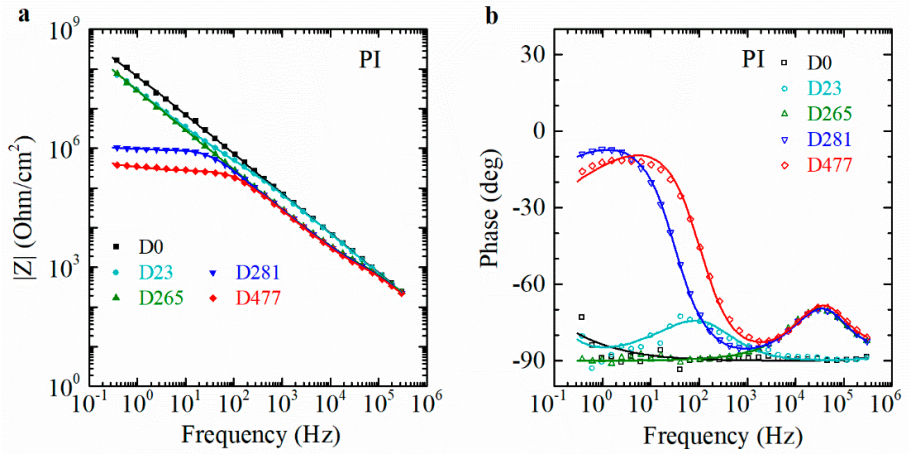

Figure S4. Fitting results for PI barrier based on equivalent circuit model in Figure 7: (**a**) bode plot of impedance spectra against frequency; (**b**) bode plot of phase against frequency.

Table S1. Fitted equivalent circuit model parameters of 20 nm ALD Al_2O_3 coating layer on copper.

Soaking Time (days)	R_{PBS} (Ohm/cm²)	Q_b (F s^{n-1}/cm²)	n	R_{pore} (Ohm/cm²)	Error
0	12	2.3×10^{-7}	1.00	7.7×10^{7}	2%
1	10	2.6×10^{-7}	0.99	1.2×10^{7}	2%
2	12	3.3×10^{-7}	0.98	1.8×10^{6}	4%
3	9	6.5×10^{-7}	0.91	1.8×10^{5}	9%
4	8	1.4×10^{-5}	0.72	1.8×10^{3}	15%

Table S2. Fitted equivalent circuit model parameters of ALD ALD-3 coating layer on copper

Soaking Time (days)	R_{PBS} (Ohm/cm²)	Q_b (F s^{n-1}/cm²)	n	R_{pore} (Ohm/cm²)	Error
0	5	2.1×10^{-7}	1.00	2.9×10^{7}	2%
10	12	2.2×10^{-7}	1.00	1.7×10^{7}	3%
20	8	2.2×10^{-7}	1.00	4.2×10^{6}	4%
29	9	2.1×10^{-7}	1.00	5.8×10^{6}	4%
40	11	2.4×10^{-7}	0.98	5.5×10^{5}	4%
62	9	2.6×10^{-7}	0.97	3.6×10^{5}	5%
95	7	3.3×10^{-7}	0.95	3.5×10^{5}	5%
125	8	4.0×10^{-7}	0.93	1.2×10^{5}	12%
147	6	4.9×10^{-7}	0.92	7.9×10^{4}	18%

Table S3. Fitted equivalent circuit model parameters of PI coating layer on copper

Soaking Time (days)	R_{PBS} (Ohm/cm²)	C_b (F/cm²)	R_{pore} (Ohm/cm²)	C_{dl} (F/cm²)	R_{ct} (Ohm/cm²)	W (Ohm $s^{-1/2}$/cm²)	Error
0	5	5.7×10^{-10}	7.3×10^{8}	–	–	–	9%
23	5	5.8×10^{-10}	1.2×10^{6}	7.0×10^{-10}	2.5×10^{9}	–	8%
62	19	5.9×10^{-10}	1.1×10^{4}	7.0×10^{-10}	3.3×10^{9}	–	6%
115	11	5.7×10^{-10}	4.7×10^{4}	7.2×10^{-10}	2.8×10^{9}	–	5%
168	22	5.9×10^{-10}	5.2×10^{3}	7.2×10^{-10}	4.3×10^{9}	–	8%
219	21	6.0×10^{-10}	5.4×10^{3}	7.1×10^{-10}	2.4×10^{9}	–	6%
265	21	6.1×10^{-10}	3.1×10^{3}	7.6×10^{-10}	2.5×10^{9}	–	4%
281	17	6.2×10^{-10}	2.7×10^{3}	7.5×10^{-10}	8.7×10^{5}	2.3×10^{5}	3%
321	24	6.3×10^{-10}	2.4×10^{3}	7.7×10^{-10}	2.4×10^{5}	2.3×10^{5}	6%
392	27	6.5×10^{-10}	2.2×10^{3}	7.7×10^{-10}	2.0×10^{5}	1.7×10^{5}	5%
477	20	6.4×10^{-10}	2.4×10^{3}	8.0×10^{-10}	2.5×10^{5}	1.9×10^{5}	4%

Table S4. Fitted equivalent circuit model parameters of PI/ALD-3/PI coating layer on copper[a].

Soaking Time (days)	R_{PBS} (Ohm/cm²)	C_b (F/cm²)	R_{pore} (Ohm/cm²)	Error
0	6	2.8×10^{-10}	9.3×10^{8}	30%
56	8	2.7×10^{-10}	4.3×10^{8}	28%
115	5	2.7×10^{-10}	9.2×10^{8}	22%
155	7	2.7×10^{-10}	4.5×10^{8}	28%
192	6	2.6×10^{-10}	5.5×10^{8}	25%
281	5	2.6×10^{-10}	5.2×10^{8}	20%
366	7	2.8×10^{-10}	4.0×10^{8}	32%
477	14	2.6×10^{-10}	2.9×10^{8}	21%
681	13	2.7×10^{-10}	3.3×10^{8}	23%
776	10	2.7×10^{-10}	4.5×10^{8}	21%
868	15	3.0×10^{-10}	4.0×10^{8}	31%
973	12	2.6×10^{-10}	2.4×10^{10}	4%
1028	11	2.6×10^{-10}	7.5×10^{9}	5%

[a] The fitting error for the data fits of PI/ALD-3/PI is relative high during the first 868 test days because the impedance value is out of detection limits of the potentiostat. The error drops down to 5% at the last two measurements (the last two shaded rows) since the "low current module" installed to increase detection limits of the potentiostat.