## Flexible MoS<sub>2</sub>-Polyimide Electrode for Electrochemical Biosensors and their Applications for the highly sensitive Quantification of Endocrine Hormones: PTH, T3, and T4

*Hyeong-U Kim*,<sup>†, ‡, #</sup> *Hye Youn Kim*,<sup>§,#</sup> *Hyunho Seok*,<sup>†</sup> *Vinit Kanade*,<sup>†</sup> *Hocheon Yoo*,  $\parallel$  *Kyu-Young Park*,<sup>‡</sup> *Jae-Hyun Lee*, <sup> $\perp$ </sup> *Min-Ho Lee*, <sup>\*,§</sup> *and Taesung Kim*, <sup>\*,†, τ</sup>

† SKKU Advanced Institute of Nanotechnology (SAINT), Sungkyunkwan University, Suwon
 16419, Republic of Korea

‡ Department of Materials Science and Engineering, Northwestern University, Evanston60208, United States

§ School of Integrative Engineering, Chung-Ang University, Seoul 06973, Republic of Korea
<sup>II</sup>Department of Electronic Engineering, Gachon University, Seongnam 13120, Republic of Korea

⊥Department of Energy Systems Research and Department of Materials Science and

Engineering, Ajou University, Suwon 16499, Republic of Korea <sup>\*</sup>School of Mechanical Engineering, Sungkyunkwan University, Suwon 16419, Republic of Korea \*Corresponding authors: tkim@skku.edu, mhlee7@cau.ac.kr

**Preparation of Au-PI electrode for MoS<sub>2</sub>.** Polyimide (PI) can be used as a substrate for MoS<sub>2</sub> since its melting point is around 200 °C. The thickness of polyimide film is 175 μm and tensile strength is larger than 130 MPa. (Suzhou Kying Industrial Materials Co., Ltd, China) A4 size of PI was patterned using a shadow mask with an e-beam evaporator. The shadow mask shows a working electrode (circle) and contact electrode (Rectangular) shape as shown in **Figure S2**a. A double line of the electrode was designed, and 12 electrodes can deposit at the same time in a 4-inch wafer as shown in **Figure S2**b. It is loaded in an e-beam evaporator for deposition of the 1nm Mo metal layer. It is called the Au-PI electrode for synthesizing of MoS<sub>2</sub>.

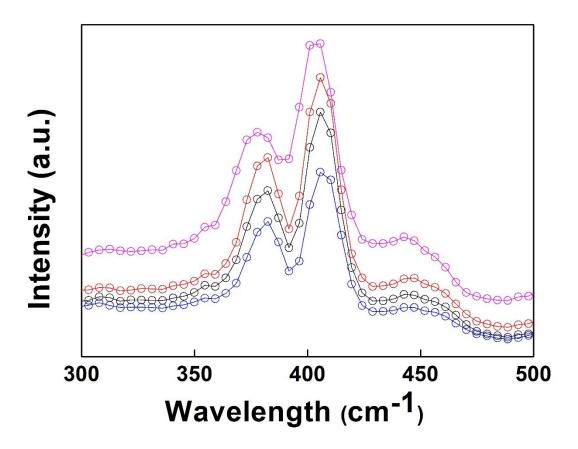
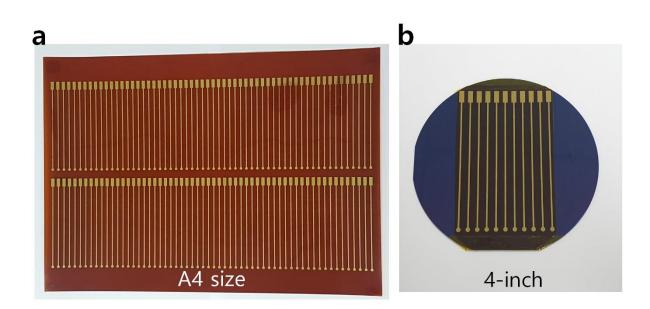


Figure S1. Uniformity test for Raman spectrum with four different MoS<sub>2</sub>-Au-PI electrodes



**Figure S2.** Photograph of Au patterned PI as a substrate for the  $MoS_2$ -Au-PI electrode. a) Au patterned on the PI substrate. b) 12 of Au-PI electrode before Mo deposition using e-beam evaporator.

**Correlation Analysis.** The ALP-(PTH, T3, T4)-MoS<sub>2</sub>-Au-PI flexible was analyzed in terms of its linear range, reproducibility, and repeatability. The concentrations of endocrine hormones (PTH, T3, T4) from 30 subjects' serum were evaluated using E 170 as reference standards. The results measured by the standard equipment for each clinical sample were determined by standard Pearson correlation coefficients using electrochemical with ALP-(PTH, T3, T4)-MoS<sub>2</sub>-Au-PI flexible complex. As a measurement of the average correlation between samples, the number of average correlations for all standard results were calculated. The slope of the fitted line, Roche E 170, used to compare expression levels of Real Samples to the rest of the data set, was calculated using linear regression minimizing the sum of the squared errors.

**Table S1.** Analytical evaluation of clinical samples using electrochemical with ALP-(PTH, T3, T4)-MoS<sub>2</sub>-Au-PI flexible complex and their accuracy of correlation with the standard results using E 170 (n = 30). a) PTH; b) T3; c) T4. The Pearson correlation factor between the measured and standard results for PTH, T3, and T4 were 0.98, 0.80, and 0.82, respectively.

a

No.	Mean of PTH concentration of clinical samples	Mean of PTH concentration of E 170	Accuracy (%)
1	1.11	1.2	92.5
2	4.61	7.7	59.87
3	16.14	8.96	19.87
4	8.28	9.16	90.39
5	9.94	10.73	92.64
6	25.3	15.95	41.38
7	25.78	20.2	72.38
8	5.5	20.83	26.40
9	21.45	21.7	98.85
10	26.63	22.92	83.81
11	10	24.79	40.34
12	20.36	30.6	66.54
13	31.93	32.07	99.56
14	33.01	32.92	99.73
15	46.14	35.08	68.47
16	28.19	38.83	72.6
17	34.34	42.01	81.74
18	48	42.41	86.82
19	40.72	50.55	80.55
20	62.5	62.42	99.87
21	30.84	78.38	39.35
22	85.63	95.42	89.74
23	83.13	110.9	74.96
24	160	144	88.88

b			
30	390	399.7	97.57
29	353.33	384.9	91.8
28	333.33	351.9	94.72
27	236.67	280.8	84.28
26	183.33	189.4	96.8
25	213.33	172.5	76.33

No.	Mean of T3 concentration of clinical samples	Mean of T3 concentration of E 170	Accuracy (%)
1	0.86	0.71	78.87
2	0.87	0.72	79.17
3	0.67	0.79	84.81
4	1.2	0.8	50
5	0.5	0.9	55.56
6	1.17	0.93	74.19
7	1.65	0.95	26.32
8	1.5	1	50
9	0.69	1.02	67.65
10	1.07	1.02	95.1
11	1.62	1.04	44.23
12	0.8	1.05	76.19
13	1.32	1.09	78.9
14	1.15	1.09	94.5
15	1.68	1.1	47.27
16	1.15	1.1	95.45
17	1.56	1.18	67.8
18	1.83	1.21	48.76
19	0.48	1.32	36.36
20	1.25	1.52	82.24
21	1.48	1.61	91.93
22	1.76	1.78	98.88
23	1.46	1.84	79.35
24	2.41	2.11	85.78

c			
30	3.82	4.64	82.33
29	4.36	3.91	88.49
28	3.65	3.83	95.30
27	1.73	2.54	68.11
26	2.76	2.49	89.16
25	1.58	2.44	64.75

No.	Mean of T4 concentration of clinical samples	Mean of T4 concentration of E 170	Accuracy (%)
1	4.53	3.1	53.87
2	3.15	4.6	68.48
3	4.62	5.1	90.59
4	3.92	5.3	73.96
5	5.1	5.3	96.23
6	7.3	6	78.33
7	7.13	6.4	88.59
8	6.44	6.7	96.12
9	6.17	6.8	90.74
10	4.76	6.8	70
11	7.46	7	93.43
12	6.79	7.1	95.63
13	7.03	7.2	97.64
14	4.96	7.3	67.95
15	6.75	7.5	90
16	8.85	7.8	86.54
17	9.55	8	80.63
18	7.87	8.8	89.43
19	6.32	8.8	71.82
20	8.09	8.9	90.9
21	9.02	9.4	95.96
22	8.97	10	89.7
23	9.59	10.2	94.02
24	10.47	10.4	99.33

25	8.09	10.5	77.05
26	9.86	11.4	86.49
27	13.31	11.5	84.26
28	14.5	12.3	82.11
29	13.6	12.5	91.2
30	14.3	14.4	99.31