

Electronic Supplementary Information

Tuning atomic-scale mixing of Pd-Hf nanoparticles by atmospheric-pressure spark ablation

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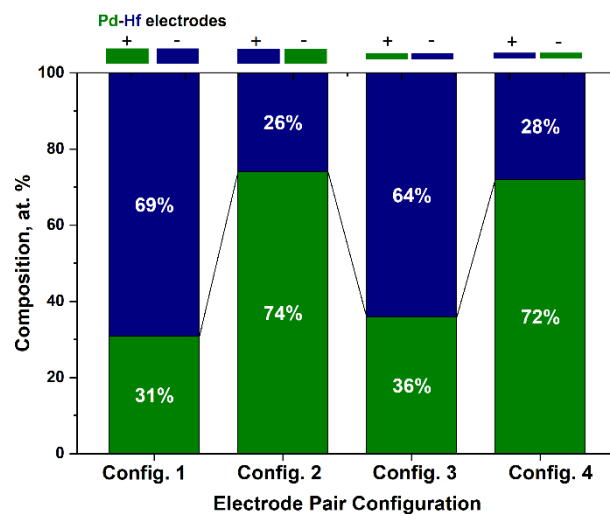


Figure S1. Fraction of Pd (green) and Hf (blue), estimated by the corresponding at.%, determined by gravimetric analysis for different electrode configurations, different polarity but of the same thickness. In all cases, the thick and thin electrodes had diameters of 2 and 0.25 mm, respectively.

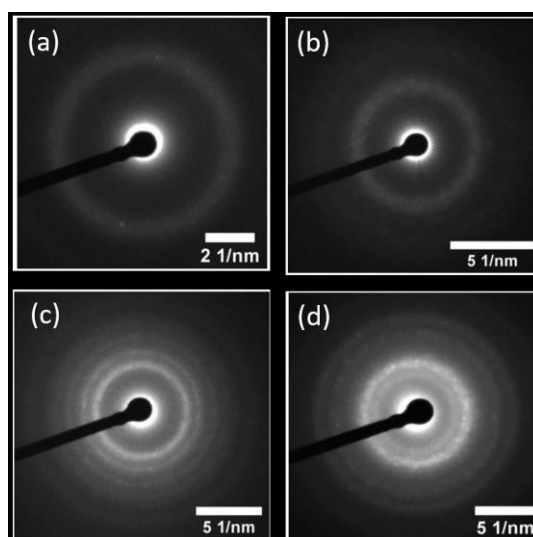


Figure S2. SAED patterns for different electrode configurations (a) Configuration 1: Pd (2 mm, positive) and Hf (0.25 mm, negative); (b) Configuration 2: Pd (2 mm, negative) and Hf (0.25 mm, positive); (c) Configuration 3: Pd (0.25 mm, positive) and Hf (2 mm, negative); (d) Configuration 4: Pd (0.25 mm, positive) and Hf (2mm, negative).

Table S1. Pd:Hf composition (at. %) estimated from TEM-EDS for different electrode configurations, different polarity and thickness. **Configuration 1:** Pd (2 mm, positive) and Hf (0.25 mm, negative); **Configuration 2:** Pd (2 mm, negative) and Hf (0.25 mm, positive); **Configuration 3:** Pd (0.25 mm, positive) and Hf (2 mm, negative); **Configuration 4:** Pd (0.25 mm, positive) and Hf (2mm, negative).

Spectrum	Configuration 1		Configuration 2		Configuration 3		Configuration 4	
	Pd (at.%)	Hf (at.%)	Pd (at.%)	Hf (at.%)	Pd (at.%)	Hf (at.%)	Pd (at.%)	Hf (at.%)
1	46.32	53.68	44.72	55.28	57.92	42.08	75.77	24.23
2	44.94	55.06	37.53	62.47	58.94	41.06	78.46	21.54
3	46.08	53.92	48.61	51.39	57.61	42.39	73.35	26.65
4	47.38	52.62	48.40	51.60	57.00	43.00	73.67	26.33
5	61.57	38.43	35.73	64.27	61.00	39.00	72.67	27.33
6	34.00	66.00	33.03	66.97	57.20	42.80	76.92	23.08
7	41.42	58.58	36.15	63.85	55.08	44.92	73.31	26.69
8	45.71	54.29	44.96	55.04	58.25	41.75	71.40	28.60
9	41.50	58.50	40.17	59.83	63.34	36.66	74.38	25.62
10	33.18	66.82	36.36	63.64	62.28	37.72	71.91	28.09
11	34.01	65.99	41.22	58.78	57.87	42.13	77.94	22.06
12	29.61	70.39	44.67	55.33	54.01	45.99	76.43	23.57
13	33.55	66.45	38.73	61.27	59.11	40.89	73.37	26.63
14	38.47	61.53	46.47	53.53	53.65	46.35	68.97	31.03
15	29.73	70.27	42.91	57.09	60.14	39.86	72.12	27.88
16	31.15	68.85	36.14	63.86	60.86	39.14	76.50	23.50
17	26.40	73.60	33.81	66.19	59.22	40.78	71.09	28.91
18	31.86	68.14	33.78	66.22	54.82	45.18	74.08	25.92
19	35.03	64.97	34.07	65.93	61.86	38.14	72.12	27.88
20	37.80	62.20	40.87	59.13	56.12	43.88	73.16	26.84

Table S2. Pd:Hf composition (at. %) estimated from STEM-EDS for different electrode configurations, different polarity and thickness. **Configuration 1:** Pd (2 mm, positive) and Hf (0.25 mm, negative);

Spectrum	Pd (at.%)	Hf (at.%)
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1	40.03	59.97
2	41.56	58.44
3	55.74	44.26
4	57.10	42.90
5	71.62	28.38
6	48.09	51.91
7	55.88	44.12
8	49.20	50.80
9	33.49	66.51
10	26.12	73.88
11	45.41	54.59
12	56.29	43.71
13	36.43	63.57
14	50.22	49.78
15	55.18	44.82
16	49.58	50.42
17	50.45	49.55
18	47.00	53.00
19	41.88	58.12
20	51.87	48.13
21	52.55	47.45
22	50.11	49.89
23	67.61	32.39
24	67.00	33.00
25	44.34	55.66
26	29.06	70.94
27	59.36	40.64

28	27.69	72.31
29	28.38	71.62
30	41.53	58.47
31	67.55	32.45
32	47.60	52.40
33	35.24	64.76
34	23.59	76.41
35	43.72	56.28

Table S3. Pd:Hf composition (at. %) estimated from STEM-EDS for different electrode configurations, different polarity and thickness. **Configuration 2:** Pd (2 mm, negative) and Hf (0.25 mm, positive)

Spectrum	Pd (at.%)	Hf (at.%)
1	40.43	59.57
2	41.17	58.83
3	46.56	53.44
4	53.03	46.97
5	66.79	33.21
6	25.86	74.14
7	43.23	56.77
8	37.45	62.55
9	35.51	64.49
10	48.48	51.52
11	54.65	45.35
12	40.93	59.07
13	52.49	47.51
14	51.87	48.13
15	41.50	58.50
16	58.50	41.50

17	52.01	47.99
18	48.85	51.15
19	34.35	65.65
20	56.31	43.69
21	61.12	38.88
22	39.91	60.09
23	54.78	45.22
24	49.26	50.74
25	45.43	54.57
26	41.32	58.68
27	38.63	61.37
28	64.23	35.77
29	48.32	51.68
30	39.10	60.90
31	41.23	58.77
32	49.88	50.12
33	73.48	26.52
34	53.58	46.42
35	50.39	49.61
36	48.40	51.60
37	50.59	49.41
38	55.36	44.64
39	43.58	56.42
40	51.87	48.13
41	61.58	38.42
42	52.74	47.26
43	42.71	57.29

44	52.14	47.86
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Table S4. Pd:Hf composition (at. %) estimated from STEM-EDS for different electrode configurations, different polarity and thickness. **Configuration 3:** Pd (0.25 mm, positive) and Hf (2 mm, negative)

Spectrum	Pd (at.%)	Hf (at.%)
1	64.35	35.65
2	71.14	28.86
3	68.77	31.23
4	58.22	41.78
5	78.22	21.78
6	57.00	43.00
7	79.63	20.37
8	58.68	41.32
9	64.87	35.13
10	66.42	33.58
11	81.68	18.32
12	76.16	23.84
13	81.34	18.66
14	59.37	40.63
15	70.57	29.43
16	65.34	34.66
17	68.57	31.43
18	65.51	34.49
19	82.51	17.49
20	64.63	35.37
21	67.03	32.97
22	62.67	37.33
23	68.36	31.64

24	75.33	24.67
25	62.62	37.38
26	79.29	20.71
27	80.02	19.98
28	56.07	43.93
29	32.67	67.33
30	81.74	18.26
31	64.55	35.45

Table S5. Pd:Hf composition (at. %) estimated from STEM-EDS for different electrode configurations, different polarity and thickness. **Configuration 4:** Pd (0.25 mm, positive) and Hf (2mm, negative).

Spectrum	Pd (at.%)	Hf (at.%)
1	84.51	15.49
2	74.14	25.86
3	75.29	24.71
4	61.47	38.53
5	79.47	20.53
6	85.24	14.76
7	79.11	20.89
8	84.27	15.73
9	87.52	12.48
10	75.14	24.86
11	76.20	23.80
12	60.84	39.16
13	78.81	21.19
14	77.76	22.24
15	73.03	26.97
16	83.12	16.88

17	63.24	36.76
18	78.26	21.74
19	80.79	19.21
20	87.33	12.67