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Supporting Information

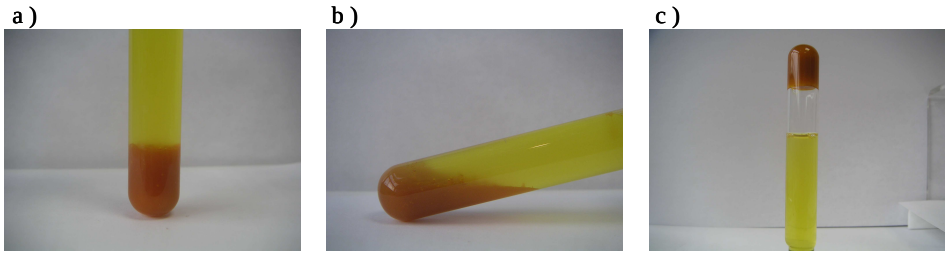

Fig. 1 Photographs of the same sample prepared at 40°C and pH 3.4 with [gelatin] = 8 wt.% / [vanadates] = 120 mM: a) and b) just after preparation at 40°C ; c) after 3 h at 20°C.

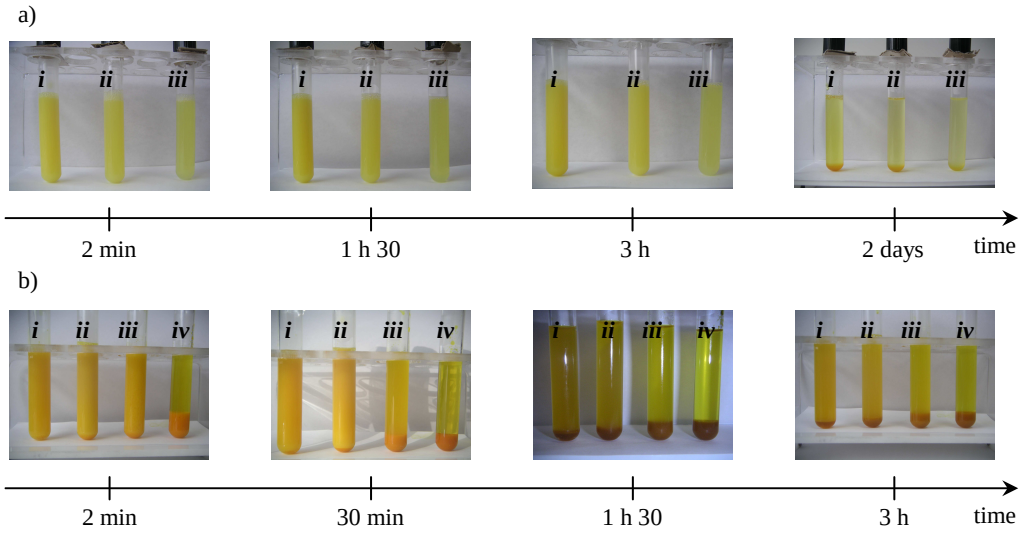

Fig. 2 Photographs taken upon ageing conditions without stirring (T = 40°C, pH 3.4) with gelatin in: a) dilute regime (*i*. c = 1 wt.%, *ii*. c = 0.5 wt.%, *iii*. c = 0.2 wt.%) and different vanadate concentrations: *i*. c = 5 mM, *ii*. c = 3.5 mM, *iii*. c = 2.5 mM ; b) semi-dilute regime (c = 8 wt.%) and different vanadate concentrations: *i*. c = 50 mM, *ii*. c = 60 mM, *iii*. c = 70 mM, *iv*. c = 75 mM.

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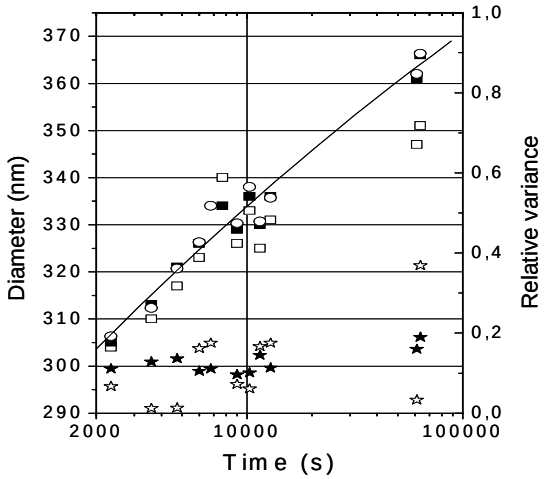

Fig 3. Evolution of colloids size as measured by DLS upon ageing conditions without stirring (T = 40°C, pH 3.4) for [G] = 0.5 wt.% and [V] = 3.5 mM. The correlation function have been fitted by different methods: Cumulant Quartic (□), Cumulant Quadratic (■) and NNLS (○). The stars represent respectively the relative variance associated to the quadratic data (full symbols) and the relative variance associated with NNLS data (open symbols). The continuous line corresponds to a second order polynomial fit of the Cumulant quadratic data and should be used as a guide for the eyes.

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Experimental

Materials. Gelatin, $NaVO_3$, HCl, and NaCl were purchased from Sigma-Aldrich in their reagent grade and used without further purification. Deionized water obtained with a MilliQ system (Millipore) was used as solvent in all the experiments.

- *Preparation of vanadate solutions.* Polyoxovanadate solutions have been prepared by acidification of an aqueous solution of sodium metavanadate $NaVO_3$ (Sigma, >99%, 0.1 $mol.L^{-1}$, pH = 8) with an aqueous HCl solution (2N). Once the polyoxovanadate solution is prepared with the desired temperature and pH, this solution is added to an aqueous gelatin solution.

- *Preparation of Gelatin solutions.* Gelatin extracted from porcine skin (type A with pI close to 8 and a bloom of ~175 g corresponding to an average molecular weight of ~40000 g/mol according to Sigma-Aldrich). Gelatin solutions were prepared by swelling the gelatin granules in an aqueous solution during a minimum of 3 h at 5 °C. Gelatin was then dissolved at 40 °C using a magnetic stirrer for 30 min at 300 rpm. When gelatin is dissolved, the pH and ionic strength are adjusted with respectively an aqueous HCl solution (2N) and NaCl.

- *Preparation of mixtures.* Mixtures were prepared by slowly adding dilute vanadate solutions in gelatin solutions under stirring (v = 300 rpm). The final solutions were stirred during 2 min prior to any observations. In fact, we have checked that when turbid samples are still present after 2 min of stirring, further stirring periods lead to macroscopic phase separation and never to redissolution. After these initial observations, the samples are aged in an oven at 40°C.

Methods. ^{51}V liquid NMR spectroscopy. ^{51}V liquid NMR spectra were recorded at 78.9 MHz on a Bruker Avance 300 spectrometer. The chemical shifts were referenced to $VOCl_3$ (δ = 0 ppm). A spectral width of 31 kHz, a pulse width of 16 µs, with an accumulation time of 0.25 s and no relaxation delay, were used. An accumulation of 1000 transients was usually performed on each sample. The 10 mm NMR tubes were filled with 4 ml of vanadate / gelatin solutions.

Dynamical light scattering (DLS). Dynamical light scattering was performed on a Brookhaven spectrometer (BI-9000AT autocorrelator). The autocorrelation functions were recorded at θ = 90°C and analyzed by the Cumulants technique for the determination of the diffusion constant $D(c)$. From the extrapolation of the quadratic diffusion coefficient to infinite dilution, the hydrodynamic radius (R_H) of the colloids was determined through the

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Stokes-Einstein relation, $R_H = k_B T/6\pi\eta_S D_{(c \rightarrow 0)}$, where k_B is the Boltzmann constant, T the temperature ($T = 338\ K$) *and* η_S the solvent viscosity at 338 K.