

## Electronic Supporting information

### Polyvinylpyrrolidone-mediated synthesis of ultra-stable gold nanoparticles in nonaqueous choline chloride-urea deep eutectic solvent

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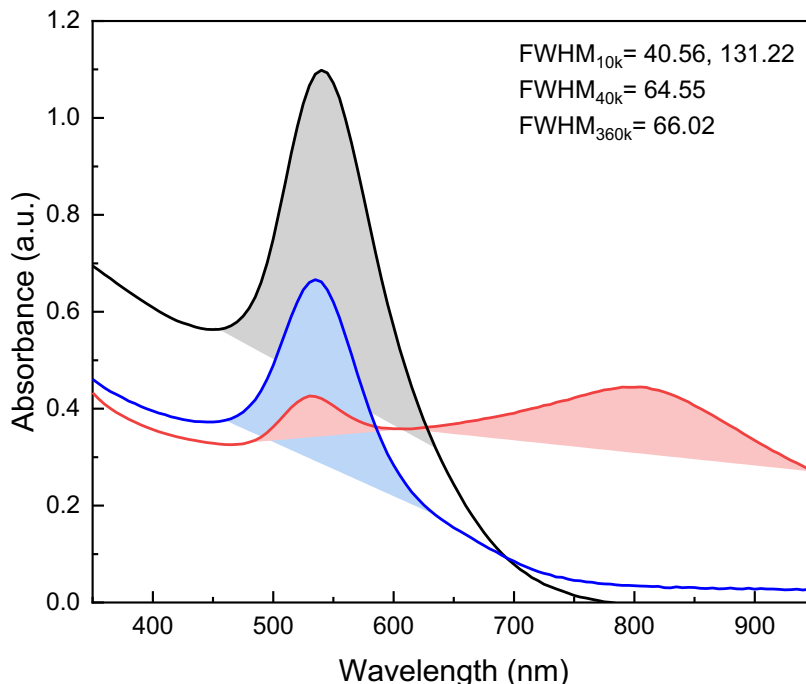


Figure S1. UV-Vis absorption spectra of the AuNPs synthesized with different  $M_n$  of PVP (10k-red, 40k-black and 360k-blue) highlighting the area under the curve utilized to calculate the FWHM values in the graph.

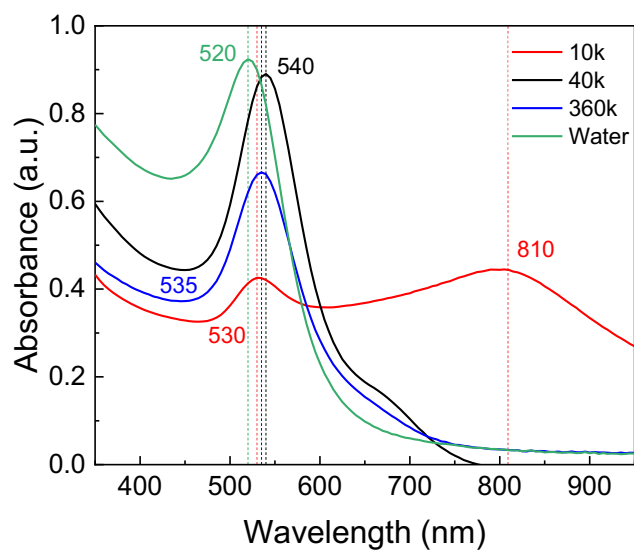


Figure S2. Digital photograph showcasing the different gold colloids in DES, prepared with PVP 10k, 40k and 360k and 40k in water, along with their respective UV-Vis spectra.

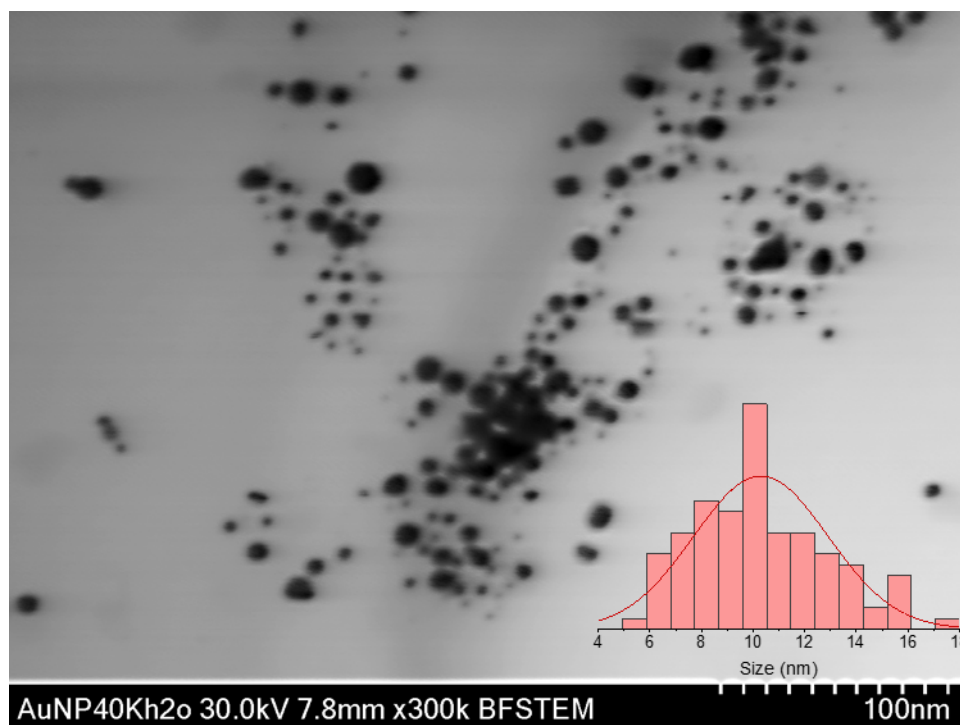


Figure S3. STEM micrograph of AuNPs stabilized with 40k PVP in water. Inset: particle size distribution histogram ( $10.3 \pm 2.6$  nm).

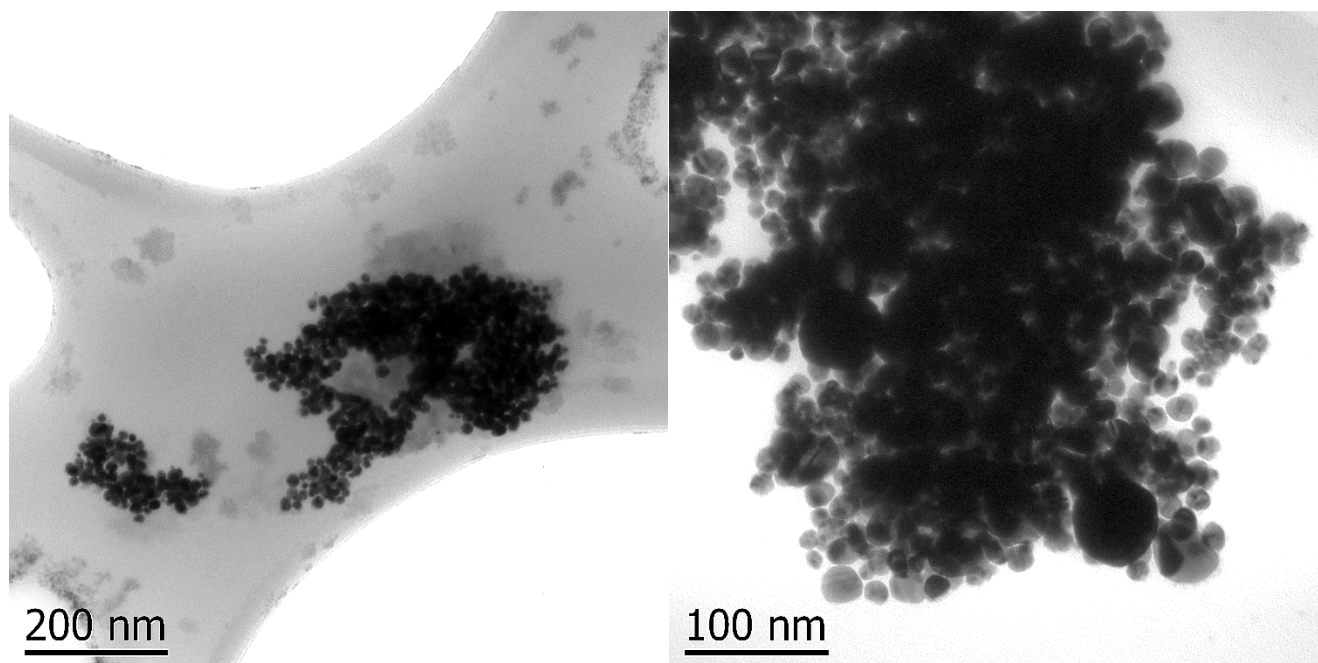


Figure S4. TEM micrograph of AuNPs stabilized with 10 kDa PVP, showing polydispersed nanoparticles and agglomerations responsible for the plasmon around 810 nm.

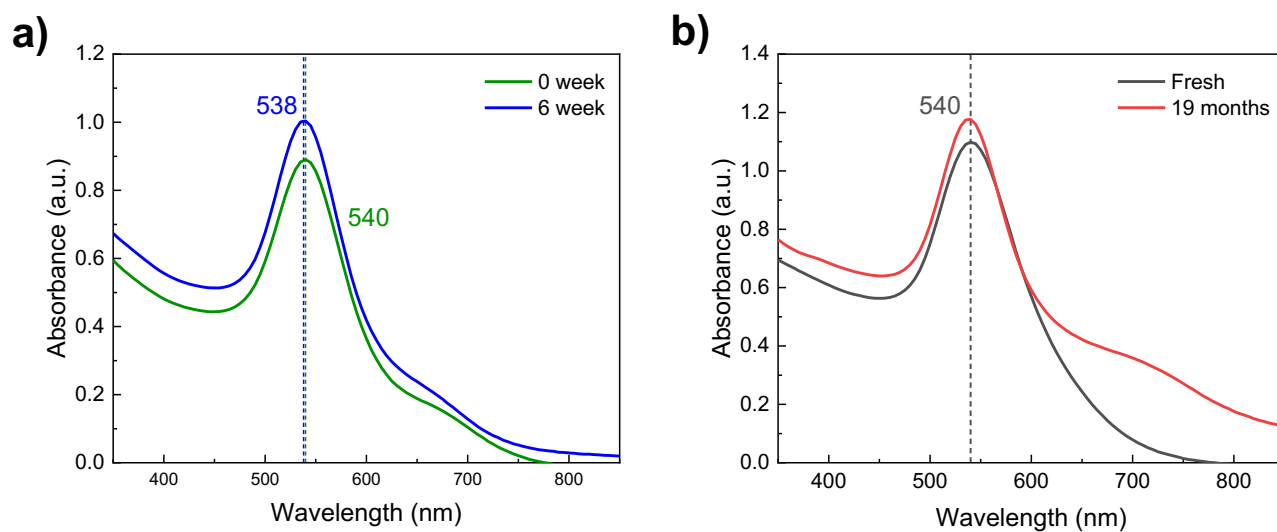
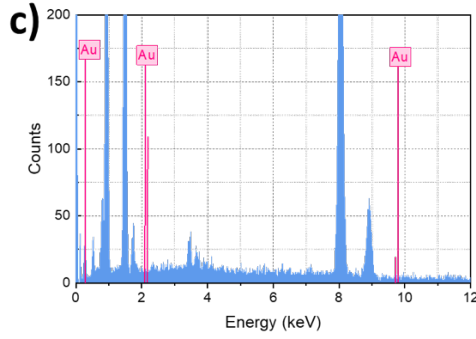
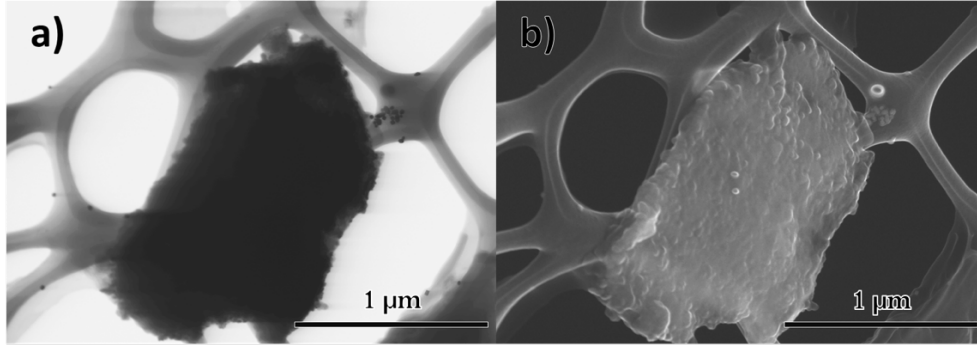
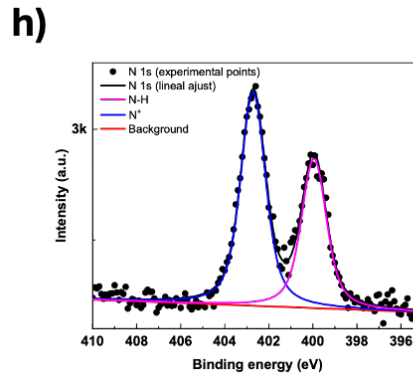
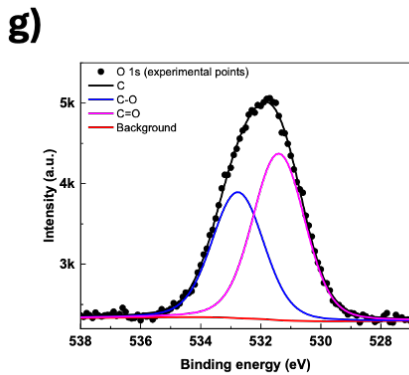
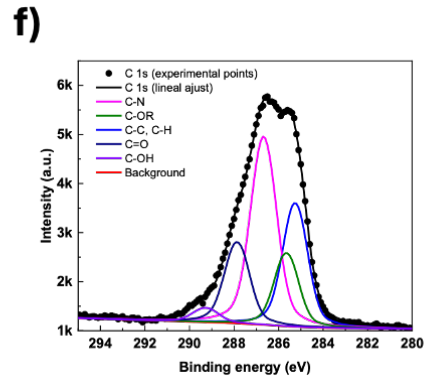
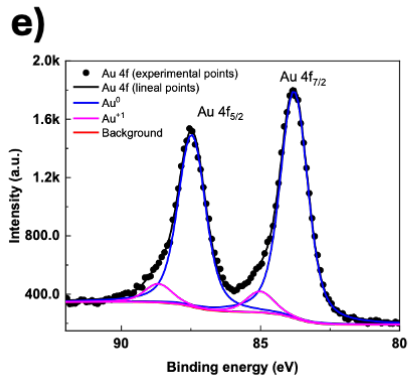


Figure S5. Comparison of the stability of different batches of AuNPs synthesized in a ChCl-U DES using 40k PVP (a) after synthesizing and 6 weeks later and (b) 19 months later.



**d)**

| El   | AN | Series   | [wt. %] | [norm. wt. %] | [norm. at. %] | Error in wt. % (1 Sigma) |
|------|----|----------|---------|---------------|---------------|--------------------------|
| C    | 6  | K-series | 17.902  | 17.755        | 43.042        | 9.32866                  |
| O    | 8  | K-series | 6.8693  | 6.813         | 12.399        | 3.72228                  |
| Al   | 13 | K-series | 16.461  | 16.326        | 17.618        | 1.13385                  |
| Si   | 14 | K-series | 1.1094  | 1.1003        | 1.1407        | 0.19272                  |
| Cu   | 29 | K-series | 54.896  | 54.446        | 24.948        | 1.72243                  |
| Sn   | 50 | L-series | 3.3674  | 3.3398        | 0.8192        | 0.28468                  |
| Au   | 79 | L-series | 0.2223  | 0.2204        | 0.0326        | 0.13036                  |
| Sum: |    |          | 100.83  | 100           | 100           |                          |



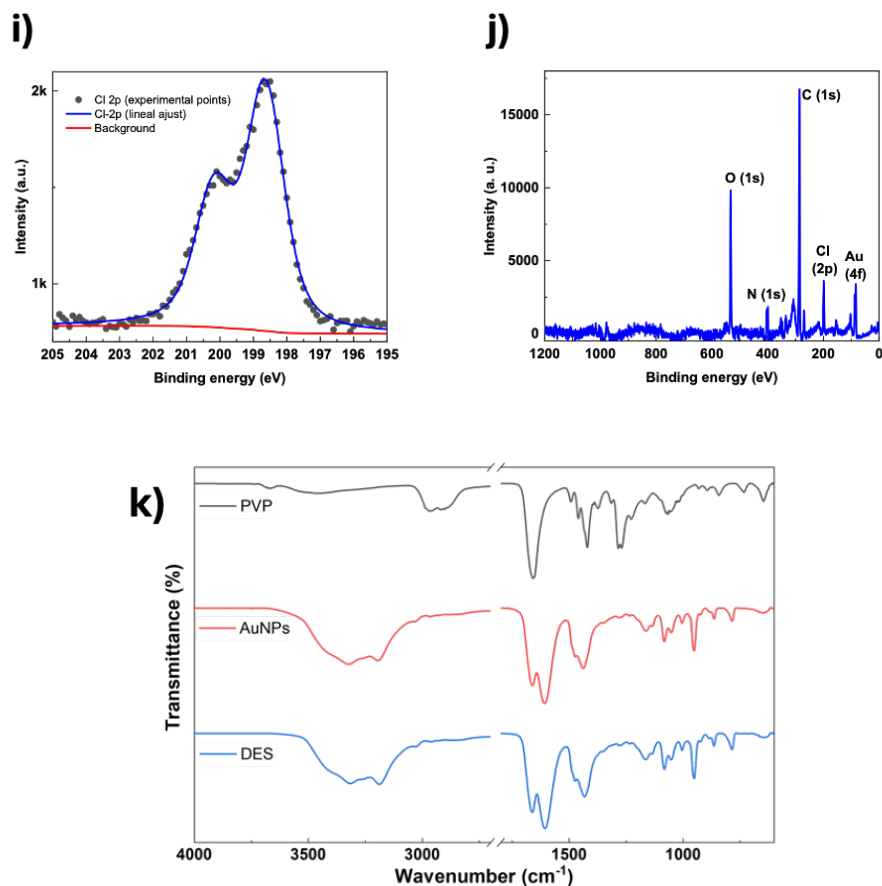


Figure S6. (a) Transmission and (b) scanning electron micrographs of the AuNPs synthesized with 40kDa PVP, (c) energy dispersive spectroscopy (EDS) energy graph of the selected area, highlighting the energies corresponding to gold and (d) element quantities, (e-i) high-resolution spectra of Au, C, O, N, Cl and (j) general survey analyzed by XPS, and (k) ATR-FTIR of PVP, AuNPs precipitated from DES and U-ChCl DES.

For SEM/EDS characterization of 40k PVP AuNPs it can be observed (Fig. Sb), through secondary and backscattered electrons, the presence of gold nanoparticles, appearing as bright dots due to their higher atomic number in Z-contrast mode, surrounded and capped by the PVP polymer. The morphology of the nanoparticles can be appreciated more clearly in Figure S6a, where the polymer appears almost transparent, sharpening the nanoparticles' borders.

On the other hand, XPS traces above show the presence of zero-valent gold due to the AuNPs and C, O, N and Cl due to PVP and DES residues.

For the FTIR-ATR spectra of 40k PVP AuNPs precipitated from the DES, it can be observed that all PVP signals from the PVP-capped AuNPs overlap with those from the remaining DES. Specifically, it is evident that the intense carbonyl signals from PVP at 1669 and 1620 cm<sup>-1</sup> are due to C=O stretching and N-H scissoring vibrations, respectively, of urea. Additionally, the intense CH<sub>2</sub> bending of an alkyl group at 1447 cm<sup>-1</sup> is also observed.

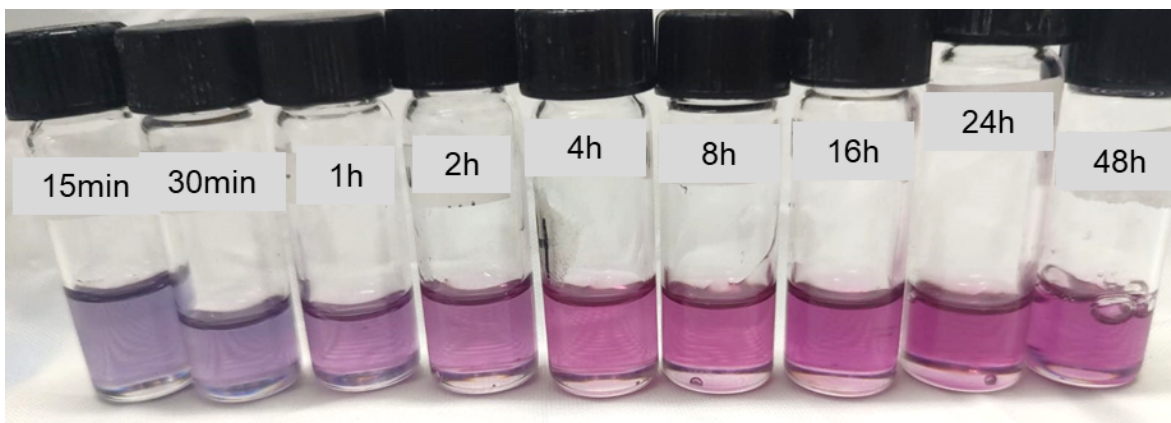
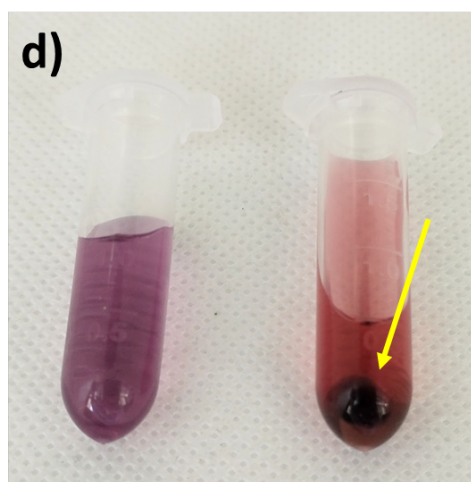
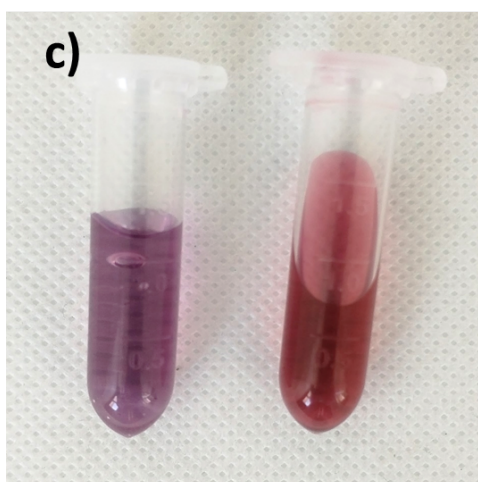
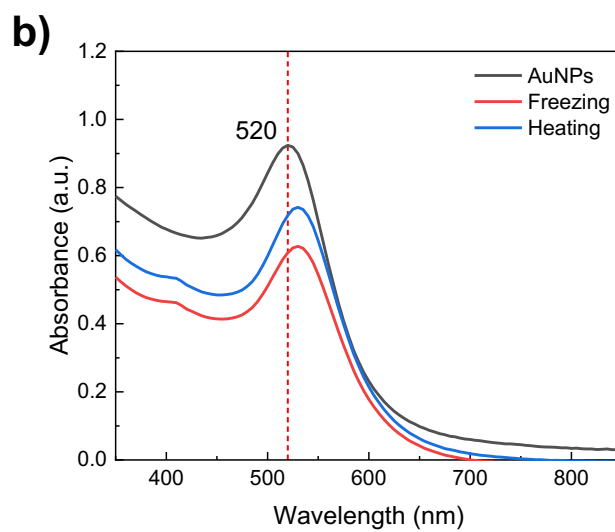
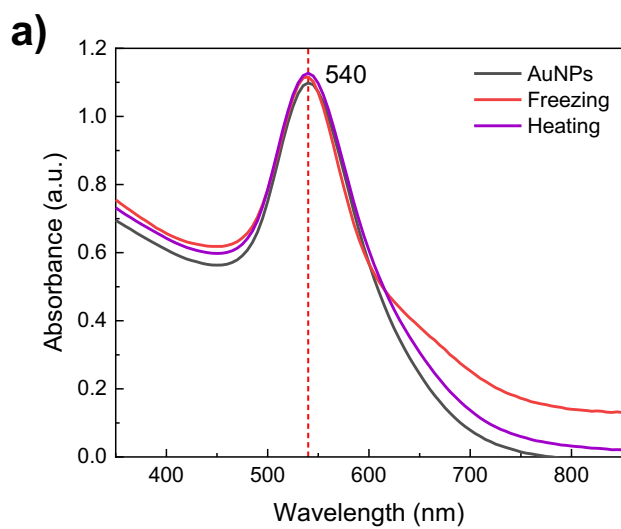


Figure S7. Digital photography of the gold colloids as the reaction time increases (40 k PVP).



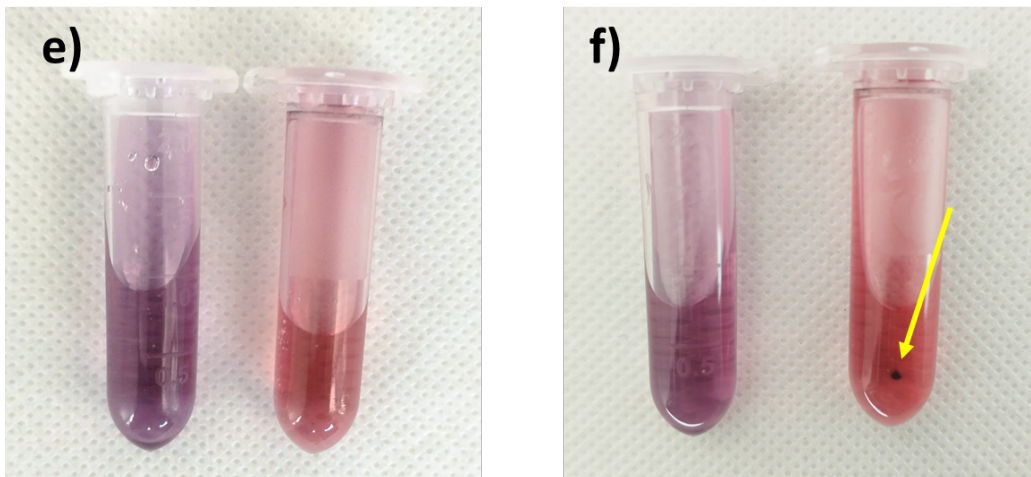


Figure S8. Comparison of the changes in the plasmon after freezing ( $-4^{\circ}\text{C}$ ) and heating ( $90^{\circ}\text{C}$ ) of AuNPs synthesized with 40k PVP in DES (a) and water (b). Contrast in the colloidal stability of AuNPs synthesized with 40k PVP in DES (purple) and water (red), (c) before and (d) after centrifugating at 10,000 RPM for 10 minutes, and (e) before and after (f) 2 months of sunlight exposure.

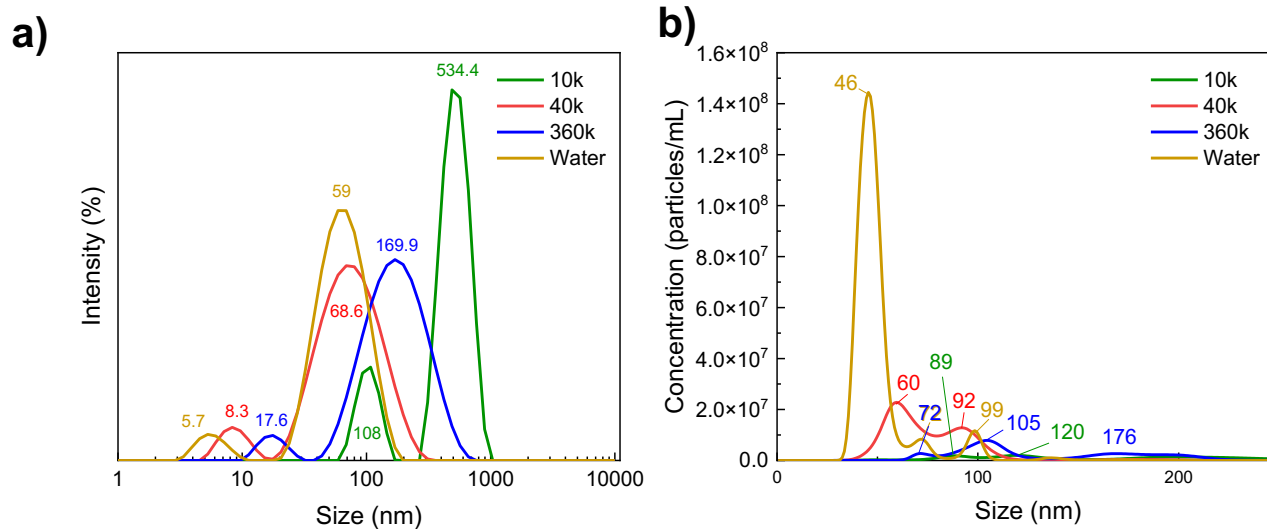


Figure S9. Comparisons of the (a) Dynamic light scattering (DLS) and (b) nanoparticle tracking analysis (NTA) of the AuNPs synthesized in water (40 kDa PVP) and in DES using 10, 40 and 360 kDa PVP.

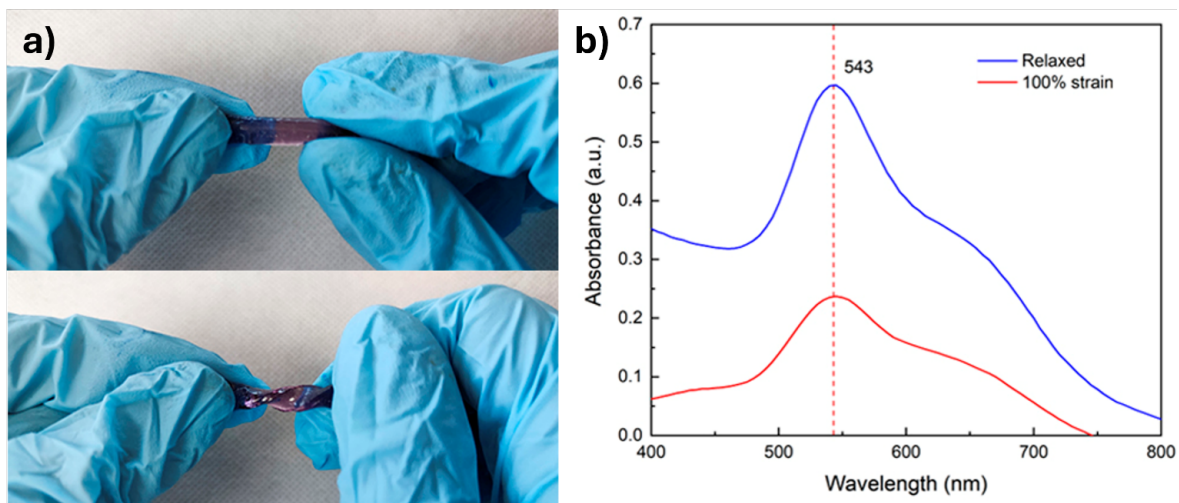


Figure S10. a) eutectogel prepared with 22 wt% gelatin in the gold colloid in DES and b) UV-Vis spectra of the plasmonic eutectogel relaxed and stretched at 100% of its original length.

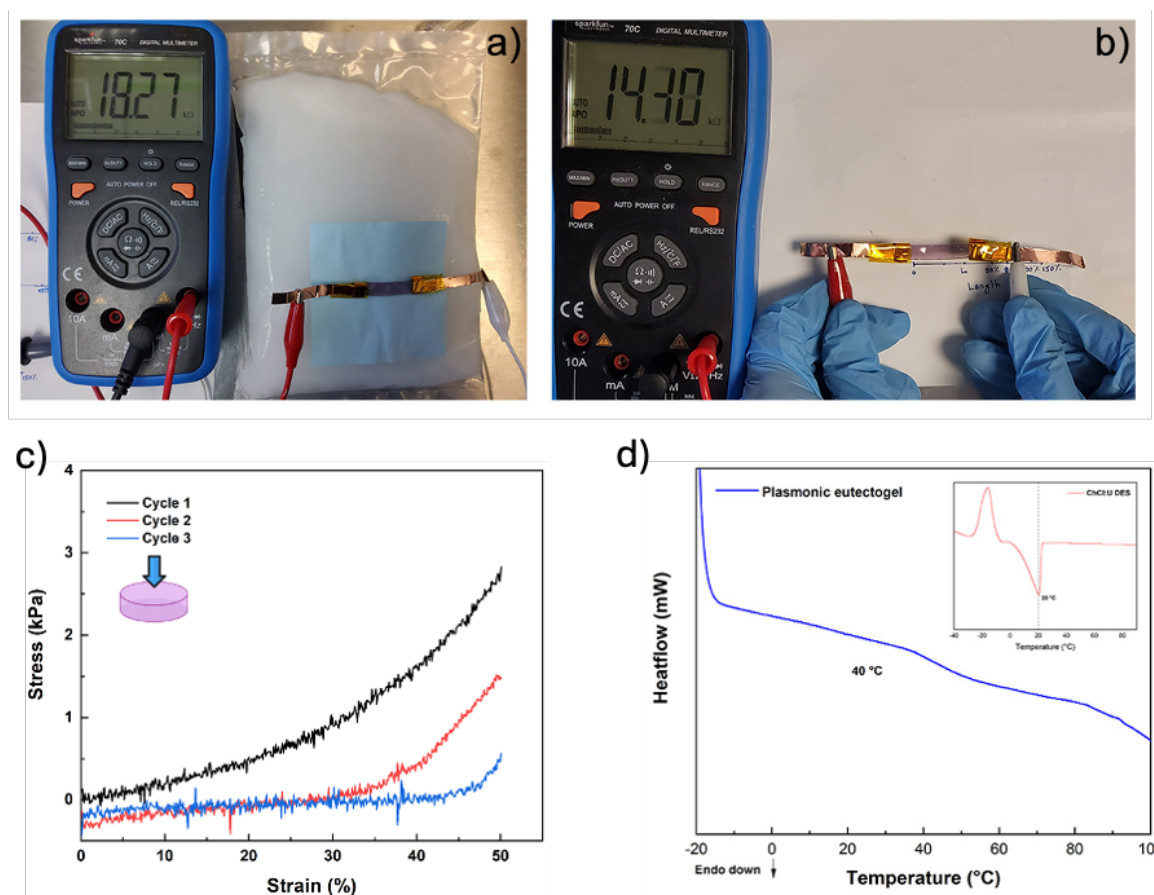


Figure S11. (a) Set up for the GF calculations: Copper tape is attached to the sides of the eutectogel sample to measure the resistance change upon stretching. b) The gel is stretched at 50, 80, 100 and 150% of its original length, and the resistance change is recorded with a



USB digital multimeter. (c) Compressive stress-strain cycles and (b) thermogram of the plasmonic aerogel (mp=40 °C) with an inset of the thermogram for U-ChCl DES (mp=20 °C).

Table S1. Recorded data from the digital multimeter at different deformations of the plasmonic eutectogel.

| Length (cm) | Strain (%) | R (kOhm) | $\Delta R$ (%) |
|-------------|------------|----------|----------------|
| 2.4         | 0          | 13       | 0              |
| 3.6         | 50         | 18       | 34.8           |
| 4.3         | 80         | 20       | 49.8           |
| 4.8         | 100        | 22       | 64.8           |
| 6.0         | 150        | 26       | 94.8           |