Supporting Information

In-situ plasma-assisted synthesis of polydopamine-functionalized gold nanoparticles for biomedical applications

Linh Nhat Nguyen,^{a,b} Neha Kaushik,^c Pradeep Lamichhane,^a Sohail Mumtaz,^a Ramhari Paneru,^a Pradeep Bhartiya,^a Jae Sung Kwon,^d Yogendra Kumar Mishra,^e Liem Quang Nguyen,^b Nagendra Kumar Kaushik,^{a,*} Eun Ha Choi^{a,*}

^aDepartment of Electrical and Biological Physics, Plasma Bioscience Research Centre, and Applied Medicine Research Center, Kwangwoon University, Seoul 01987, Korea

^bLaboratory of Plasma Technology, Institute of Materials Science, Vietnam Academy of Science and Technology, 18 Hoang Quoc Viet, Cau Giay, Hanoi 100000, Vietnam

^cDepartment of Biotechnology, University of Suwon, Hwaseong, 18223, Korea

^dDepartment and Research Institute of Dental Biomaterials and Bioengineering Yonsei University, College of Dentistry, Seoul 03722, Korea

^eMads Clausen Institute, NanoSYD, University of Southern Denmark, Alsion 2, Denmark



Fig. S1. OES lines of O+ at 441 nm and Atomic O at 777 nm.



Fig. S2. SEM analysis and photograph (inset) of Au@PDA NPs (0.2 mM HAuCl¬4 and 0.05 mM DA) synthesized with different plasma power. Scale bar: 200 nm.



Fig. S3: Hydrodynamic diameter and PDI analysis of of plasma synthesized Au@PDA NPs



Mobility Distribution

No File Name	Repet. No	MeasTime	pН	Zeta Potential (mV)
1 #2_20200609_154116	1	15:41:16	NA	-24.70
2 #2_20200609_154116	2	15:41:16	NA	-21.23
3 #2_20200609_154116	3	15:41:16	NA	-19.09
Average				-21.67

Fig. S4: Zeta potential analysis of plasma synthesized Au@PDA NPs



Fig. S 5: XPS spectra of pristine DA sample for C 1s, O 1s and N 1s core-levels

Table S1: C 1s, O 1s, and N 1s energy biding peak analysis for pristine DA and Au@PDA NPs

Sample	C 1s (eV)		O 1s (eV)		N 1s (eV)				
	C-C	C-0/C-N	C=0	он	C=0	-NH ₃ +	-NH ₂	R-NH-R	-NO ₃ -
DA	284.1 eV	285.6 eV	287.3	532.2	530.4	401.1	399.4		
Au@PDA NPs	284.7 eV	286.4 eV	288.4	532.6	530.8			399.9	407.2



Fig. S6: Absorption spectra of DA solution before (black line) and after (red line) the plasma treatment. The peak at 280 nm corresponds to the absorption of DA molecule. Under the plasma treatment, DA undergoes oxidation into PDA.



Fig. S7: UV-vis absorption of Au@PDA NPs when stored in refrigerator for 2 months



Fig. S8: a) Photograph and b) SEM image of gold thin film formed at plasma-water interface



Fig. S9: SEM image of Au@PDA NPs after 10 minutes plasma treatment



Fig. S10: Size distribution and average diameter size of Au@PDA NPs synthesized with different DA concentration and 0.2 mM HAuCl4. The samples were measure without centrifugation



Fig. S11: SEM analysis of Au@PDA NPs synthesized with different DA concentration and 0.2 mM HAuCl4. The samples were measure without centrifugation. (Scale bar: 200 nm)



Fig. S12: UV-vis absorption spectra of Au@PDA NPs synthesized with different DA concentrations and 0.2 mM HAuCl4.



Fig. S13: Temporal evolution UV-vis absorption spectra of HAuCl4-DA solution with different DA concentration under 5 minutes

of plasma treatment.



Fig. S14: Photographs of HAuCl₄-DA 0.1 mM synthesized with 4 minutes of plasma treatment. The solution gradually changed 5 min, 30 min, and 2h after the plasma treatment finished.



、

Fig. S15: SEM analysis of Au@PDA NPs synthesized with 0.2 mM HAuCl4 and 0.05 mM DA under different plasma reaction time. The samples were measure without centrifugation. Scale bar: 200 nm.



Fig. S16: Size distribution and average diameter size of Au@PDA NPs synthesized with 0.2 mM HAuCl4 and 0.05 mM DA under different plasma treatment time. The samples were measure without centrifugation.



Fig. S17: UV-Vis spectra and photographs of Au@PDA NPs synthesized with different HAuCl4 concentration (0.1 -1mM) and 0.05 mM DA



Fig. S 18: SEM analysis of Au@PDA NPs synthesized with different HAuCl4 concentration (0.1 -1mM) and 0.05 mM DA (Scale bar:

200 nm)



Fig. S19 SEM of AuNPs provided by CytoDiasnostics, Inc

Table S2: Current advances in the fabrication of Au@PDA nanoparticles.

Year	Synthesis methods		Core Size Shell Thickness		PDA	Ref
	Au Core	PDA Shell			coating time	
2013	Chemical	Self-	50.6 ± 5.9 [nm]	19.0 [nm] - 36.4 [nm]	1 [h]	[1]
	synthesis	polymerization				
2014	Chemical	Self-	80 [nm]	5 [nm]	4 [h]	[2]
	synthesis	polymerization				
2015	Chemical	Self-	42.3 ± 2.7 [nm]	4.6 ± 0.3 [nm]	1 [h]	[3]
	synthesis	polymerization				
2016	UV radiation	Self-	110 [nm]	31 [nm]	24 [h]	[4]
		polymerization				
2017	Chemical	Self-	45 [nm]	1.3 [nm]	1 [h]	[5]
	synthesis	polymerization				
2018	Chemical	Sonicate-	30 [nm]	5 – 6 [nm]	96 [h]	[6]
	synthesis	assisted				
2018	Chemical	Self-	19 8 + 2 0 [nm]	10 [am]	20 [min]	[7]
2010	synthesis	polymerization	15.0 ± 2.0 [mm]	10 [nm]	20 [min]	[/]
2019	Chemical	Solf	20 + 3 nm [nm]		4 [1-]	[0]
2015	synthesis	polymerization	20 ± 5 min [min]		4 [n]	[8]
2010	O		20,400 []			
2019	One step Cher	nical synthesis	20-400 [nm]	-	90 [min]*	[9]
2019	One step Chemical synthesis		8 [nm]	-	24 [h]	[10]
2020	One step Plasma synthesis		44.4 + 4.8 [nm]	4 []	۲ (maim)*	Thissul
2020	one-step i las	ind synthesis		4 [nm]	5 [min]*	This work

References

- X. Liu, L. Angeles, Q. Jin, K. Ren, J. Xu, J. Cao, H. Li, J. Li, Q. Jin, K. Ren, J. Ji, ACS Nano 2013, 7, 9384.
- [2] A. Kumar, S. Kumar, W. K. Rhim, G. H. Kim, J. M. Nam, J. Am. Chem. Soc. 2014, 136, 16317.
- [3] C. H. J. K. K. Choi, J. J. Li, Y. J. Xu, L. W. C. Ho, K. Kin, W. To, C. H. J. K. K. Choi, L.
 Bian, C. H. J. K. K. Choi, J. J. Li, Y. J. Xu, L. W. C. Ho, M. Zhu, K. K. W. To, *J. Am*.

Chem. Soc. 2015, 137, 7337.

- [4] C. Li, Z. Liu, P. Yao, *RSC Adv.* **2016**, *6*, 33083.
- [5] W. Ye, H. Huang, W. Yang, X. Wang, C. Ren, Q. Hu, Y. Li, B. Ren, *Analyst* 2017, 142, 3459.
- [6] K. H. S. Sy, L. W. C. Ho, W. C. Y. Lau, H. Ko, C. H. J. Choi, *Langmuir* **2018**, *34*, 14033.
- [7] Y. Wu, H. Wang, F. Gao, Z. Xu, F. Dai, W. Liu, Adv. Funct. Mater. 2018, 28, 1.
- [8] X. Liu, G. Liao, L. Zou, Y. Zheng, X. Yang, Q. Wang, X. Geng, S. Li, Y. Liu, K. Wang, ACS Appl. Mater. Interfaces 2019, 11, 40817.
- [9] J. Du, C. Jing, Anal. Chim. Acta 2019, 1062, 131.
- [10] X. Luo, X. Xie, Y. Meng, T. Sun, J. Ding, W. Zhou, Anal. Chim. Acta 2019, 1087, 76.