Supporting Information

Uncovering Gold Nanoparticle Synthesis Using Microchip Laser System through Pulsed Laser Ablation in Aqueous Solution

Barana Sandakelum Hettiarachchi,[a] Yusuke Takaoka,[b] Yuta Uetake,[ac] Yumi Yakiyama,*[ac] Hwan Hong Lim,[d] Takunori Taira,[de] Mihoko Maruyama,[f] Yusuke Mori,[f] Hiroshi Y. Yoshikawa[b] and Hidehiro Sakurai*[ac]

[a]Division of Applied Chemistry, Graduate School of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan. E-mail: yakiyama@chem.eng.osaka-u.ac.jp, hsakurai@chem.eng.osaka-u.ac.jp

[b]Division of Applied Physics, Graduate School of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan.
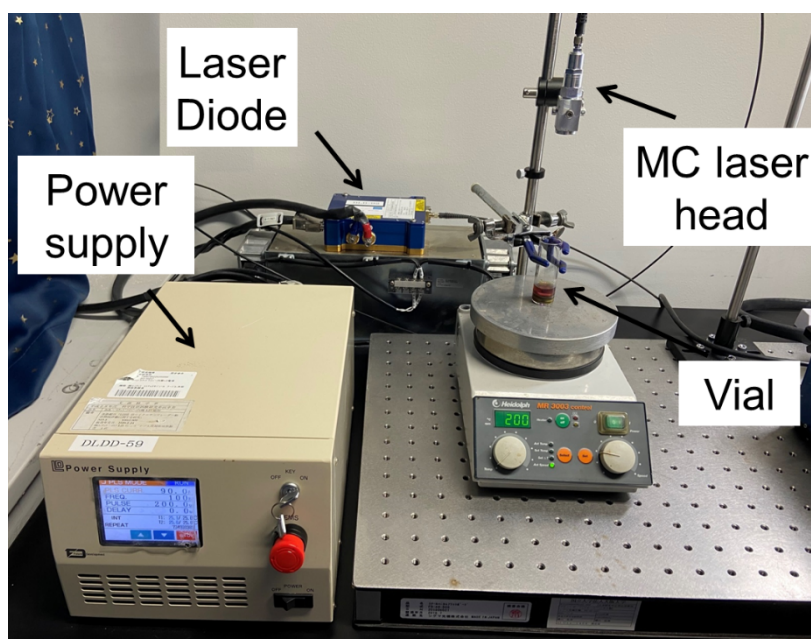
[c]Innovative Catalysis Science Division, Institute for Open and Transdisciplinary Research Initiatives (ICS-OTRI), Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan.

[d]Division of Research Innovation and Collaboration, Institute for Molecular Science, 38 Nishigonaka, Myodaiji, Okazaki 444-8585, Japan.
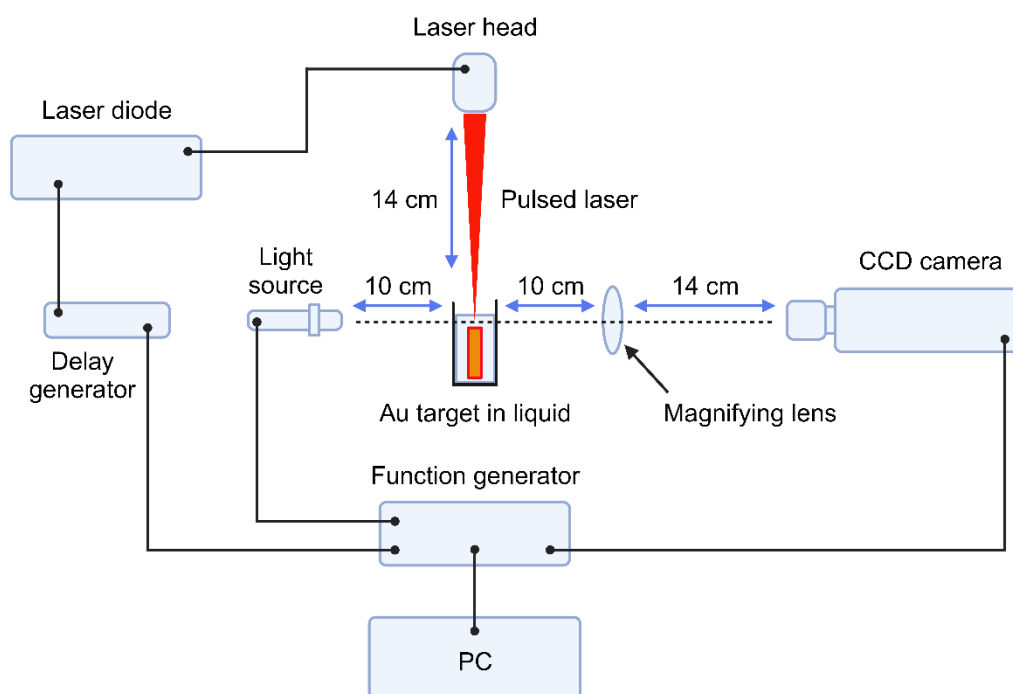
[e]Laser-Driven Electron-Acceleration Technology Group, RIKEN SPring-8 Center, 1-1-1 Kouto, Sayo-cho, Sayo-gun, Hyogo 679-5148, Japan.

[f]Division of Electrical, Electronic and Infocommunications Engineering, Graduate School of Engineering, Osaka University, 2-1 Yamada-oka, Suita, Osaka 565-0871, Japan.
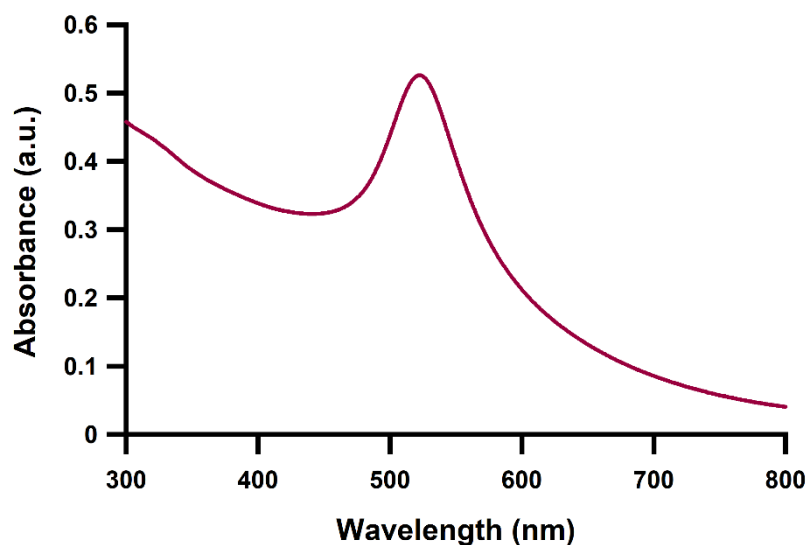
Figure S1. Setup of PLAL system.

Figure S2. Experimental setup for monitoring the bubble formation with the aid of videography.
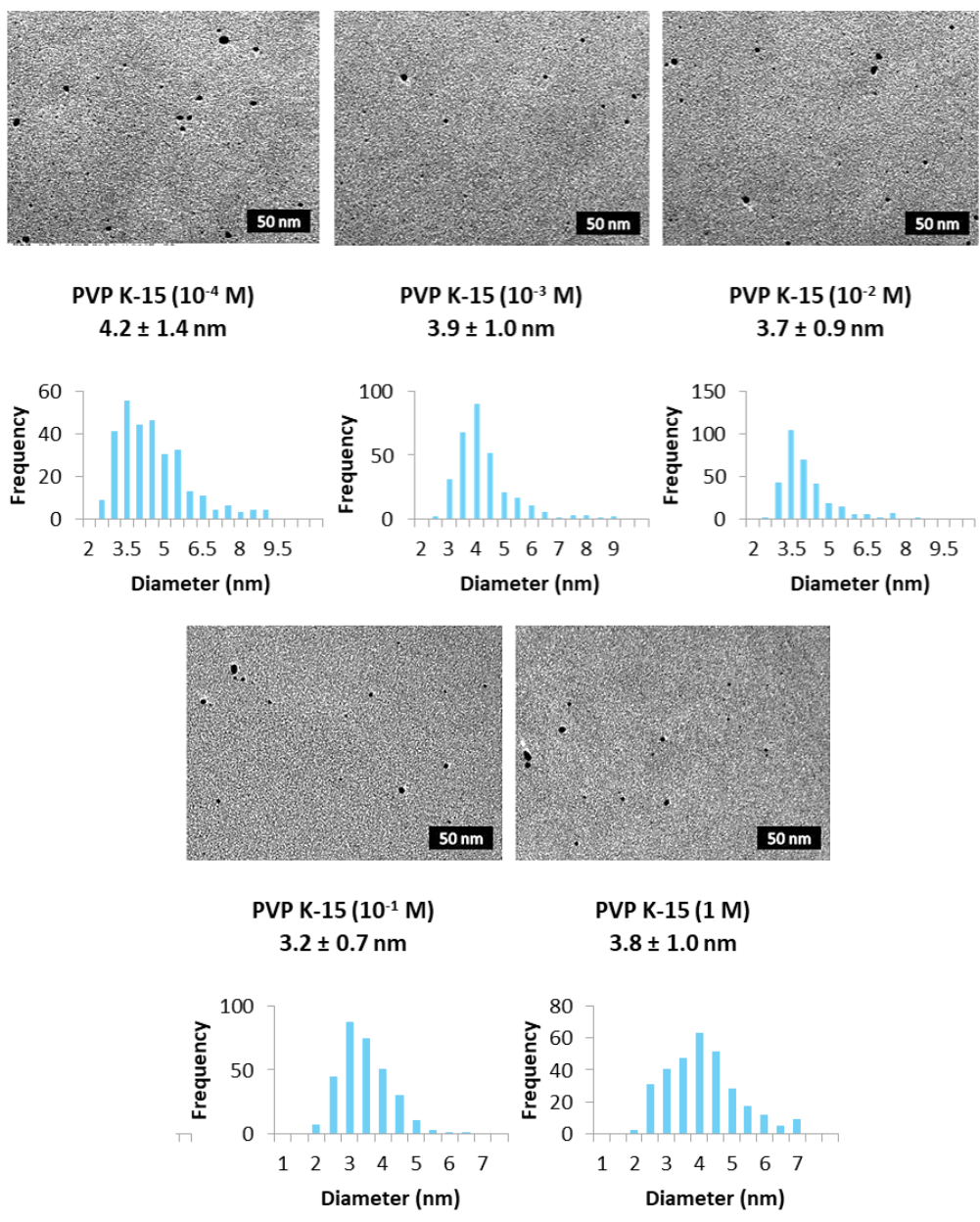
Figure S3. UV/Vis absorption spectrum of the Au NPs produced by MCL ablation in water.
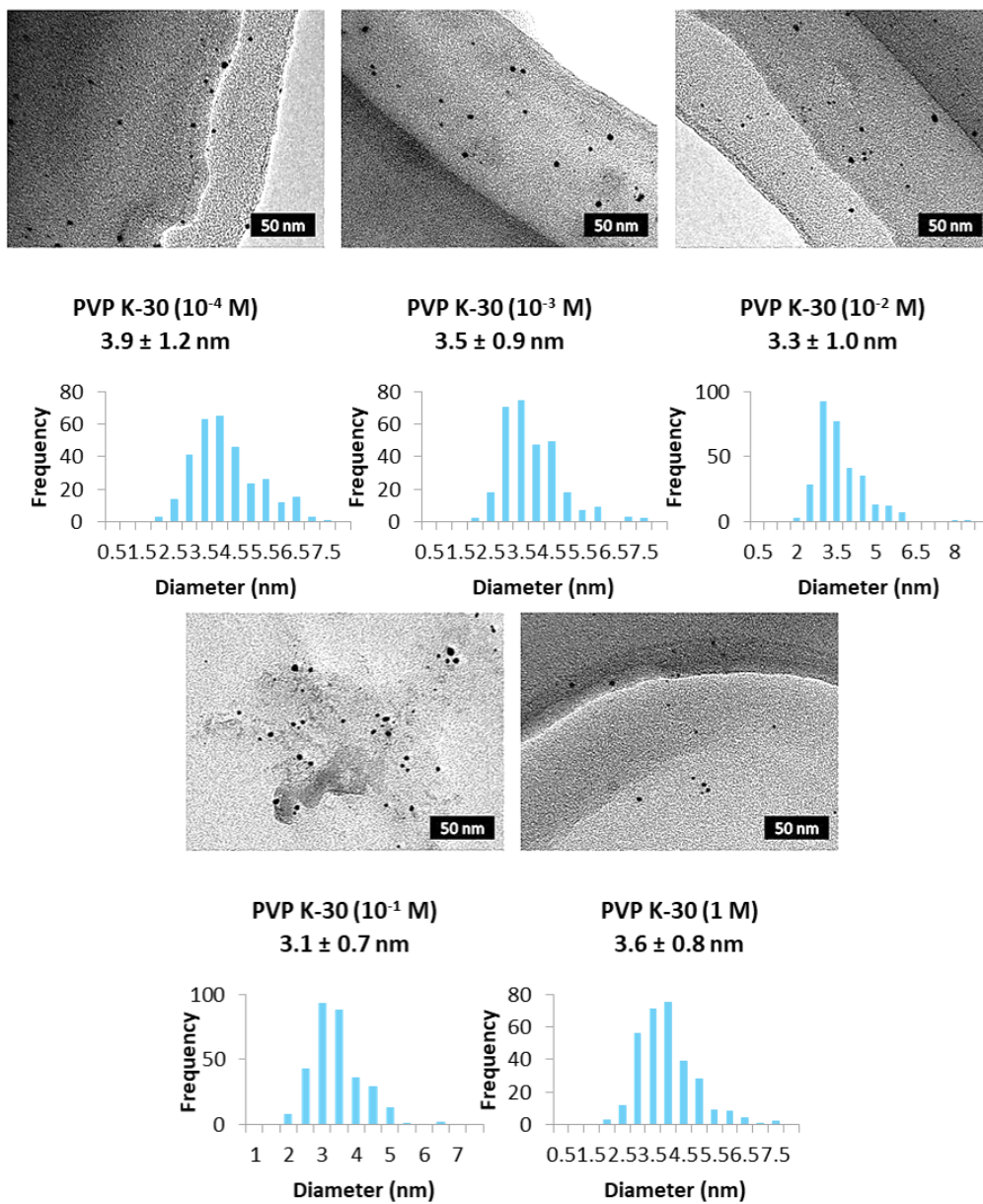
Table S1. Present and reported results of PLAL generated particle sizes.

| Metal	Matrix	Matrix conc. (mM)	Laser condition	Particle size (nm)[a]	Ref.
Au	PVP	0 0.1 1 10 100 1000	900 ps, 1064 nm, 100 Hz, 27 J/pulse	4.9 ± 1.3 3-4 ± 0.7-1.4	This work
Au[b]	PVP	0.0001[c] 0.001[c] 0.01[c]	100 fs, 780 nm, 10 Hz, 6 mJ/pulse	3-5	1
Au[d]	PVP	0.01[c]	100 fs, 800 nm, 1 kHz, 1 mJ/pulse	7-27	2
Au	PVP	0.5	7 ns, 1064 nm, 10 Hz, 11 mJ/pulse	20-50[e]	3
Au	PVP	0.4	8 ns, 335 nm, 10 Hz, 17 mJ/pulse[f]	720[g]	4
Au	SDS[h]	10 0.1	12 ns, 1064 nm, 10 Hz, 80 mJ/pulse	4.6 14.4	5
Au	β-CD[i]	0 0.1 1 10	110 fs, 800 nm, 1 kHz, 1 mJ/pulse	40-60 10 5.3 2.3	6
Ag	PVP	0 2 6 12 18	8 ns, 1064 nm, 10 Hz, 12 mJ/pulse	18 13 10 11 11	7

Figure S4. TEM images of Au:PVP (K-15) prepared in water.

**Figure S5.** TEM images of Au:PVP (K-30) prepared in water.
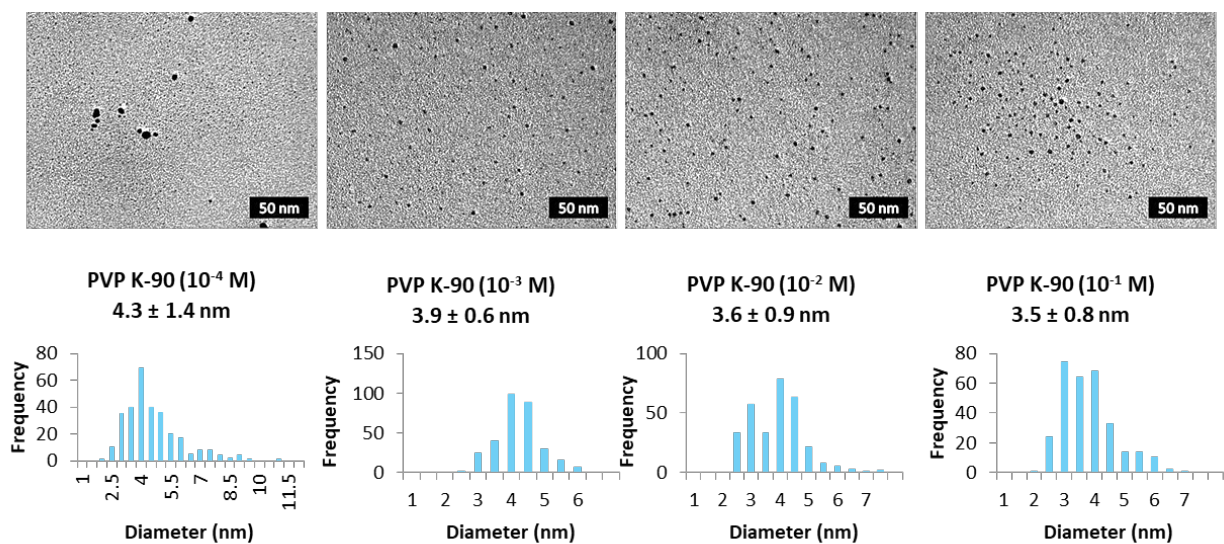
Figure S6. TEM images of Au:PVP (K-90) prepared in water.
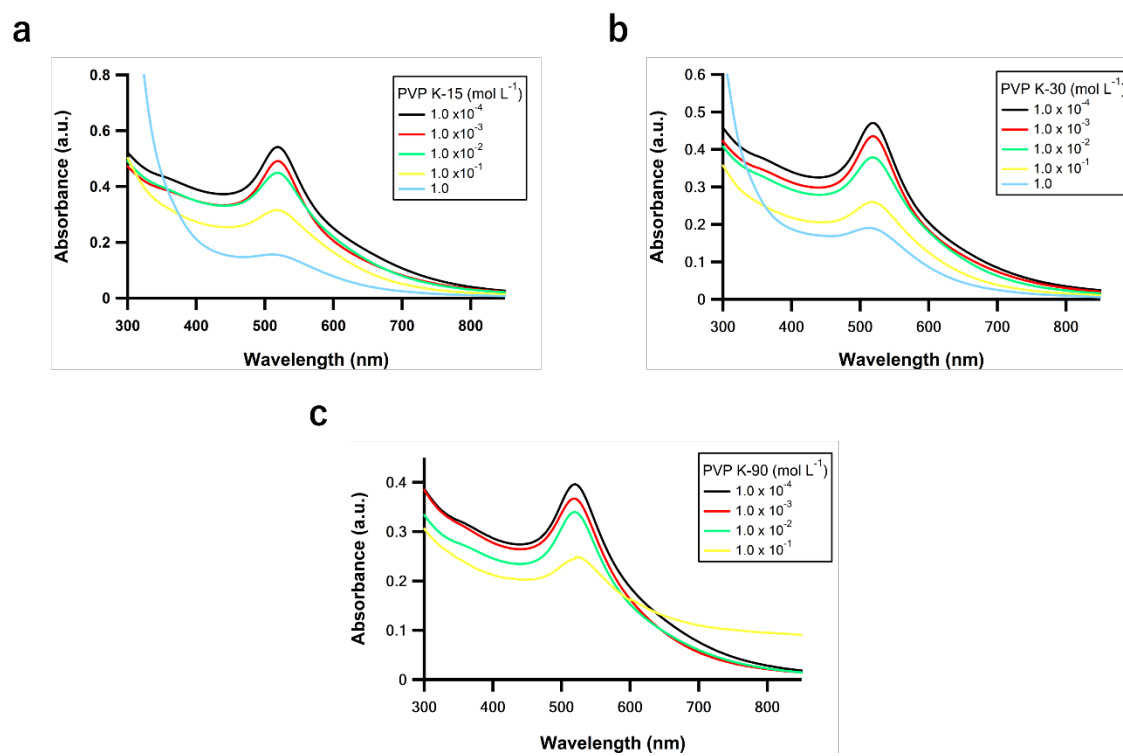
Figure S7. UV/Vis absorption spectrum of Au NPs in different concentration of (**a**) PVP K-15, (**b**) PVP K-30 and (**c**) PVP K-90.

Table S2. Ablated total Au amount in 15 mL solvent with the same viscosity.

PVP type	Viscosity (mPa·s)	Au productivity ($\mu g\ h^{-1}$)
PVP K-15	2.20 ± 0.01	93
PVP K-30	2.23 ± 0.01	92
PVP K-90	2.14 ± 0.12	104
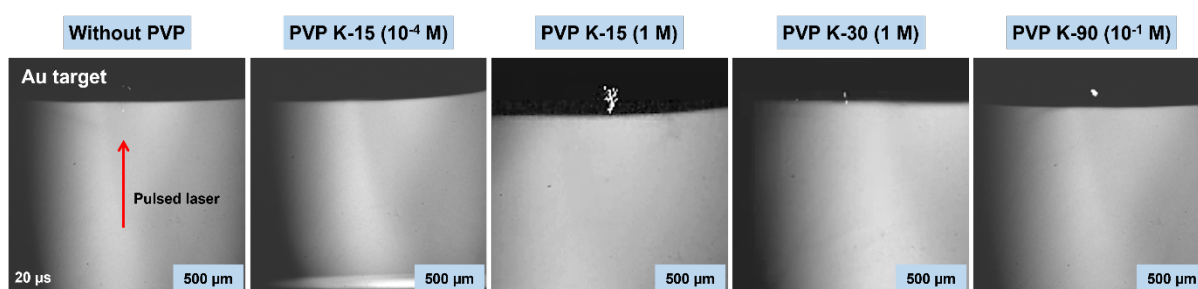
Figure S8. Snapshot images of the target surface corrected at 2 μs after the induction of the laser pulse. From left, without PVP, PVP K-15 (10^{-4} M), PVP K-15 (1 M), PVP K-30 (1 M), and PVP K-90 (10^{-1}M).

References

1) T. Nakamura, Y. Mochidzuki and S. Sato, Fabrication of gold nanoparticles in intense optical field by femtosecond laser irradiation of aqueous solution, *J. Mater. Res*., 2008, **23**, 968–974.

2) Affi N. Hidayah, D. Triyono, Y. Herbani, and R. Saleh, Tuning size and shape of gold nanoparticles using seed-mediated growth by unfocused femtosecond laser-induced plasma, *Opt. Lett*., 2023, **48**, 2126-2129.

3) A. Letzel, S. Reich, T. dos Santos Rolo, A. Kanitz, J. Hoppius, A. Rack, M. P. Olbinado, A. Ostendorf, B. Gokce, A. Plech and S. Barcikowski, Time and mechanism of nanoparticle functionalization by macromolecular ligands during pulsed laser ablation in liquids, *Langmuir*, 2019, **35**, 3038–3047.

4) J. Larez, R. Castell and C. Rojas, Colloids and composite materials Au/PVP and Ag/PVP generated by laser ablation in polymeric liquid environment, *Revista Mexicana de Fisica*, 2016, **62**, 188-192.

5) F. Mafuné, J. Kohno, Y. Takeda, T. Kondow and H. Sawabe, Formation of gold nanoparticles by laser ablation in aqueous solution of surfactant, *J. Phys. Chem. B*, 2001, **105**, 5114–5120.

6) A. V. Kabashin, M. Meunier, C. Kingston and J. H. T. Luong, Fabrication and characterization of gold nanoparticles by femtosecond laser ablation in an aqueous solution of cyclodextrins, *J. Phys. Chem. B*, 2003, **107**, 4527–4531.

7) T. Tsuji, D.-H. Thang, Y. Okazaki, M. Nakanishi, Y. Tsuboi and M. Tsuji, Preparation of silver nanoparticles by laser ablation in polyvinylpyrrolidone solutions, *Appl. Surf. Sci*., 2008, **254**, 5224-5230.