

Yield precursor in primary creep of colloidal gels

Electronic Supplementary Information

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1 Quantification of sample aging

A rheometric measure of the rate of sample aging is given by the mutation time τ_{mu} defined as

$$\tau_{mu} \equiv \left(\frac{1}{G^*} \frac{dG^*}{dt_s} \right)^{-1} = \left(\frac{d \ln G^*}{dt_s} \right)^{-1}, \quad (S1)$$

where $G^* \equiv \sqrt{G'^2 + G''^2}$ denotes the complex modulus and t_s the sample time since the initiation of gelation.^{S1} For each experiment, starting from $t_s = 0$ s we perform (1) a time sweep with a small oscillatory strain amplitude γ_0 in the linear deformation regime ($\gamma_0 = 4 \times 10^{-3}$ and 8×10^{-2} for the strain-softening and the strain-hardening gel, respectively) at a frequency $\omega = 6.3 \text{ rad s}^{-1}$ for 1800 s, (2) a frequency sweep at the same strain amplitude γ_0 for approximately 400 s, and (3) the creep test. By fitting a linear function to $\ln G^*(t)$ obtained during the last 100 or 200 s of the time sweep step, as shown in Fig. S1, we estimate the mutation times τ_{mu} to be $(4.37 \pm 0.36) \times 10^3$ s and $(1.34 \pm 0.70) \times 10^4$ s for the strain-softening and the strain-hardening gel, respectively. We underscore that these values are conservative estimates of the mutation time τ_{mu} . As $\ln G^*/dt_s$ keeps decreasing with t_s , the true value of τ_{mu} at the beginning of the creep step that starts ~ 400 s after the end of the time sweep is likely greater than the values obtained here.

The extent of sample aging during a measurement can be quantified as the mutation number $N_{mu} \equiv \Delta t/\tau_{mu}$, where Δt denotes the duration of the experiment.^{S1} Effects of aging on the yield precursor $\dot{\gamma} \sim t^{-0.6}$

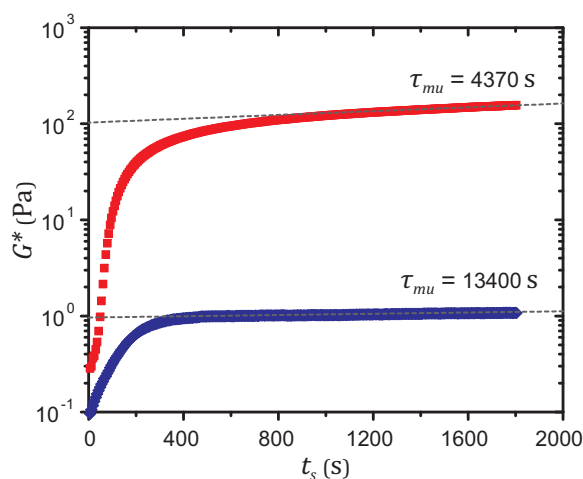


Fig. S1: Temporal change in the complex modulus G^* for a frequency $\omega = 6.3 \text{ rad s}^{-1}$ of the strain-softening (red squares) and the strain-hardening (blue diamonds) gels. The mutation time τ_{mu} is obtained from a linear fit to the last 100 s (strain-softening) or 200 s (strain-hardening) of the data sets.

can be neglected if $N_{mu} \ll 1$, where Δt is defined as the end time of the yield precursor, corresponding to the time at which the strain γ reaches 0.5 and 1.0 for the strain-softening and the strain-hardening gel, respectively. For the strain-softening gel, N_{mu} is lower than 0.10, while for the strain-hardening gel, $N_{mu} < 0.04$. A frequently accepted threshold value of N_{mu} below which aging can be neglected is 0.15,^{S1} which justifies our assumption that aging does not affect the yield precursor during primary creep. For the longest experiments presented in Fig. 2 of the main text, however, yield is not observed until $N_{mu} \simeq 1.21$ and 0.33 for the strain-softening (at the applied stress $\sigma_0 = 1.5$ Pa) and the strain-hardening gel (at $\sigma_0 = 1.0$ Pa), respectively. In these two experiments, therefore, aging of the gels likely affects the later-stage deformations after the power law $\dot{\gamma} \sim t^{-0.6}$, suppressing yield even at large strains γ .

2 Full ranges of the strain $\gamma(t)$ and the shear rate $\dot{\gamma}(t)$

The full ranges of the strain $\gamma(t)$ and the shear rate $\dot{\gamma}(t)$ of the strain-softening and the strain-hardening gels are shown in Fig. S2(A to D) for a particle volume fraction $\phi = 5.0\%$. The shear rate $\dot{\gamma}(t)$ after delayed yielding approaches a constant value for fluidized samples.

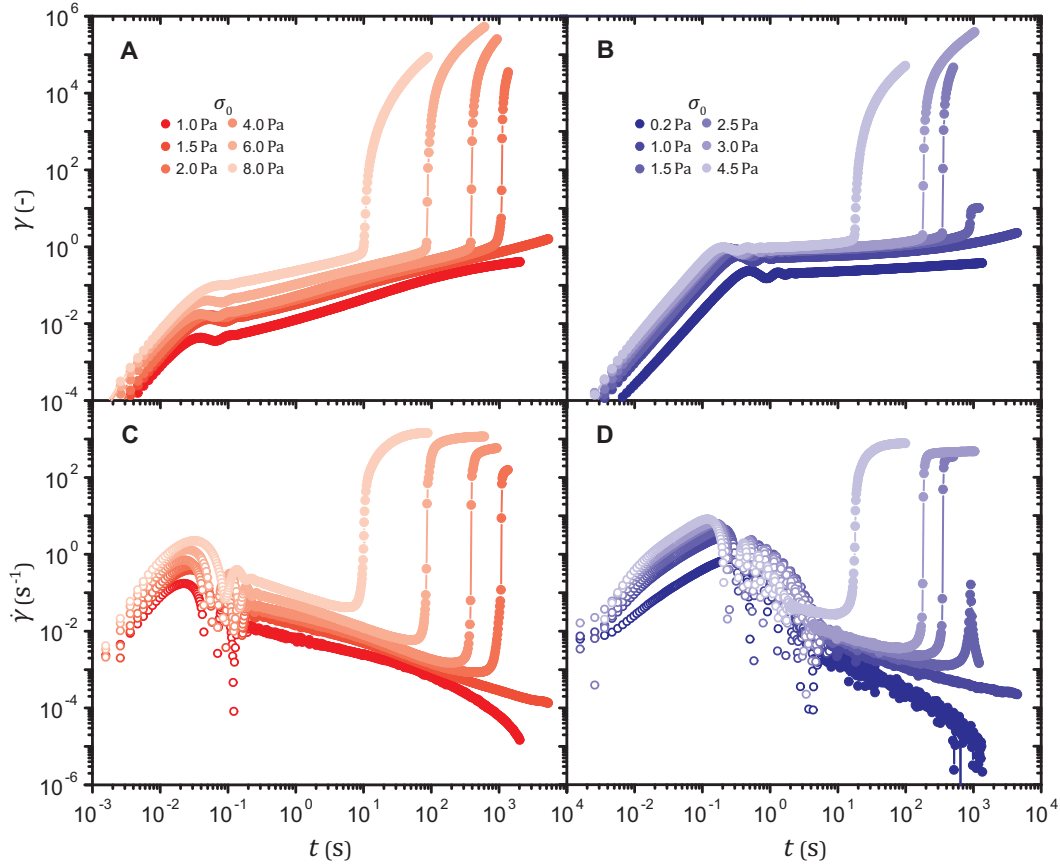


Fig. S2: (A and B) Strain $\gamma(t)$ and (C and D) shear rate $\dot{\gamma}(t)$ of (A and C) the strain-softening and (B and D) the strain-hardening gels for a particle volume fraction $\phi = 5.0\%$ during creep under different stresses σ_0 . Data points of $\dot{\gamma}$ at early times dominated by the instrument inertia are marked by open symbols.

3 Calculation of the dimensionless shear rate $\dot{\Gamma}(n)$ in the one-dimensional model

In our one-dimensional (1D) model for the creep deformation of colloidal gels, the macroscopic strain Γ at dimensionless time n is obtained as $\Gamma(n) = N_p(n)\Gamma_l/N$, where N_p denotes the number of plastically deformed mesoscopic units, N the total number of mesoscopic units in the system, and Γ_l the local strain of each plastically deformed mesoscopic unit. We assume $\Gamma_l = 1$ for all units such that Γ is equal to the fraction of yielded units. Therefore, $\Gamma = 1$ when all units are plastically deformed. We let each unit yield once at most.

Adopting the approach presented in refs. S2,S3, the dimensionless local yield stress Σ_y of each mesoscopic unit is sampled from a Weibull distribution, whose probability density function f_{wb} can be expressed as

$$f_{wb}(\Sigma_y) = \frac{\beta}{\Sigma_y^*} \left(\frac{\Sigma_y}{\Sigma_y^*} \right)^{\beta-1} \exp \left[- \left(\frac{\Sigma_y}{\Sigma_y^*} \right)^\beta \right], \quad \Sigma_y \in [0, \infty), \quad (\text{S2})$$

where β denotes the shape parameter and Σ_y^* the scale parameter. Numerical values of the local yield stress Σ_y are insignificant, and hence we assume a scale parameter $\Sigma_y^* = 1$. The larger the shape parameter β , the sharper f_{wb} is around $\Sigma_y = \Sigma_y^* = 1$, as shown in Fig. S3; the local yield stresses of different units are more narrowly distributed around $\Sigma_y^* = 1$ for larger β .

Upon the application of a constant dimensionless stress Σ_0 at $n = 0$, every unit with a local yield stress $\Sigma_y < \Sigma_0$ plastically deforms. Given a sufficiently large number of units N , the resultant initial macroscopic strain Γ_0 is the same as the fraction of the units with $\Sigma_y < \Sigma_0$, or $\Gamma_0 = 1 - \exp(-\Sigma_0^\beta)$, which is equal to the cumulative distribution function $F_{wb}(\Sigma_y)$ of the local yield stress evaluated at $\Sigma_y = \Sigma_0$.

For each unyielded unit, the probability of plastic deformation over a unit time $\Delta n = 1$ is assumed to be

$$p(\Sigma_y, \Sigma_0) = \nu \exp \left[- \frac{(\Sigma_y - \Sigma_0) V_a}{k_B T} \right], \quad (\text{S3})$$

where $\nu = 1$ denotes the attempt frequency, V_a the dimensionless activation volume, and $k_B T$ the dimensionless thermal energy. In the absence of spatiotemporal correlations, the probability of an arbitrary mesoscopic unit with $\Sigma_y > \Sigma_0$ being plastically deformed at time $n \geq 1$ is $p^1 (1 - p)^{n-1}$. Thus, the averaged probability

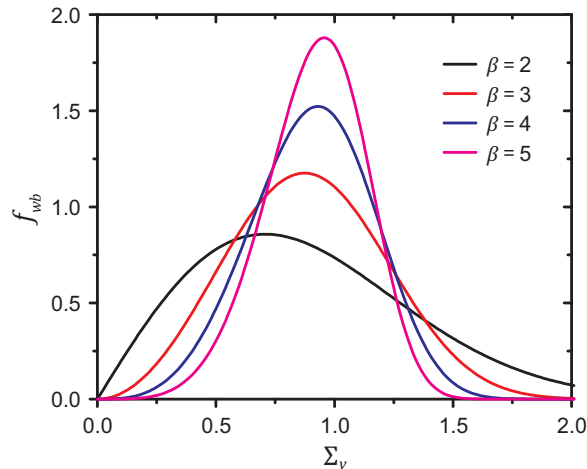


Fig. S3: Probability density functions of the Weibull distribution f_{wb} for a scale parameter $\Sigma_y^* = 1$ and shape parameters $\beta = 2, 3, 4$, and 5.

of local yield at $n \geq 1$ for a mesoscopic unit with $\Sigma_y > \Sigma_0$ can be expressed as

$$\begin{aligned}
P(n, \Sigma_0) &= \frac{\int_{\Sigma_0}^{\infty} p(\Sigma_y, \Sigma_0) [1 - p(\Sigma_y, \Sigma_0)]^{n-1} f_{wb}(\Sigma_y) d\Sigma_y}{\int_{\Sigma_0}^{\infty} f_{wb}(\Sigma_y) d\Sigma_y} \\
&= \frac{\int_{\Sigma_0}^{\infty} p(\Sigma_y, \Sigma_0) [1 - p(\Sigma_y, \Sigma_0)]^{n-1} f_{wb}(\Sigma_y) d\Sigma_y}{1 - F_{wb}(\Sigma_0)}. \tag{S4}
\end{aligned}$$

The average macroscopic shear rate at $n \geq 1$ is equal to the number of units with $\Sigma_y > \Sigma_0$ times the macroscopic strain increment due to a single plastic event, multiplied by $P(n, \Sigma_0)$:

$$\begin{aligned}
\dot{\Gamma}(n, \Sigma_0) &= N [1 - F_{wb}(\Sigma_0)] \times \frac{\Gamma_l}{N} \times P(n, \Sigma_0) \\
&= \int_{\Sigma_0}^{\infty} p(\Sigma_y, \Sigma_0) [1 - p(\Sigma_y, \Sigma_0)]^{n-1} f_{wb}(\Sigma_y) d\Sigma_y. \tag{S5}
\end{aligned}$$

Finally, the average total strain at $n \geq 1$ can be obtained from

$$\Gamma(n, \Sigma_0) = \Gamma_0(\Sigma_0) + \sum_{j=1}^n \dot{\Gamma}(j, \Sigma_0). \tag{S6}$$

4 Stress independence of power-law exponents for different model parameters

In the main text, the shear rate $\dot{\Gamma}$ in the model is shown to exhibit a power law $\dot{\Gamma} \sim n^{-0.6}$, independent of the applied stress Σ_0 when the strain falls within the range of $\Gamma = 0.01 - 0.04$ for a Weibull shape parameter $\beta = 3$ and a thermal energy density $k_B T/V_a = 0.06$. Altering the values of β or $k_B T/V_a$ changes the power-law exponent that best describes the temporal decrease in the shear rate $\dot{\Gamma}(n)$ in a given strain range. Yet, the stress independence of an exponent holds true regardless of the parameter values, as shown in Fig. S4(A and B).

A larger shape parameter β corresponds to a narrower distribution of the local yield stress Σ_y around the scale parameter $\Sigma_y^* = 1$, as displayed in Fig. S3, and thus leads to a reduced fraction of mesoscopic units whose local yield stresses are close to the applied stress $\Sigma_0 \ll 1$. The scarcity of readily deformable units renders plastic events less likely from the outset, partially obscuring the effect of statistical hardening. Therefore, a larger β results in a smaller magnitude of the power-law exponent in a fixed strain range (Fig. S4(A)). A smaller thermal energy density $k_B T/V_a$ strengthens the effect of statistical hardening by decreasing the probability of a plastic event for every unyielded unit after the initial deformation. A smaller $k_B T/V_a$ hence induces a larger magnitude of the power-law exponent in a fixed strain range (Fig. S4(B)).

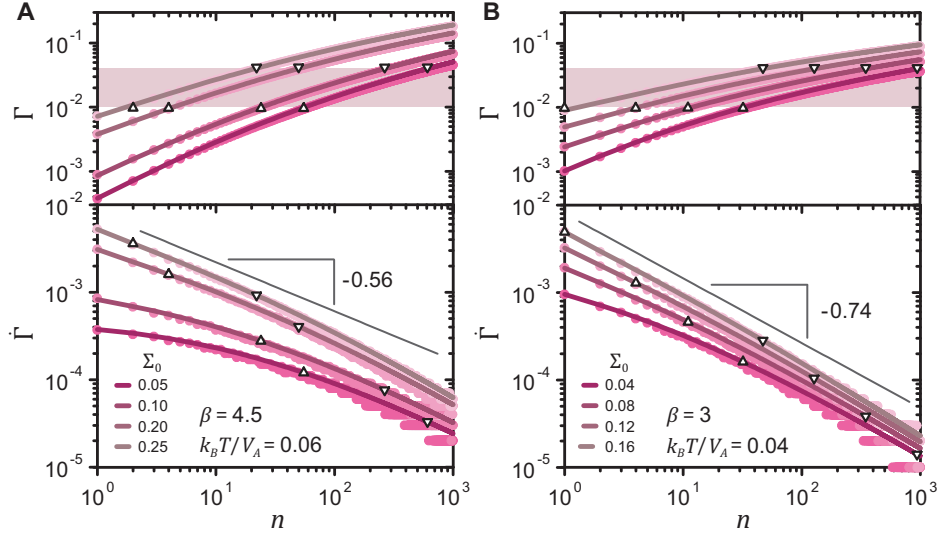


Fig. S4: Strain Γ and shear rate $\dot{\Gamma}$ of the 1D model (lines) and the 2D model (circles) under various stresses Σ_0 for different Weibull shape parameters β and thermal energy densities $k_B T/V_a$. Triangles and inverted triangles represent data points corresponding to the lower and the upper bounds, respectively, of the highlighted strain range $\Gamma = 0.01 - 0.04$. For the 1D model, the results are exact averaged quantities numerically evaluated by Eqs. (S5) and (S6). For the 2D model, the results are obtained from simulations for $N = 10^5$ and $M = 10^2$.

5 Model shear rate $\dot{\Gamma}(n)$ for different distributions

The same integral expression in Eq. (S5) can be used for other distributions than the Weibull distribution, except that $f_{wb}(\Sigma_y)$ needs to be replaced with the appropriate probability density function. Different distributions of the local yield stress lead to slightly different behaviors in the model in terms of the shear rate $\dot{\Gamma}(n)$.

5.1 Normal distribution

The normal (Gaussian) distribution gives rise to a near power-law creep with its exponent dependent on the strain, as observed for the Weibull distribution. The average shear rate $\dot{\Gamma}(n)$ in Eq. (S5) can be numerically calculated using the probability density function:

$$f_G(\Sigma_y) = \frac{1}{ZS\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{\Sigma_y - \mu}{S}\right)^2\right], \quad \Sigma_y \in [0, \infty), \quad (\text{S7})$$

where

$$Z \equiv \int_0^\infty f_G(\Sigma_y) d\Sigma_y \quad (\text{S8})$$

denotes the normalization parameter, μ the mean, and S the standard deviation. The normalization parameter is needed as we consider a distribution truncated at $\Sigma_y = 0$, such that f_G is defined only for $\Sigma_y \geq 0$. For the choice of $\mu = 1$ and $S = 0.3$, the shear rate decrease can be described as $\dot{\Gamma} \sim n^{-0.63}$ independent of the applied stress Σ_0 , when the strain Γ falls within the range 0.01 – 0.04, as shown in Fig. S5(A). The similarity between the normal and the Weibull distributions of the local yield threshold attests to the generality of the dependence of the power-law exponent on the total strain.

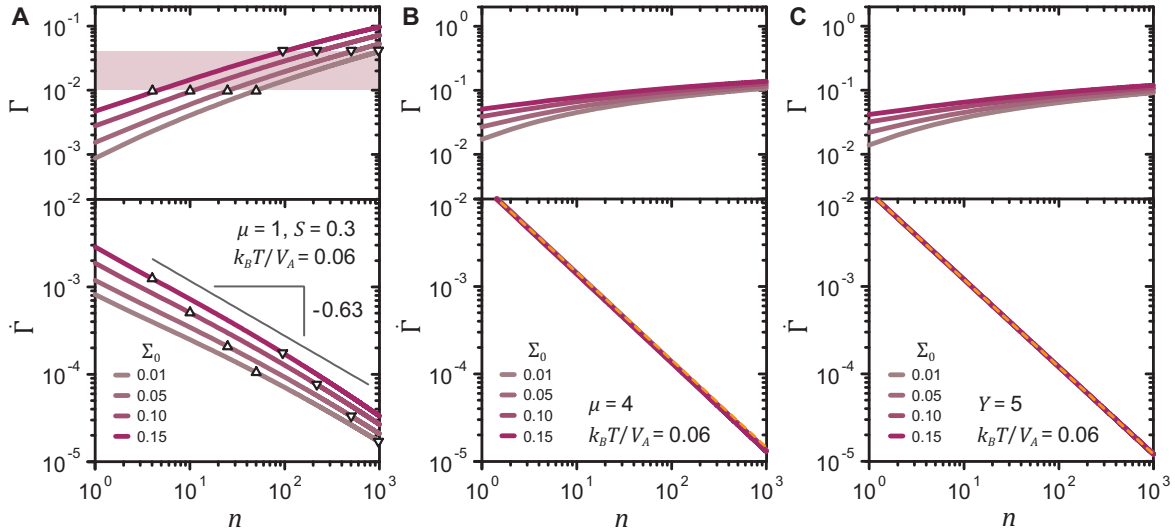


Fig. S5: Strain $\Gamma(n)$ and shear rate $\dot{\Gamma}(n)$ under different stresses Σ_0 for (A) the normal distribution, (B) the exponential distribution, and (C) the uniform distribution of local yield stress Σ_y . Triangles and inverted triangles in (A) represent data points corresponding to the lower and the upper bounds, respectively, of the highlighted strain range $\Gamma = 0.01 - 0.04$. Purple lines denote numerical results and orange dashed lines for $\dot{\Gamma}$ in (B and C) denote analytical results.

5.2 Exponential distribution

For an exponential distribution of the local yield stress, an analytical expression for $\dot{\Gamma}(n)$ can be obtained. The probability density function of an exponential distribution can be expressed as

$$f_{exp}(\Sigma_y) = \frac{1}{\mu} \exp\left(-\frac{\Sigma_y}{\mu}\right), \quad \Sigma_y \in [0, \infty). \quad (\text{S9})$$

We derive a closed-form expression for the shear rate by first applying the binomial theorem to the integrand in

$$\dot{\Gamma}(n, \Sigma_0) = \int_{\Sigma_0}^{\infty} p(1-p)^{n-1} f_{exp} d\Sigma_y, \quad (\text{S10})$$

where

$$\begin{aligned} p(1-p)^{n-1} &= p \left[\binom{n-1}{0} p^0 - \binom{n-1}{1} p^1 + \binom{n-1}{2} p^2 - \dots + (-1)^{n-1} \binom{n-1}{n-1} p^{n-1} \right] \\ &= \binom{n-1}{0} p - \binom{n-1}{1} p^2 + \binom{n-1}{2} p^3 - \dots + (-1)^{n-1} \binom{n-1}{n-1} p^n. \end{aligned} \quad (\text{S11})$$

Substituting $\xi \equiv k_B T / V_a$, such that

$$p = \exp\left[-\frac{\Sigma_y - \Sigma_0}{\xi}\right], \quad (\text{S12})$$

the integral can be calculated for each term in Eq. (S10) after the expansion. For the first term,

$$\begin{aligned} \int_{\Sigma_0}^{\infty} \binom{n-1}{0} p f_{exp} d\Sigma_y &= \binom{n-1}{0} \frac{1}{\mu} \exp\left(\frac{\Sigma_0}{\xi}\right) \int_{\Sigma_0}^{\infty} \exp\left[-\left(\frac{1}{\xi} + \frac{1}{\mu}\right)\Sigma_y\right] d\Sigma_y \\ &= \binom{n-1}{0} \frac{X_1}{\mu} \exp\left(\frac{\Sigma_0}{\xi}\right) \exp\left(-\frac{\Sigma_0}{X_1}\right) \\ &= \binom{n-1}{0} \frac{X_1}{\mu} \exp\left(-\frac{\Sigma_0}{\mu}\right), \end{aligned} \quad (\text{S13})$$

where

$$\frac{1}{X_q} \equiv \frac{q}{\xi} + \frac{1}{\mu}, \quad (\text{S14})$$

for a positive integer q . For the second term,

$$\begin{aligned} \int_{\Sigma_0}^{\infty} -\binom{n-1}{1} p^2 f_{exp} d\Sigma_y &= -\binom{n-1}{1} \frac{1}{\mu} \exp\left(\frac{2\Sigma_0}{\xi}\right) \int_{\Sigma_0}^{\infty} \exp\left[-\left(\frac{2}{\xi} + \frac{1}{\mu}\right)\Sigma_y\right] d\Sigma_y \\ &= -\binom{n-1}{1} \frac{X_2}{\mu} \exp\left(\frac{2\Sigma_0}{\xi}\right) \exp\left(-\frac{\Sigma_0}{X_2}\right) \\ &= -\binom{n-1}{1} \frac{X_2}{\mu} \exp\left(-\frac{\Sigma_0}{\mu}\right). \end{aligned} \quad (\text{S15})$$

Evaluating the integrals of all the terms yields

$$\dot{\Gamma}(n, \Sigma_0) = \frac{1}{\mu} \exp\left(-\frac{\Sigma_0}{\mu}\right) \left[\binom{n-1}{0} X_1 - \binom{n-1}{1} X_2 + \dots + (-1)^{n-1} \binom{n-1}{n-1} X_n \right]. \quad (\text{S16})$$

For $\mu q \gg \xi$, which holds true for relevant domains of the parameters that enable prolonged deformation,

$$X_q = \frac{\mu\xi}{\mu q + \xi} \simeq \frac{\mu\xi}{\mu q} = \frac{\xi}{q}. \quad (\text{S17})$$

The shear rate can be approximated as

$$\begin{aligned}\dot{\Gamma}(n, \Sigma_0) &\simeq \frac{\xi}{\mu} \exp\left(-\frac{\Sigma_0}{\mu}\right) \left[\binom{n-1}{0} \frac{1}{1} - \binom{n-1}{1} \frac{1}{2} + \dots + (-1)^{n-1} \binom{n-1}{n-1} \frac{1}{n} \right] \\ &\simeq \frac{\xi}{\mu} \exp\left(-\frac{\Sigma_0}{\mu}\right) \sum_{m=0}^{n-1} \frac{(-1)^m}{m+1} \binom{n-1}{m}.\end{aligned}\quad (\text{S18})$$

It can be shown, as in ref. S4, that

$$\sum_{m=0}^{n-1} \frac{(-1)^m}{m+1} \binom{n-1}{m} = \frac{1}{(n-1)+1} = n^{-1}, \quad (\text{S19})$$

which leads to

$$\dot{\Gamma}(n, \Sigma_0) \simeq \frac{k_B T}{V_a \mu} \exp\left(-\frac{\Sigma_0}{\mu}\right) n^{-1}. \quad (\text{S20})$$

This result indicates that an exponential distribution of the local yield stress in our model induces a logarithmic creep $\Gamma \sim \ln n$, as shown in Fig. S5(B), during which a single power-law exponent (-1) of $\dot{\Gamma}(n)$ satisfactorily describes the viscoplastic deformation for all accessible ranges of the strain $\Gamma(n)$.

5.3 Uniform distribution

An approximate analytical expression for $\dot{\Gamma}(n)$ can be obtained also for a uniform distribution of the local yield stress. The probability density function of a uniform distribution can be expressed as

$$f_{uni}(\Sigma_y) = \begin{cases} 1/Y & \text{if } 0 \leq \Sigma_y \leq Y \\ 0 & \text{otherwise} \end{cases} \quad (\text{S21})$$

where Y denotes the maximum local yield stress. Term-by-term integration of the integral

$$\dot{\Gamma}(n, \Sigma_0) = \int_{\Sigma_0}^{\infty} p(1-p)^{n-1} f_{uni} d\Sigma_y, \quad (\text{S22})$$

where $p = \exp[-(\Sigma_y - \Sigma_0)/\xi]$ and $\xi \equiv k_B T/V_a$, can be performed after the binomial expansion, as shown for the exponential distribution. The first term is

$$\begin{aligned}\int_{\Sigma_0}^{\infty} \binom{n-1}{0} p f_{uni} d\Sigma_y &= \binom{n-1}{0} \frac{1}{Y} \exp\left(\frac{\Sigma_0}{\xi}\right) \int_{\Sigma_0}^Y \exp\left(-\frac{\Sigma_y}{\xi}\right) d\Sigma_y \\ &= -\binom{n-1}{0} \frac{\xi}{Y} \exp\left(\frac{\Sigma_0}{\xi}\right) \left[\exp\left(-\frac{Y}{\xi}\right) - \exp\left(-\frac{\Sigma_0}{\xi}\right) \right] \\ &= -\frac{\xi}{Y} \exp\left(\frac{\Sigma_0}{\xi}\right) \exp\left(-\frac{Y}{\xi}\right) + \frac{\xi}{Y}.\end{aligned}\quad (\text{S23})$$

The second term is

$$\begin{aligned}\int_{\Sigma_0}^{\infty} -\binom{n-1}{1} p^2 f_{uni} d\Sigma_y &= -\binom{n-1}{1} \frac{1}{Y} \exp\left(\frac{2\Sigma_0}{\xi}\right) \int_{\Sigma_0}^Y \exp\left(-\frac{2\Sigma_y}{\xi}\right) d\Sigma_y \\ &= \frac{1}{2} \binom{n-1}{1} \frac{\xi}{Y} \exp\left(\frac{2\Sigma_0}{\xi}\right) \left[\exp\left(-\frac{2Y}{\xi}\right) - \exp\left(-\frac{2\Sigma_0}{\xi}\right) \right] \\ &= \frac{1}{2} \binom{n-1}{1} \frac{\xi}{Y} \exp\left(\frac{2\Sigma_0}{\xi}\right) \exp\left(-\frac{2Y}{\xi}\right) - \frac{1}{2} \binom{n-1}{1} \frac{\xi}{Y}.\end{aligned}\quad (\text{S24})$$

Evaluating all the terms and rearranging the resulting series leads to

$$\begin{aligned}
& \dot{\Gamma}(n, \Sigma_0) \\
&= \frac{\xi}{Y} \left[-\exp\left(-\frac{Y - \Sigma_0}{\xi}\right) + \frac{1}{2} \binom{n-1}{1} \exp\left(-\frac{2(Y - \Sigma_0)}{\xi}\right) - \dots + \frac{(-1)^n}{n} \binom{n-1}{n-1} \exp\left(-\frac{n(Y - \Sigma_0)}{\xi}\right) \right] \\
&\quad + \frac{\xi}{Y} \left[1 - \frac{1}{2} \binom{n-1}{1} + \frac{1}{3} \binom{n-1}{2} - \dots + \frac{(-1)^{n-1}}{n} \binom{n-1}{n-1} \right] \\
&= \frac{\xi}{Y} \left\{ \sum_{m=0}^{n-1} \left[\frac{(-1)^{m+1}}{m+1} \binom{n-1}{m} \exp\left(-\frac{(m+1)(Y - \Sigma_0)}{\xi}\right) \right] + \sum_{l=0}^{n-1} \frac{(-1)^l}{l+1} \binom{n-1}{l} \right\}. \tag{S25}
\end{aligned}$$

Given $Y \gg \Sigma_0$ and $\xi \ll 1$, which hold true for the relevant ranges of the parameters that enable prolonged deformation, the term in the second summation dominates its counterpart in the first summation for any $m = l \geq 1$. Thus, the expression simplifies to

$$\dot{\Gamma}(n, \Sigma_0) \simeq \frac{k_B T}{V_a Y} n^{-1}. \tag{S26}$$

This result indicates that a uniform distribution of the local yield stress, similar to an exponential distribution, leads to a logarithmic creep $\Gamma \sim \ln n$, as shown in Fig. S5(C), during which a single power-law exponent (-1) of $\dot{\Gamma}(n)$ satisfactorily describes the viscoplastic deformation for all accessible ranges of the strain $\Gamma(n)$.

6 Model elapsed time τ_p vs. stress Σ_0 for different distributions

As reported in the main text, the Weibull distribution of the local yield stress results in the exponential dependence of the elapsed time τ_p to reach an arbitrary value of strain Γ_p on the applied stress Σ_0 . We find that such exponential dependence likewise results from the normal, exponential, and uniform distributions.

6.1 Normal distribution

We numerically calculate the elapsed time τ_p for the model to reach an arbitrary strain Γ_p with a normal distribution of the local yield stress, for $\mu = 1$, $S = 0.3$, and $k_B T/V_a = 0.06$. As shown in Fig. S6(A), the elapsed time exhibits an exponential dependence $\tau_p \sim \exp(-\Sigma_0 V_a/k_B T)$, regardless of the value of Γ_p .

6.2 Exponential distribution

For the exponential distribution of the local yield stress Σ_y , the exponential dependence of the elapsed time τ_p on the stress Σ_0 can be derived analytically. Assuming τ_p is a positive integer, substituting $\Gamma = \Gamma_p$ into Eq. (S6) and rearranging the equation leads to

$$\sum_{j=1}^{\tau_p} \dot{\Gamma}(j, \Sigma_0) = \Gamma_p - \Gamma_0(\Sigma_0), \quad (\text{S27})$$

where the left-hand side can be expressed as

$$\begin{aligned} \sum_{j=1}^{\tau_p} \dot{\Gamma}(j, \Sigma_0) &= \sum_{j=1}^{\tau_p} \int_{\Sigma_0}^{\infty} p [1-p]^{j-1} f_{exp} d\Sigma_y \\ &= \int_{\Sigma_0}^{\infty} p f_{exp} \sum_{j=1}^{\tau_p} [(1-p)^{j-1}] d\Sigma_y. \end{aligned} \quad (\text{S28})$$

Simplifying the geometric series in the integrand,

$$\sum_{j=1}^{\tau_p} [(1-p)^{j-1}] = \frac{1 - (1-p)^{\tau_p}}{p}, \quad (\text{S29})$$

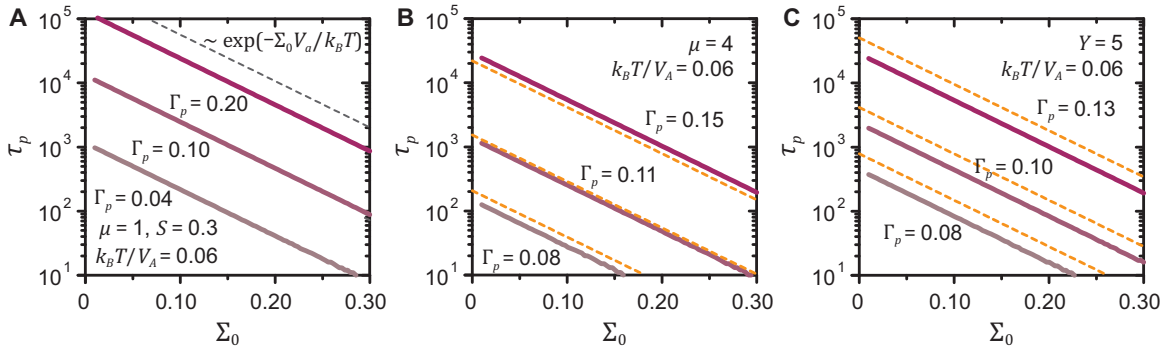


Fig. S6: Elapsed time τ_p for the model to reach arbitrary total strains Γ_p as a function of stress Σ_0 for (A) the normal distribution, (B) the exponential distribution, and (C) the uniform distribution of local yield stress Σ_y . Purple lines denote numerical results and orange dashed lines in (B and C) denote analytical results.

Eq. (S27) can be expressed as

$$\begin{aligned}
& \int_{\Sigma_0}^{\infty} [1 - (1-p)^{\tau_p}] f_{exp} d\Sigma_y = \Gamma_p - \Gamma_0(\Sigma_0) \\
& \Rightarrow \int_{\Sigma_0}^{\infty} f_{exp} d\Sigma_y - \int_{\Sigma_0}^{\infty} (1-p)^{\tau_p} f_{exp} d\Sigma_y = \Gamma_p - \Gamma_0(\Sigma_0) \\
& \Rightarrow \int_{\Sigma_0}^{\infty} (1-p)^{\tau_p} f_{exp} d\Sigma_y = 1 - \Gamma_p,
\end{aligned} \tag{S30}$$

since $\Gamma_0 = \int_0^{\Sigma_0} f d\Sigma_y$. Therefore, an expression for τ_p can be found by solving Eq. (S30). After the binomial expansion,

$$(1-p)^{\tau_p} = \binom{\tau_p}{0} p^0 - \binom{\tau_p}{1} p^1 + \binom{\tau_p}{2} p^2 - \dots + (-1)^{\tau_p} \binom{\tau_p}{\tau_p} p^{\tau_p}, \tag{S31}$$

the integral can be evaluated term-by-term. The first term is

$$\int_{\Sigma_0}^{\infty} f_{exp} d\Sigma_y = \exp\left(-\frac{\Sigma_0}{\mu}\right). \tag{S32}$$

The second term is

$$\begin{aligned}
\int_{\Sigma_0}^{\infty} -\binom{\tau_p}{1} p f_{exp} d\Sigma_y &= -\frac{1}{\mu} \binom{\tau_p}{1} \exp\left(\frac{\Sigma_0}{\xi}\right) \int_{\Sigma_0}^{\infty} \exp\left(-\frac{\Sigma_y}{X_1}\right) d\Sigma_y \\
&= -\frac{1}{\mu} \binom{\tau_p}{1} X_1 \exp\left(-\frac{\Sigma_0}{\mu}\right),
\end{aligned} \tag{S33}$$

where $\xi \equiv k_B T / V_a$ and $1/X_q \equiv q/\xi + 1/\mu$, for a positive integer q . The third term is

$$\int_{\Sigma_0}^{\infty} \binom{\tau_p}{2} p^2 f_{exp} d\Sigma_y = \frac{1}{\mu} \binom{\tau_p}{2} X_2 \exp\left(-\frac{\Sigma_0}{\mu}\right). \tag{S34}$$

The left-hand side of Eq. (S30) is equal to a series

$$\begin{aligned}
& \int_{\Sigma_0}^{\infty} (1-p)^{\tau_p} f_{exp} d\Sigma_y \\
&= \exp\left(-\frac{\Sigma_0}{\mu}\right) \left[1 - \frac{1}{\mu} \binom{\tau_p}{1} X_1 + \frac{1}{\mu} \binom{\tau_p}{2} X_2 - \dots + \frac{(-1)^{\tau_p}}{\mu} \binom{\tau_p}{\tau_p} X_{\tau_p} \right] \\
&\simeq \exp\left(-\frac{\Sigma_0}{\mu}\right) \left[1 + \frac{\xi}{\mu} \sum_{m=1}^{\tau_p} \frac{(-1)^m}{m} \binom{\tau_p}{m} \right],
\end{aligned} \tag{S35}$$

where the last line assumes that $X_q \simeq \xi/q$ as $q\mu \gg \xi$ for the relevant ranges of the parameters that enable prolonged deformation. Since the summation of the last term is the negated τ_p -th harmonic number,

$$\int_{\Sigma_0}^{\infty} (1-p)^{\tau_p} f_{exp} d\Sigma_y \simeq \exp\left(-\frac{\Sigma_0}{\mu}\right) \left[1 - \frac{\xi}{\mu} \sum_{j=1}^{\tau_p} \frac{1}{j} \right]. \tag{S36}$$

For sufficiently large τ_p ,

$$\sum_{j=1}^{\tau_p} \frac{1}{j} \approx \int_1^{\tau_p+1} \frac{dx}{x} = \ln(\tau_p + 1). \tag{S37}$$

Therefore, Eq. (S30) can be written as

$$1 - \frac{\xi}{\mu} \ln(\tau_p + 1) \simeq (1 - \Gamma_p) \exp\left(\frac{\Sigma_0}{\mu}\right). \quad (\text{S38})$$

Given $\Sigma_0 \ll \mu$, $\exp(\Sigma_0/\mu) \approx 1 + \Sigma_0/\mu$. Hence,

$$\begin{aligned} -\frac{\xi}{\mu} \ln(\tau_p + 1) &\simeq \frac{\Sigma_0}{\mu} - \Gamma_p - \frac{\Sigma_0}{\mu} \Gamma_p \\ \Rightarrow \ln(\tau_p + 1) &\simeq \frac{-\Sigma_0 + \mu\Gamma_p + \Sigma_0\Gamma_p}{\xi} \\ \Rightarrow \tau_p &\simeq \exp\left[\frac{(\mu + \Sigma_0)\Gamma_p}{\xi}\right] \exp\left(-\frac{\Sigma_0}{\xi}\right) \\ \Rightarrow \tau_p &\simeq \exp\left(\frac{\mu\Gamma_p V_a}{k_B T}\right) \exp\left(-\frac{\Sigma_0 V_a}{k_B T}\right), \end{aligned} \quad (\text{S39})$$

which indeed indicates that $\tau_p \sim \exp(-\Sigma_0 V_a/k_B T)$. The numerically evaluated and analytically approximated values of the elapsed time τ_p are in close agreement, and both exhibit an exponential dependence on Σ_0 , as shown in Fig. S6(B) for $\mu = 4$ and $k_B T/V_a = 0.06$.

6.3 Uniform distribution

An analytical approximation of the elapsed time τ_p can also be found for the uniform distribution of the local yield stress, using a similar approach as the one presented for the exponential distribution. We start from

$$\int_{\Sigma_0}^{\infty} (1-p)^{\tau_p} f_{uni} d\Sigma_y = 1 - \Gamma_p, \quad (\text{S40})$$

which is identical to Eq. (S30), except that f_{exp} is replaced with f_{uni} . The left-hand side can be evaluated by term-by-term calculation of the integrals after the binomial expansion. The first term is

$$\int_{\Sigma_0}^{\infty} f_{uni} d\Sigma_y = \frac{Y - \Sigma_0}{Y}. \quad (\text{S41})$$

The second term is

$$\begin{aligned} \int_{\Sigma_0}^{\infty} -\binom{\tau_p}{1} p f_{uni} d\Sigma_y &= -\binom{\tau_p}{1} \frac{1}{Y} \exp\left(\frac{\Sigma_0}{\xi}\right) \int_{\Sigma_0}^Y \exp\left(-\frac{\Sigma_y}{\xi}\right) d\Sigma_y \\ &= \binom{\tau_p}{1} \frac{\xi}{Y} \left[\exp\left(-\frac{Y - \Sigma_0}{\xi}\right) - 1 \right]. \end{aligned} \quad (\text{S42})$$

The third term is

$$\int_{\Sigma_0}^{\infty} \binom{\tau_p}{2} p^2 f_{uni} d\Sigma_y = -\binom{\tau_p}{2} \frac{1}{Y} \frac{\xi}{2} \left[\exp\left(-\frac{2(Y - \Sigma_0)}{\xi}\right) - 1 \right]. \quad (\text{S43})$$

By introducing a new variable $X \equiv Y - \Sigma_0$, the left-hand side of Eq. (S40) can be expressed as

$$\begin{aligned} &\int_{\Sigma_0}^{\infty} (1-p)^{\tau_p} f_{uni} d\Sigma_y \\ &= \frac{1}{Y} \left\{ X + \binom{\tau_p}{1} \frac{\xi}{1} \left[\exp\left(-\frac{X}{\xi}\right) - 1 \right] - \dots + (-1)^{\tau_p+1} \binom{\tau_p}{\tau_p} \frac{\xi}{\tau_p} \left[\exp\left(-\frac{\tau_p X}{\xi}\right) - 1 \right] \right\} \\ &= \frac{1}{Y} \left\{ X + \xi \left[\sum_{m=1}^{\tau_p} \frac{(-1)^{m+1}}{m} \binom{\tau_p}{m} \exp\left(-\frac{mX}{\xi}\right) + \sum_{l=1}^{\tau_p} \frac{(-1)^l}{l} \binom{\tau_p}{l} \right] \right\}. \end{aligned} \quad (\text{S44})$$

For $X \gg \xi$, which holds true for the relevant ranges of the parameters that enable prolonged deformation, the term in the second summation dominates the term in the first summation for any $m = l \geq 1$. Hence,

$$\begin{aligned}
\int_{\Sigma_0}^{\infty} (1-p)^{\tau_p} f_{uni} d\Sigma_y &\simeq \frac{1}{Y} \left[X + \xi \sum_{l=1}^{\tau_p} \frac{(-1)^l}{l} \binom{\tau_p}{l} \right] \\
&\simeq \frac{1}{Y} \left[X - \xi \sum_{j=1}^{\tau_p} \frac{1}{j} \right] \\
&\simeq \frac{1}{Y} [X - \xi \ln(\tau_p + 1)].
\end{aligned} \tag{S45}$$

Substituting this expression into Eq. (S40),

$$\begin{aligned}
X - \xi \ln(\tau_p + 1) &\simeq Y(1 - \Gamma_p) \\
\Rightarrow \xi \ln(\tau_p + 1) &\simeq (Y - \Sigma_0) - Y(1 - \Gamma_p) \\
\Rightarrow \ln(\tau_p + 1) &\simeq \frac{Y\Gamma_p - \Sigma_0}{\xi} \\
\Rightarrow \tau_p &\simeq \exp\left(\frac{Y\Gamma_p V_a}{k_B T}\right) \exp\left(-\frac{\Sigma_0 V_a}{k_B T}\right),
\end{aligned} \tag{S46}$$

which shows $\tau_p \sim \exp(-\Sigma_0 V_a / k_B T)$. This analytical approximation slightly overestimates the numerically calculated values of τ_p , as displayed in Fig. S6(C) for $Y = 5$ and $k_B T / V_a = 0.06$, but the exponential dependence of τ_p on Σ_0 is robust also in the numerical results for different arbitrary strains Γ_p .

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