

Substrate Cleaning Protocol

For basic cleaning, the glass substrates (soda-lime 0215 Corning Glass, Corning, Corning, NY, USA) 75x50x1 mm in size were sonicated in a bath with Micro-90 cleaning solution (International Products Corporation, Burlington Township, NJ, USA), diluted with bi-distilled water (DI) at 1:50 at 80 °C for 10 min in an ultrasonic cleaner (GT Sonic-P3, GT Sonic, Shenzhen, China). The substrates were rinsed with deionized water to remove the residual of the cleaning solution and then kept in an ultrasonic bath with deionized water for 10 min. Further, the substrates were dried in airflow and sonicated in isopropanol for 10 min at 80 °C. After that, the substrates were blown with dry nitrogen, immersed in boiling bi-distilled water (Milli-Q® IQ 7003, Merck, Darmstadt, Germany) for 5 s, dried in a nitrogen flow, and finally heated in an oven (MRC MSF 11-4, MRC, Holon, Israel) for 20 min at 120 °C. Clean substrates were stored in a desiccator under a vacuum. Before use, the glasses were cleaved into squares of 25 mm × 25 mm. Each substrate was rinsed in the spincoater (WS-650HZ-23NPPB, Laurell Technologies Inc, North Wales, PA, USA) subsequently with 3 mL bi-distilled water, 0.5 mL isopropanol, 3 mL DI water, 0.5 mL isopropanol, 3 mL bi-distilled water (×2 times) at 4000 rpm, and finally spin-dried at 6000 rpm for 15 s. After rinsing, glasses were dried in the oven for 20 min at 120 °C. Just before the vacuum deposition, the substrates were processed in a vacuum plasma cleaner (MTI Corp. EQ-PCE3, Richmond, CA, USA) for 2 min at the “High” regime in residual air at 0.18 Torr.

The Detailed Description of Samples N1–N5

To reveal the composition of the ink, five types of solutions were prepared, referred to in the text as N1–N5, and their parameters were measured and compared.

- N1—The ethylene glycol/water (EG/H₂O) solution (3:2 by volume) was prepared by mixing 4 mL DI water with 6 mL EG under stirring until complete dissolution.
- N2—The colloidal solution of 0.125 g titania nanoparticles (TNP) in 10 mL EG/H₂O (3:2) was prepared, with the same concentration as in ink, except that the solution was not boiled and no H₂AuCl₄ was added.
- N3—The complex gold nanoparticles (AuCNP) precursor, all the ink components were mixed and simply stirred at room temperature, without any ink preparation procedures presented in the Experimental Section.
- N4—The AuCNP ink without EG—0.125 gr of TNP in 8 mL DI water was heated until boiling point. The acid solution, which consisted of 0.0677 g H₂AuCl₄·3H₂O mixed with 2 mL DI water and 1.2 µl Tween 80, was quickly added to the flask with boiling TNP colloid. After 10 min of constant stirring, the dispersion was cooled to room temperature.
- N5—The AuCNP ink—the preparation is presented in Experimental Section.

The images of the samples N1, N2, N3, and N5 are presented in Figure S1.

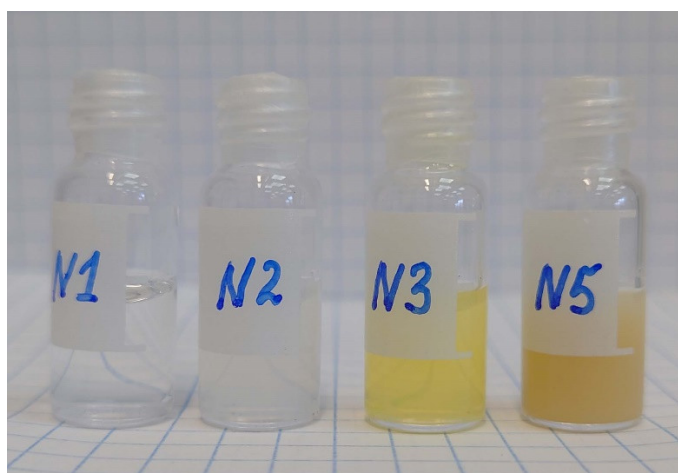


Figure S1. The images of the samples N1, N2, N3, and N5.

Preparation of TiO₂ Nanoparticles

A low-temperature peptization method was used to synthesize TiO₂ NPs (TNP) [1,2]. First, 0.7 mL of HNO₃ were mixed in 100 mL DI water. The solution was heated to 70 °C during intensive stirring with a magnetic stirrer. Then, 12 mL of isopropanol was added to the heated solution. After this, the 14 mL of titanium isopropoxide was slowly dripped into the dispersion. The resulting mixture was stirred at 600 rpm for 45 min at 80 °C, covered with foil, and stirred for five days at room temperature. After that, a colloid solution of titania nanoparticles was transferred to a plastic cup for slow fluids evaporation for five days so that only the solid phase remained. Then, the obtained xerogel of the nanoparticles of about 40 nm was collected (without calcination) and saved at room temperature for further use. The hydrodynamic diameter (HDD) of TNP nanoparticles is presented in Figure S2.

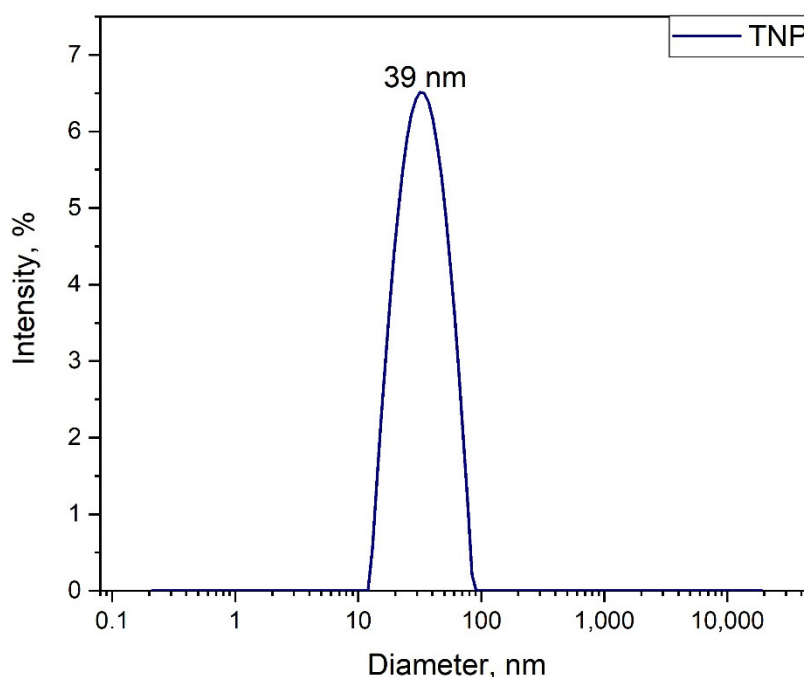


Figure S2. Particle size distribution of colloidal TNP obtained by DLS.

The image of the TiO₂/AuNP film ink-jet printed with 10-day-aged AuCNP ink on a glass substrate is presented in Figure S3.

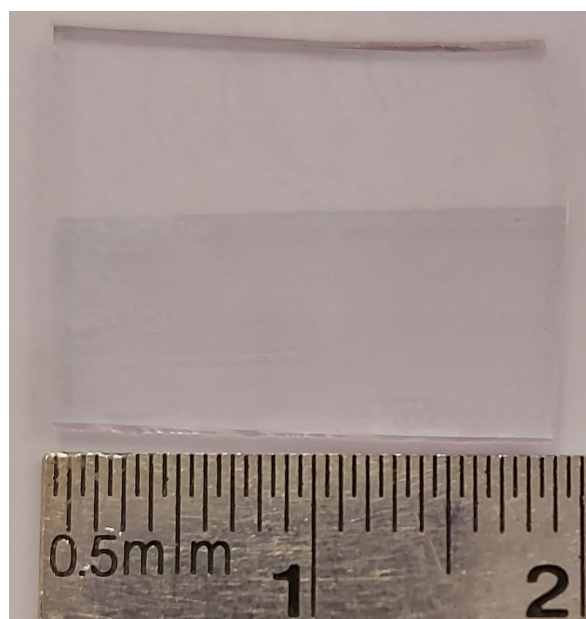


Figure S3. The image of the TiO₂/AuNP film ink-jet printed with 10-day-aged AuCNP ink on a glass substrate.

SEM images of the cross-section of the TiO₂/AuNP film ink-jet printed with 10-day-aged AuCNP ink on a glass substrate are presented in Figure S4. The images were recorded simultaneously with secondary electrons (SE) and backscattering electrons (BSE) In-Beam detectors from the same area of the sample. The bright spots on the BSE image correspond to the AuNPs.

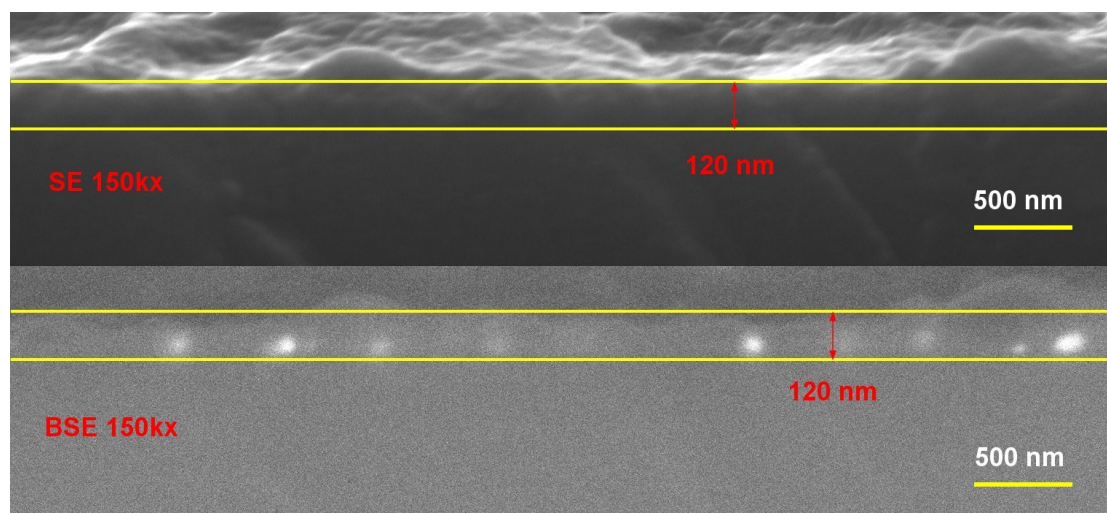


Figure S4. The SEM images of the cross-section of the TiO₂/AuNP film.

The results of twenty-five weeks of monitoring the complex gold nanoparticles (AuCNP) HDD in ink are presented in Figure S5. It was revealed that over time the dispersion remained stable, and there is a clear tendency towards a decrease in both HDD of particles and the standard deviation.

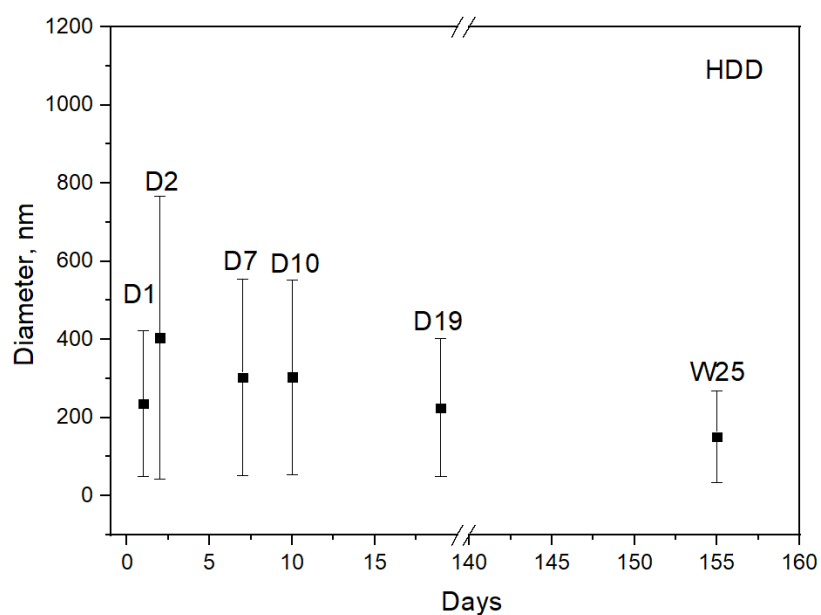


Figure S5. The day-age dependence of the AuCNP nanoparticles' HDD. The bars on the plot correspond to standard deviation.

References

1. Keller, K.; Khramenkova, E.V.; Slabov, V.; Musin, A.; Kalashnikov, A.; Vinogradov, A.V.; Pidko, E.A. Inkjet Printing of Sc-Doped TiO₂ with Enhanced Photoactivity. *Coatings* **2019**, *9*, 78, doi:10.3390/coatings9020078.
2. Behnajady, M.A.; Eskandarloo, H.; Modirshahla, N.; Shokri, M. Sol-gel low-temperature synthesis of stable anatase-type TiO₂ nanoparticles under different conditions and its photocatalytic activity. *Photochem. Photobiol.* **2011**, *87*, 1002–1008, doi:10.1111/j.1751-1097.2011.00954.x.