Performance and Ageing of Self-Assembled Metal Electrodes

L. F. Engel 1 [a], J. H. M. Maurer 2 [a], T. Kister 3 [a],
L. González-García 4 [a], T. Kraus 5 [a, b]

[a] *INM - Leibniz Institute for New Materials, Saarbrücken, 66123, Germany*
[b] *Colloid and Interface Chemistry, Saarland University, Saarbrücken, 66123, Germany*

e-mail: tobias.kraus@leibniz-inm.de

Thin metal grids are promising candidates for flexible, transparent electrodes in next-generation optoelectronics. We use a roll-to-roll compatible process to manufacture such grids by nanoimprinting and self-assembly of gold nanowires (AuNWs) and nanospheres (AuNPs) in a reusable silicone stamp [1, 2]. Such nanoscale colloids contain a relatively large fraction of insulating ligand that stabilizes the dispersion. Unless the ligand itself is conductive [3], it has to be removed in order to make the imprinted metal grid conductive. Controlled plasma treatments can remove ligands while retaining the imprinted structure [4]. The combination of self-assembly and plasma treatment leads to complex structures with performances and ageing processes that are not yet well understood.
Here, we discuss the effect of low-power Ar/H_2 room temperature plasma on the performance and ageing of imprinted meshes of AuNPs or AuNWs with oleylamine (OAm) ligand. The plasma yields conducting paths with a metal shell around a metal-OAm core which age by de-wetting on the surfaces and de-mixing of the cores. Ageing seems accelerated by the remaining oleylamine that increases the surface mobility of Au.
We find that the conductivity, optical transmission, and ageing of the electrodes depend on the imprinted nano-object and its concentration. Gold nanowire concentrations ≥ 4 mg/mL led to sheet resistances (R_{sh}) that strongly *in*creased within 24 h. Sheet resistances remained nearly constant for wires and spheres imprinted at 3 mg/mL. Nanospheres imprinted at ≥ 6 mg/ml formed electrodes with sheet resistances that *de*creased after 3-5 days by up to 80 % (Fig. 1). We correlated these findings with differences in pre- and post-plasma morphology (Fig. 2) from SAXS, TEM, SEM, AFM and laser confocal microscopy. The insights gained from these studies enabled us to develop strategies against electrode degradation.
Our results (Fig. 3) show that the performance of electrodes made from wires are initially superior to those made from spheres. After ageing, electrodes made from spheres outperformed those made from wires because they use the metal more efficiently.

[1] J. H. M. Maurer et al., Adv. Mater. Technol., 2 (2017), 1700034
[2] T. Kister and J. H. M. Maurer et al., ACS Appl. Mater. Interface, 10 (2018), 6079-6083
[3] B. Reiser et al., Chem. Sci., 7 (2016), 4190
[4] J. H. M. Maurer et al., Nano Lett., 16 (2016), 2921-2925

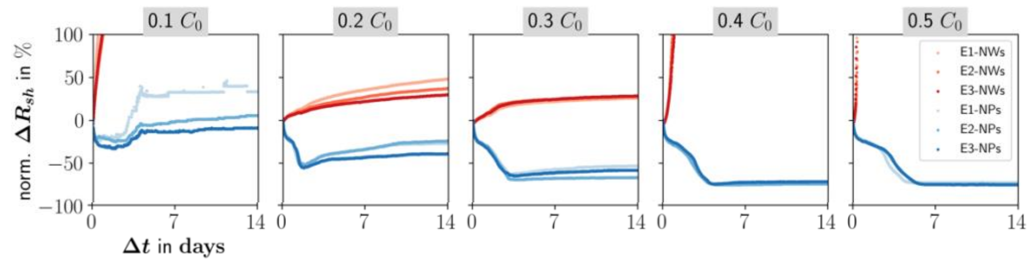

Figure 1: Normalized change in R_{sh} with time for AuNW (in red) and AuNP (in blue) based electrodes imprinted at different C_{Au} (AuNWs: C_0 = 10 mg/ml, AuNPs: C_0 = 30 mg/ml). Three samples (E1-3) were tested for both AuNW and AuNP based electrodes at any given C_{Au} .

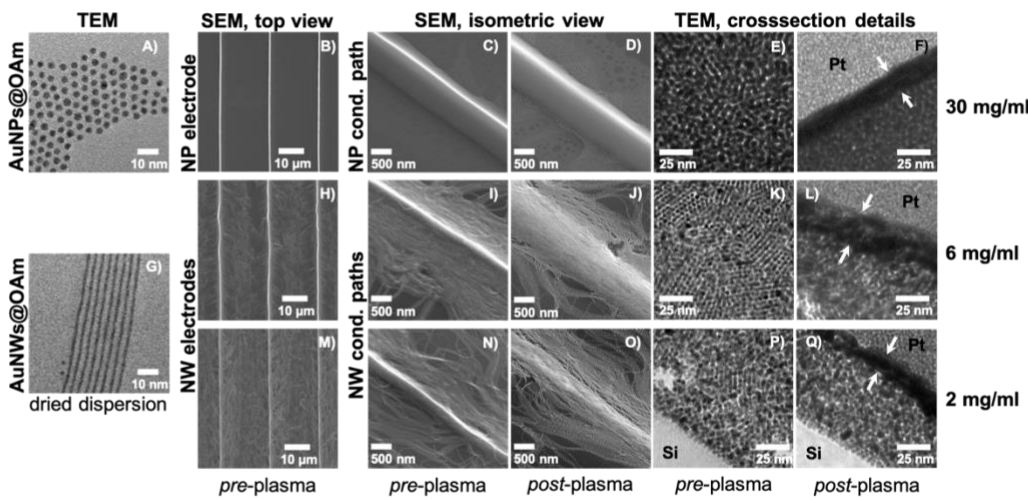

Figure 2: Morphological analysis of electrode grids printed from AuNWs and AuNPs before and after plasma treatment.

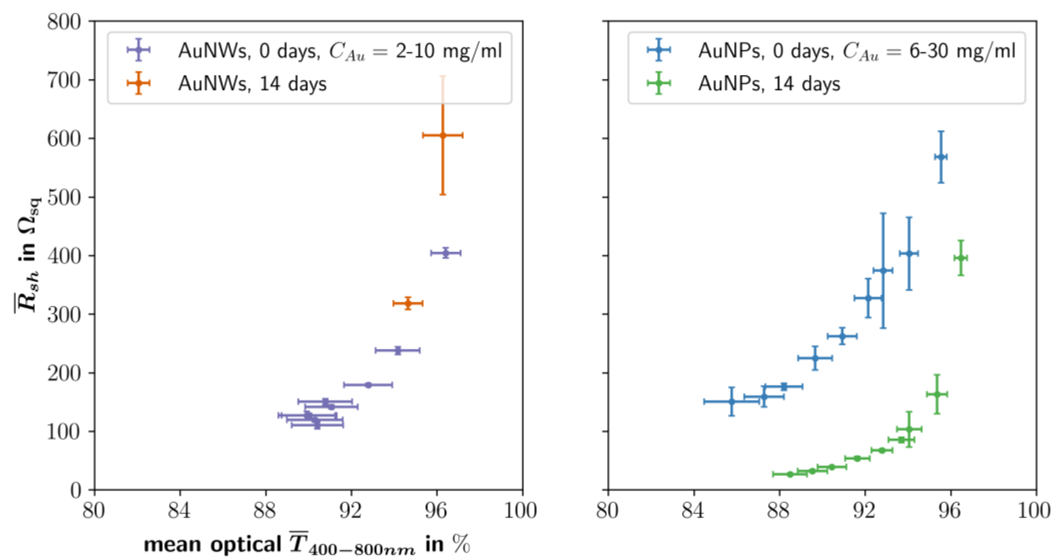

Figure 3: R_{sh} and mean optical transmittance before and after ageing imprinted electrodes made from self-assembled AuNWs (left) and AuNPs (right).