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Rheology of animal and plant milks featuring dripping-onto-substrate protocols

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ABSTRACT

Animal milk is a multicomponent colloidal dispersion that contains fat globules and proteins dispersed in an aqueous continuous phase that contains dissolved sugars and minerals. The rapid increase in demand and production of plant milks, i.e., the plant-based alternatives to animal milks, is driven by lactose intolerance, allergens, and religious, cultural, environmental, and health choices or concerns. Many challenges for emulating animal milk's physicochemical, functional, and sensory attributes can be translated as problems related to the characterization and analysis of flow behavior, motivating this study. Only a few studies characterize shear rheology and almost none show characterization of extensional rheology of animal milks (cow and goat) and plant milks (soy, almond, rice, coconut, pea, and oat). We find that the two animal milks have rate-independent shear viscosity responses similar to Newtonian fluids. In contrast, the plant milks, except rice milk, display shear thinning. We characterize the pinching dynamics and extensional rheology response by employing the dripping-onto-substrate (DoS) protocols that we have shown are suitable for formulations such as milk that display a constant or shear thinning response but no measurable elasticity or viscoelasticity. In the last stage of pinching, plant milks appeared to show inertiocapillary, power law, or elastocapillary pinching, whereas animal milk displayed a viscocapillary response. The distinct rheology response of animal and plant milks, the illustration of suitable measurement protocols, and the discussion of the role of dispersed colloidal particles, drops, and macromolecules are presented as three key contributions of this study, aimed at facilitating the formulation and design of more nutritious and sustainable future milks.

INTRODUCTION

Animal milk refers to the nutrition-dense, calorie-rich fluid mammals produce to feed their growing offspring.[1] The color, texture, mouthfeel, creaminess, and rheology or flow behavior of animal milks depend mainly on the size, density, and concentration of the fat droplets and proteins or protein assemblies in the dispersed phase.[2] Nearly a billion tons of animal milk are consumed annually worldwide by humans as a food ingredient, savory beverage, raw material for making cheese, butter, buttermilk, milk-based desserts, and as a supplementary source of nine essential amino acids, minerals (calcium), and vitamins (D).[1–3] Milk is the source of many specialized regional food by-products like *ghee*, *lassi*, *chaas*, *paneer*, and *channa* in the Indian subcontinent and European cheeses granted protected geographical status like Havarti, Feta, Livarot, Roquefort, Piave, Taleggio, Boerenkaas, Oscypek, Queijo Serpa, Cabrales, Queso Manchego, and Syr.[1–3] Milk has a long history of use as a cosmetic ingredient, lotion, and skin softener, as an additive to many potions and formulations claimed to improve health or beauty, and as a vehicle for delivering nutrients, drugs, and minerals.[4,5] Lactose intolerance and cow milk allergies (CMA)[6,7] have ushered in demand for milk from animals like goats, sheep, camels, and yaks, or plant-based alternatives, especially as children risk malnutrition and stunted growth without adequate digestible, nutritious milk.[8–10] The recent surge in demand for plant-based substitutes or vegan alternatives is due to an increased awareness of the impact of dairy farming and agriculture on the environment, animal welfare, and climate change.[8,10,11] Additional push comes from consumers who cannot drink animal milk due to cultural, religious, or dietary restrictions due to allergens, diabetes, and cholesterol levels.[8,9] Despite the growing market for plant milks, significant challenges remain regarding flavor, mouthfeel, texture, foamability, shelf life, and consumer perception.[10–13] The critical need for formulating dairy milk alternatives without compromising on the nutritional, cultural, and functional attributes could be interpreted partially as a problem in calibrating and emulating the physiochemical properties like rheology of bovine milk.[12,13] Here, we contrast the flow behavior of two animal (cow and goat) and six plant (soy, pea, oat, coconut, almond, and rice) milks, by supplementing shear rheology characterization with a more challenging (and often neglected) analysis of pinching dynamics and extensional rheology response using dripping-onto-substrate (DoS) rheometry protocols.

Animal milks exhibit variations in the amount and types of proteins, fat, sugar, and other molecules due to the influence of the diet, habitat, lactation cycle, and species-specific physiological traits.[1,14] Bovine and goat milk have <4% fat, whereas whale and seal milk fat is >40%.[1] The fat fraction is highest in whole milk (3.3%), but the stores also carry reduced fat (2%), low fat (1%), and fat-free (skimmed) versions that are often homogenized and pasteurized. Homogenization reduces the drop sizes and narrows size distribution, enhancing shelf life and possibly influencing the state of different protein conformations and distributions in milk-fat-globule-membrane (MFGM), dispersed, or micellar states.[15–17] The fat globules in cow and goat milk contain a complex mixture of triglyceride and lipid molecules, which liquefy completely above 40 °C.[3,18–22] The emulsified large fat globules (0.1–15 μm) are enveloped by a protein-embedded milk-fat-globule-membrane (MFGM).[3,20–25] In addition to proteins in MFGM and some dispersed immunoglobulins (0.6–1 g/l), animal milk primarily contains two distinct classes of dispersed proteins: globular whey proteins (including lactoglobulins and lactalbumins) in monomeric form (<10 nm), and four types of flexible caseins as aggregates or self-assembled as micelles.[2,26,27] The casein micelles (∼50–500 nm) are like particles and contain calcium phosphate.[26–28] Bovine milk's suspending phase is an aqueous solution containing lactose and minerals. Greater than 80% of the world milk market is dominated by bovine milk. Goat milk is quite similar to bovine milk in terms of fat and protein concentration, shear viscosity and surface tension, and even nutritional and physiochemical properties, though lower lactose concentration and better digestibility are considered advantageous by its adherents.[29–31]

On the other hand, even though the demand, availability, and variety for plant milks have surged most in the last thirty years, there are attempts to reserve or preserve the term "milk" for dairy products. However, calling plant-based concoctions milk is a centuries-old tradition.[1,7,9,32] South/Southeast Asian and East Asian recipes have used coconut and soy milk for over two thousand years.[1,14,33,34] Cookbooks have suggested almond milk for over a thousand years in Central Asia and since the European Renaissance.[14,33] Fascinatingly, coconut, almond, and soy milk were used in cooking long before becoming favored milk substitutes for drinking. Samuel Johnson's dictionary (1755 and 1773 editions) offers "emulsion produced by contusion of seeds" as a secondary meaning for milk and quotes Francis Bacon for mentioning "nourishing almond milk."[35] Etymologically speaking, "emulsion" originated from the Latin verb *emulgere* (to milk out). It was first used to describe milk made by crushing almonds in water. Like almond milk, the recipes for making rice, pistachio, pea, soy, or oat milk, involve milling, grinding, and filtering processes that result in a dispersion of fine oil drops or globules, plus particles in an aqueous phase for suspending fluid. A survey of different recipes, ingredient lists, and scientific publications suggests that plant milks have displayed significant variations and diversity in size distributions, amount and type of oil drops, residual particles, extracted proteins, and polysaccharides.[7–9,12,13,32–36] The plant milk formulations are often enhanced for consumer satisfaction by fortifying with nutrients, adding flavor and aroma molecules, reducing fine particles, tuning the amount and type of sugars, changing pH and salt concentration, and adding emulsifier and rheology modifiers to influence stability, foamability, mouthfeel, and texture.[12,13,36]

We posit that the improved design of formulated milks will require understanding more than just the shear rheology response. In oral processing, dilution, digestion, and dispersion by mixing with saliva provide additional influence on perceived flow behavior, mouthfeel, or texture.[37–39] Industrial processing and bottling, eating, and appreciating milk introduce flow fields that are either extensional associated with streamwise velocity gradients, or tribological, confined within fast-moving solid surfaces.[38–41] Many plant milks are advertised using images highlighting pouring, drinking, and splashing that are free surface flows with extensional flow fields. The extensional rheology response likely influences the consumer's sensory perception and milk performance in processes like sucking through a baby bottle or a straw, pumping through a coffee nozzle or filters, splashing, pouring, spraying, and spray-drying.[41,42] The extensional viscosity that characterizes the resistance to extensional flows is three times the shear viscosity for Newtonian fluids. However, the extensional rheology response of many non-Newtonian fluids can display strong strain-rate- and strain-dependent values that cannot be computed or anticipated by measuring the shear rheology response.[41–55]

Many conventional and commercial methods, including CaBER (capillary breakup extensional rheometer), FISER (filament stretching extensional rheometer), microfluidic or capillary rheometer, orifice flow, and opposed jet, are unsuitable for characterizing extensional rheology responses of low-viscosity and low-elasticity fluids like milk.[41–50] Therefore, we characterize pinching dynamics and extensional rheology of milks using the dripping-onto-substrate (DoS) protocols we developed and have used for characterizing extensional relaxation time and apparent extensional viscosity of polymer solutions and various complex fluids (including low-viscosity systems) as detailed elsewhere.[41,48–55] The DoS protocols visualize and analyze capillarity-driven pinching of a fluid neck formed after a finite fluid volume is dispensed from a fixed nozzle onto a partially wetting or pinned substrate. Scaling analysis and detailed numerical studies have led to an understanding of how parameters captured by power law or exponential fits to radius evolution data provide the measurement of material properties, especially for polymer solutions.[53,55,56] We recently reviewed the literature on modeling drop deformation and rheology of dilute to dense emulsions.[57] Only a couple of studies have investigated capillarity-driven pinching dynamics of semi-dilute, concentrated, and jammed-dense emulsions.[58–60] However, there are no such investigations of dilute emulsions, in addition to the near absence of any extensional rheology studies of animal and plant milks.

In this contribution, we characterize the rheological response of the two animal milks to establish the baseline expectation for flow behavior and restrict attention to the standard American version of homogenized whole milks. After identifying rheological measures and methods that are most suitable for conducting a comprehensive analysis of the flow behavior of two animal milks (cow and goat), we utilized these to characterize and contrast the rheological response of six plant milks. We utilized torsional shear rheometry to characterize steady shear viscosity, which represents resistance to shear flows associated with velocity gradients perpendicular to flow direction, commonly arising when fluid moves past solid or liquid interfaces.[44,61] Surprisingly, reliable, steady shear viscosity measurements for plant milks are unavailable. On surveying the few published datasets that plot the shear viscosity of oat milk,[36,62–64] soy milk,[36,63] and coconut milk,[65] we find reports with relatively high shear viscosity (>100 × water viscosity) and strong apparent shear thinning at nominally low rates ($\sim 1\,\mathrm{s}^{-1}$). Even though polysaccharide thickeners could contribute to enhanced viscosity and shear thinning,[41,51,66] the lack of datasets perhaps attests to the shear rheology characterization challenges that arise for low-viscosity fluids. We characterize the extensional rheology response of animal and plant milks using DoS rheometry, by analyzing pinching dynamics using protocols and key equations, summarized in the Materials and Methods section. Most plant milks contain drops with coated oil–water interfaces, fine dispersed particles, and polysaccharides added to the suspending fluid. In contrast, animal milks contain large casein aggregates and fat globules surrounded with MFGM. Polymer coils and emulsion drops can exhibit much stronger deformation under strong, sustained extensional flows than shear flow.[41–50,57,67] Therefore, animal and plant milks can be expected to show distinct ingredient-dependent extensional rheology responses. Understanding flow behavior could provide a pathway to producing more nutritious alternatives with fewer ingredients and processing steps. However, there appear to be no experimental studies of the extensional rheology of animal or plant milks, and we take upon this challenge. The pinching dynamics and apparent extensional viscosity are measured for these milks for the first time. We hope the protocols and insights obtained will guide scientists and formulators who aim to create healthier, sustainable, nutritious, and texture-wise immaculate milk for the future.

MATERIALS AND METHODS

Two animal milks (cow and goat) and six plant milks (soy, pea, oat, coconut, almond, and rice)

Homogenized "whole" animal milks, and unflavored, "original" plant milks were procured and used to minimize the possible effects of added sugars, fats, or flavoring. Table I lists the lipids or oils, proteins, and polysaccharides in the milks and the amount of fat, sugars, and proteins present. The plant milks are formulated by relying on oil within the source grain or nuts and supplementing with additional oil if needed. Furthermore, additives such as polysaccharide gums are added as rheology modifiers, thickeners or stabilizers, and lecithin, saponin, and other surfactants, if present, improve the emulsification and foaming properties.[8,68] The physicochemical properties of the plant-based oils are comparable, with typical values for viscosity as 40–60 mPa·s, density ∼0.92 g/cc, surface tension 30–40 mN/m and refractive index (1.45–1.47).[69–71]

Mammalian milk primarily contains caseins, self-assembled as micelles, and globular whey proteins. Plant milk contains four primary classes of globular proteins, classified based on their solubility: globulins are soluble in salt solutions, albumins are water-soluble, prolamins dissolve in alcoholic solutions, and glutelins are soluble in dilute acidic or basic conditions. Proteins in each plant milk are listed in Table I, and their sedimentation coefficient s, the ratio of sedimentation velocity to acceleration, is measured in (S), the Svedberg unit (10^{-13} s). The sedimentation coefficient that equals the effective mass (protein or particle mass–mass of the displaced fluid $(m - m_0)$ divided by the shape- and size-dependent friction coefficient measures the hydrodynamic size and the propensity for settling or creaming, determining the shelf life. Saponins are found in soy and oat milk. Lecithin is present in soy milk but is sometimes added to plant milk to aid in emulsification and influence foamability and foam stability. Finally, the plant milks used in this study have gellan gum as the primary stabilizer and thickener. A relatively small concentration, often <0.1 wt. %, is usually sufficient to enhance the viscosity of the aqueous phase. Thus, increasing the overall viscous resistance to flow and deformation increases the timescale for diffusion, sedimentation, and coalescence, enhancing shelf life.

Torsional shear and dripping-onto-substrate (DoS) rheometry

Shear flow curves were obtained using a Couette cell (double gap, DG) on an Anton Paar MCR 302 rheometer. All samples were run at a fixed time setting of 10 s per point and a fixed temperature, 25 °C. The minimum viscosity and stress that are measurable using the DG geometry were considered before measuring steady shear viscosity and both ramp-up ($\dot{\gamma} = 0.01 - 1000\,\mathrm{s}^{-1}$) and ramp-down ($\dot{\gamma} = 1000 - 0.01\,\mathrm{s}^{-1}$) flow curves. The shear rheology characterization of milk involved several challenges often encountered in making accurate measurements for such low-viscosity fluids.[44] For example, proteins adsorbed at the liquid–air interface led to measurement artifacts like apparent shear thinning arising from the measurable interfacial viscosity, apparent interfacial yield stress,

TABLE I. Constituents and composition of animal and plant milks with polysaccharide additives and weight fractions listed based on labels on the commercial samples (grocery-store bought milks included whole for animal and unflavored original for the plant milks).

Milk	Fats, lipids, oils	Proteins and polysaccharides	Proteins (wt. %)	Fat (wt. %)	Sugars (wt. %)
Cow	Fat globules	Caseins and whey	3.33	3.33	5.00
Goat	Fat globules	Caseins and whey	3.33	2.92	4.58
Soy	Soybean oil	Glycinin (11S) and β-conglycinin (7S) *Gellan Gum*	3.33	1.88	2.08
Pea	Sunflower oil [a]Sunflower lecithin	Pea globulins: Legumin (11S), Vicilin (7S) and Convicilin (8S) *Gellan Gum and Guar Gum*	3.33	1.88	2.50
Oat	Canola/rapeseed oil	Oat globulins (12S); *Gellan Gum*	0.83	2.08	2.92
Coconut	Coconut oil [a]Sunflower lecithin	Coconut globulins: Cocosin (11S); *Gellan Gum*	0	2.08	2.08
Almond	Almond oil [a]Sunflower lecithin	Almond globulin: Amandin (14S); *Gellan Gum*	0.42	1.04	2.92
Rice	Sunflower, canola, safflower	Rice Glutelins	0	1.04	5.00

[a] Sunflower lecithin is added to improve emulsification and emulsion properties in some plant milks.

and viscoelasticity.[72,73] We verified that the apparent shear thinning observed at low shear rates for animal and plant milks was either untrustworthy as the measurement fell below the low torque limit of the rheometer transducer or displayed symptoms of the interfacial artifact, for switching to cone-and-plate geometry showed the expected upward shift in apparent viscosity plots, due to interfacial viscoelasticity and interfacial yield stress detailed in studies on aqueous solutions of bovine serum albumin and other proteins.[72–76]

The pinching dynamics and extensional rheology characterization were carried out using dripping-onto-substrate (DoS) protocols. A finite volume of liquid was pumped slowly using a NE-8000 syringe pump through a nozzle onto a partially wetting glass substrate to form an unstable bridge between a sessile drop and a liquid overhanging at the nozzle. The evolution of the liquid bridge's neck was visualized and captured with a high-speed camera (Fastcam SA3) at 19 000 fps. The nozzle radius ($2R_0 = 0.92$ mm), nozzle-to-substrate distance ($H/2R_0 = 2.25$), initial flow rate (0.02 ml/min), and temperature (20 °C) were the same for all trials. Image analysis was carried out using specially written codes using MATLAB (R2021b), and plots of normalized radius (R/R_0) as a function of time (t) were obtained and analyzed further. The quantitative radius evolution data, coupled with the image sequences that show apparent neck shape, provide insights into the extensional rheology response of each milk.

Summary of key equations and concepts used for analyzing pinching dynamics

The interplay of inertial, viscous, and capillary stresses for Newtonian fluids drives pinching flows in liquid necks spontaneously created during dripping, jetting, and spraying.[56,77–79] Similar interplay also arises in liquid necks formed after a step-strain stretches a liquid bridge between finger and thumb (a "digital" rheometer) or between two plates (in bespoke devices or commercialized techniques like CaBER and Trimaster).[38,80–84] Power laws of the form: $R(t)/R_0 = X_{xc}[(t_f - t)/t_{xc}]^{n_e}$ often capture the minimum radius vs time evolution for Newtonian fluids. Here, the prefactor X_{xc} is O(1) constant, t_{xc} a characteristic time, power-law exponent, n_e is 2/3 or 1, and t_f represents filament lifespan. The pinching dynamics of low-viscosity Newtonian fluids such as water (where $\mathrm{Oh} = \eta/\sqrt{\rho \Gamma R} < 1$) is governed by an interplay of inertial and capillary stresses, $\rho \dot{R}^2 \sim \sigma/R$, where ρ is fluid density and Γ the surface tension. The influence of this interplay manifests in the formation of a conical neck shape, with the radius evolution following the inertiocapillary (IC) pinching behavior,[56,77,79] described by a viscosity-independent power law:

$$\frac{R(t)}{R_o} = X_{IC}\left(\frac{\Gamma}{\rho R_o^3}\right)^{\frac{1}{3}}(t_f - t)^{\frac{2}{3}}, \tag{1}$$

where $n_e = 2/3$ and $t_{IC} = \left(\rho R_o^3/\Gamma\right)^{\frac{1}{2}}$. The pre-factor X_{IC} is O(1) but not a constant, but varies in the range $0.4 \leq X_{IC} \leq 0.8$ as detailed elsewhere.[77,85] A linear decay in the terminal pinching regime is characteristic of higher viscosity fluids such as glycerol, where $\mathrm{Oh} > 1$, for which viscous stresses exceed inertial stresses and their interplay with capillarity leads to the linear viscocapillary (VC) pinching behavior, described as follows:

$$\frac{R(t)}{R_o} = X_{VC}\frac{\Gamma}{\eta R_0}(t_f - t). \tag{2}$$

The interplay of capillarity and viscoelastic stresses results in elastocapillary (EC) pinching

$$\frac{R(t)}{R_o} \approx \frac{R_c}{R_o}\exp\left(-\frac{t - t_c}{3\lambda_E}\right). \tag{3}$$

Here, λ_E is the timescale that sets the pace of exponential decay and is considered as the extensional relaxation time, R_c is the radius at the onset of the EC regime, and t_c marks the transition into the EC regime. Models based on a corrected or modified Entov–Hinch theory determine that $R_c = R_0(G_E R_0/2\Gamma)^{\frac{1}{3}}$ assuming G_E represents an apparent extensional modulus. A limited few studies of pinching dynamics of power-law fluids that used CaBER or DoS show that radius evolution data exhibit $R(t) \propto (t_f - t)^{n_e}$ with the extensional exponent $n_e \neq n$, implying power laws can differ in shear and extensional rheology.[49,52,55,60,86] The radius evolution can be used to compute the transient extensional viscosity by considering flow to be driven by a tug-of-war between capillary stresses Γ/R and extensional stresses $\eta_E\dot{\varepsilon}$,

$$\eta_E = \frac{\Gamma}{\dot{\varepsilon}R} = \frac{-\Gamma}{2dR/dt}. \tag{4}$$

Capillary stress Γ/R continuously increases as neck radius decreases. In contrast, the variation in extensional rate depends on pinching dynamics, attaining a constant value for the EC regime, while Hencky strain $\varepsilon = -2\ln(R/R_0)$ continues to build up. Hence, capillarity-based measurements η_E is typically plotted as a function of ε.[56,82]

Particle size analysis and surface tension measurements using pendant drop tensiometry

The droplet size distribution of lipids found within each milk was determined by laser diffraction based on Mie light scattering theory with the use of a Horiba Partica mini-LA-350 (Horiba, Kyoto, Japan), which has a 650 nm laser light source giving the capability of detecting particle sizes within a wide range of 0.1–1000 µm. Duplicate measurements were performed for each milk sample. The dynamic surface tension was acquired for the eight milks using pendant drop tensiometry. The milk solution was pumped at 0.02 ml/min through a nozzle ($R = 0.92$ mm) into a container covered with parafilm and stopped once a stable pendant-shaped drop formed. A time-lapse was set to capture an image every minute for 60 min with a digital single-lens reflex (digital SLR or DSLR) camera. Surface tension was then calculated utilizing the drop shape analysis plug-in on ImageJ, assuming that the drop shape is determined by capillarity-gravity equilibrium.

RESULTS

Steady shear viscosity of animal and plant milks

The variation in steady-shear viscosity as a function of shear rate was characterized at 25 °C for animal and plant milks using a torsional rheometer as shown in Fig. 1. The two animal milks—cow and goat—exhibit nearly constant viscosity, $\eta = 1.8$ mPa·s and $\eta = 1.7$ mPa·s, respectively, as shown in Fig. 1(a). These are roughly twice that of water viscosity, and the flow behavior appears Newtonian with rate-independent viscosity. The overall emulsion viscosity value appears sensible since fat globules and casein micelles (plus whey proteins) are at a reasonably low volume fraction, and the suspending fluid is an aqueous solution that contains only lactose and minerals. No discernible normal stress differences are measurable, and any oscillatory shear

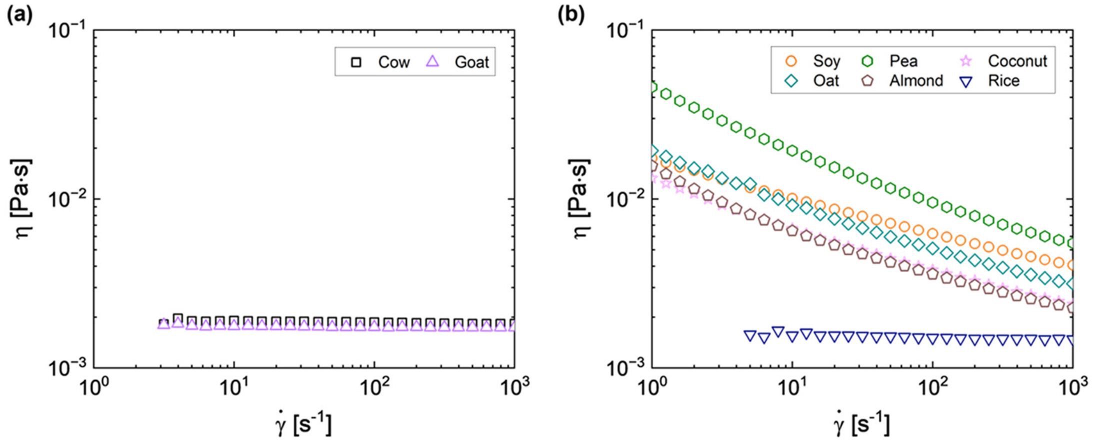


FIG. 1. Steady shear viscosity of animal and plant milks. (a) Cow and goat milks show rate-independent viscosity, similar to Newtonian fluids. (b) Plant milks show a shear thinning response, with rice milk the only exception, showing a constant value. The flow curves for these milk samples were acquired at 25 °C using a torsional (Anton Paar MCR 302) rheometer with double gap (DG) Couette geometry. The data are shown only for shear rates from $\dot{\gamma} = 1$–$1000\ \mathrm{s}^{-1}$, as the minimum torque limitation and the measurement artifacts induced by interfacial rheology (interfacial elastoviscoplasticity) contribution are at play at lower deformation rates.

measurements on this torsional rheometer are unlikely to detect any hint of elasticity for such low-viscosity systems. Figure 1(b) illustrates the flow behavior observed for all six plant milks. Only rice milk displays a single value of $\eta = 1.5$ mPas and emulates the rate-independent response of animal milk. Except rice milk, most of the plant milks are highly shear thinning, potentially due to the role played by the polysaccharide additives.

Pinching dynamics of cow and goat milks

We used the dripping-onto-substrate (DoS) protocols to visualize and analyze the capillarity-driven pinching (or thinning and breakup) of the liquid necks of the two animal milks. The image sequences obtained from high-speed videos are shown in Figs. 2(a) and 2(b) for cow and goat milk, respectively. Despite the animal milks exhibiting rate-independent shear viscosities only 1–2 times that of water ($\mathrm{Oh} \approx 0.01$), they do not form the conical neck shape expected for water-like fluids (upper cone with an acute angle, and lower with obtuse angle). In the last step, a relatively slender and thin liquid thread connecting two conical drops appears, suggesting a crossover or transition in pinching dynamics occurred in the final 2 ms. The quantitative evolution of the minimum neck radius extracted from the videos, $R(t)$, is shown in Fig. 2(c) for several representative trials. The filament lifespan, t_f, fell within the same range of 15–20 ms for all trials. Radius evolution plots show a transition in pinching dynamics consistently occurs at a critical neck radius of $R_t \approx 180\ \mu\mathrm{m}$ ($R_t/R_0 \approx 0.2$).

Like the image shapes, the radius evolution exhibits the 2/3 power-law exponent consistent with the inertiocapillary (IC) scaling in the early stages of pinching. A clear transition into linear decay is observed in the later stages, yielding remarkably similar slopes across multiple trials of cow and goat milk (≈ 0.17 m/s). The extensional strain rate, $\dot{\varepsilon} = -2\dot{R}/R$, can be computed with a numerical derivative of the data and is plotted as a function of $(t - t_f)$ in Fig. 2(d), where a rapid increase from $\dot{\varepsilon} \approx 200\ \mathrm{s}^{-1}$ to $\dot{\varepsilon} > 10\,000\ \mathrm{s}^{-1}$ is observed in the last 2 ms before the breakup. Figure 2(e) plots R/R_0 against time to breakup, $(t_f - t)$, on a log–log scale. The IC and VC regimes, with corresponding power laws of 2/3 and 1, are identified. An IC-VC transition can occur during capillary thinning in Newtonian fluids, as the local Ohnesorge number in the liquid neck continually increases due to its dependence on an external length scale (neck radius).[77,78] However, the Ohnesorge length, $l_{\mathrm{Oh}} = \eta^2/\rho\sigma$, which theoretically would mark this transition,[77–79] has a value of $l_{\mathrm{Oh}} \approx 80$ nm for the animal milks, which is three orders of magnitude lower than the observed transition point of $R \approx 180\ \mu\mathrm{m}$. Furthermore, **Eq.** (2) returns an overestimate for shear viscosity when used to fit the final pinching regime, yielding $\eta \approx 23$ mPa s for both milks instead of the observed $\eta = 1.7$–1.8 mPa·s. Thus, Newtonian viscous stresses alone cannot account for the emergence of this terminal regime, implying that other factors contribute, such as drop deformation and microstructural changes at high extension rates. Therefore, to distinguish it from the viscocapillary regime observed for Newtonian fluids, we refer to it as the terminal viscoelastocapillary (TVEC) regime.

Pinching dynamics and extensional rheology response of plant milks

Image sequences in Figs. 3(a) and 3(b) obtained using high-speed imaging and DoS protocols show the formation of a pronounced, slender, cylindrical neck for soy and oat milk, with shape evolution distinct from observation for the two animal milks, included in Figs. 2(a) and 2(b). The steady shear viscosity of both plant milks is larger than animal milks at the lowest shear rate measured (at $1\ \mathrm{s}^{-1}$, see Fig. 1) over a magnitude. However, the overall filament lifespan for the oat and soy milks is comparable to animal milks. The analysis radius evolution data in Fig. 3(c) reveals that soy and oat milk have distinct pinching dynamics. Oat milk displays a power-law response with $n_e \approx 0.76$, close to the power-law index of $n = 0.72$, observed in the shear rheology response.

In contrast, soy milk shows a short-lived power-law response followed by an elastocapillary (EC) pinching associated with the weak exponential decay observed in a linear-linear plot in Fig. 3(c). The EC regime appears as a strikingly linear region in the log-linear plot shown in the inset. The extensional rate attains a constant value of

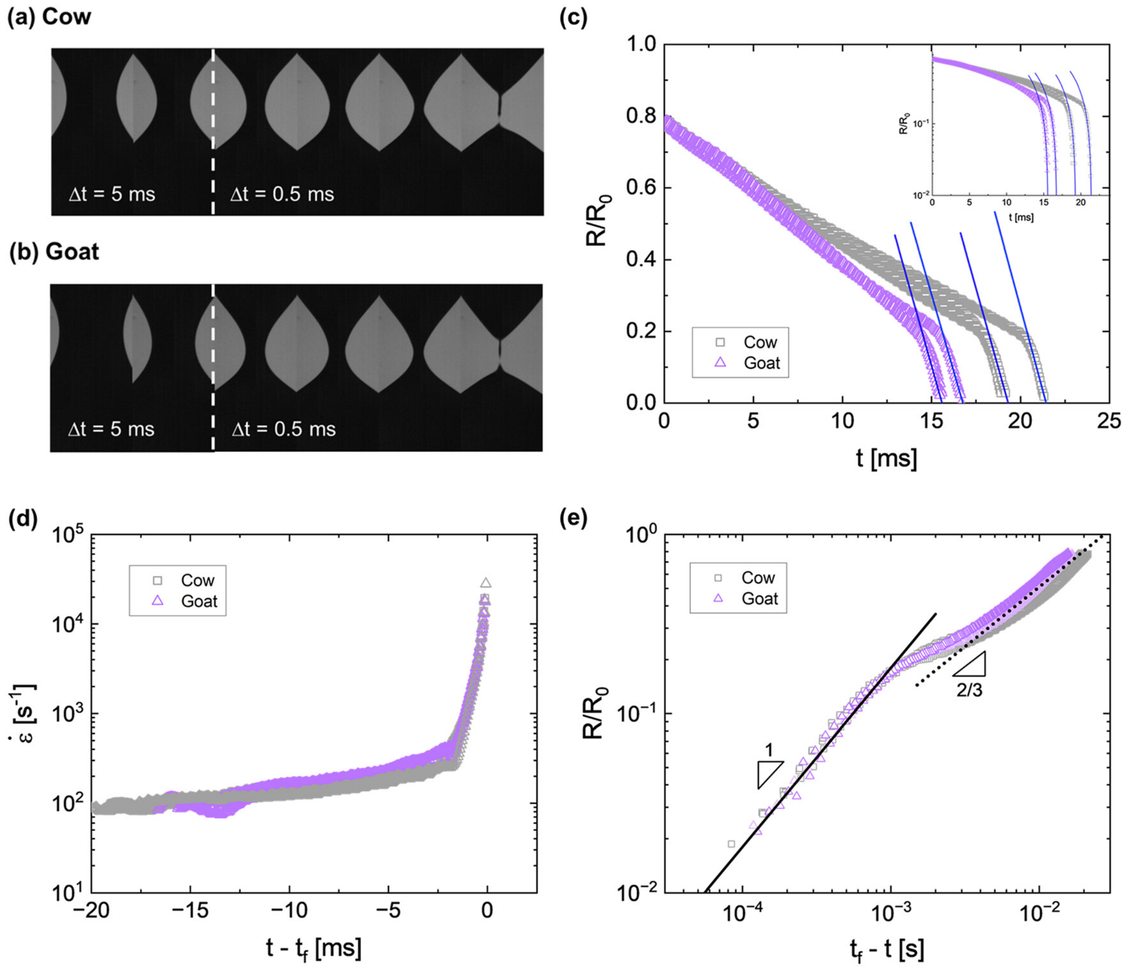


FIG. 2. Pinching dynamics of animal milks. (a) and (b) Image sequences extracted from the high-speed videos acquired at 19 000 fps were obtained using the dripping-onto-substrate (DoS) protocols for bovine and goat milk. The first two snapshots are 5 ms apart, and the next four are 0.5 ms apart. (c) Radius evolution data for bovine and goat milk, with several representative trials plotted on a linear and semi-log scale (inset). A crossover or transition in the dynamics occurs at $R/R_0 \approx 0.2$. (d) Extension rates in the thinning liquid necks are plotted as a function of $(t - t_f)$, where t_f is the filament lifespan. A sharp spike in $\dot{\varepsilon}$ is observed in the last 2 ms of the pinch-off process. (e) The normalized neck radius is plotted as a function of the time remaining until pinch-off on a log–log scale. The final pinching regimes for both milks exhibit a power law with $n_e = 1$, whereas the initial regime shows $n_e = 2/3$.

$\dot{\varepsilon}_{EC} = 2/3\lambda_E$ in the EC regime, as shown in Fig. 3(d) for the soy data. In contrast with oat milk that displayed dominant power-law behavior in the final stage before pinching, the log–log plot of radius vs time to pinch-off in Fig. 3(e) for soy milk shows the exponential decay due to elastocapillary pinching.

Thereafter, the pinching dynamics studies were conducted on the remaining four plant milks, and the results compiled in Fig. 4 are striking. The radius evolution datasets in Fig. 4(a) for almond and rice milk exhibited inertiocapillary (IC) responses, marked by a $R \propto (t_f - t)^{2/3}$ scaling in the radius evolution, and short filament lifespans ($t_f < 15$ ms). The corresponding image sequences show conical neck shapes (notice the acute and obtuse angles in the two cones) in Figs. 4(b) and 4(c), in contrast with the slender cylindrical neck observed for oat milk. Almond, and rice milk thus appear to have one pinching regime only, with power-law exponents and neck shape indicating inertiocapillary pinching and pinching dynamics strikingly distinct from the animal milks. While the IC behavior can be anticipated for low and constant viscosity rice milk, it is surprising to observe for the highly shear thinning almond milk.

Like soy, two more plant milks, pea and coconut, that display shear thinning exhibit delayed breakup, displaying a longer filament lifespan than expected for a Newtonian fluid of similar viscosity. Like soy, pea and coconut also exhibit an EC regime as shown in Fig. 4(d) and both milks form uniform cylindrical threads before pinch-off, as shown by images sequences included as Figs. 4(e) and 4(f). This is typical for polymer solutions, where the stress contributed by stretched polymer chains resists capillarity-driven pinching at high extension rates, or when the Weissenberg number $\mathrm{Wi} = \lambda_E\dot{\varepsilon} > 1/2$. Extensional relaxation times of $\lambda_E = 0.3$–0.4 ms are obtained for plant milks by fitting the radius evolution with the elastocapillary pinching expression. These timescales are comparable to those seen in dilute/semidilute solutions of cellulose-derivative thickeners such as hydroxyethyl cellulose

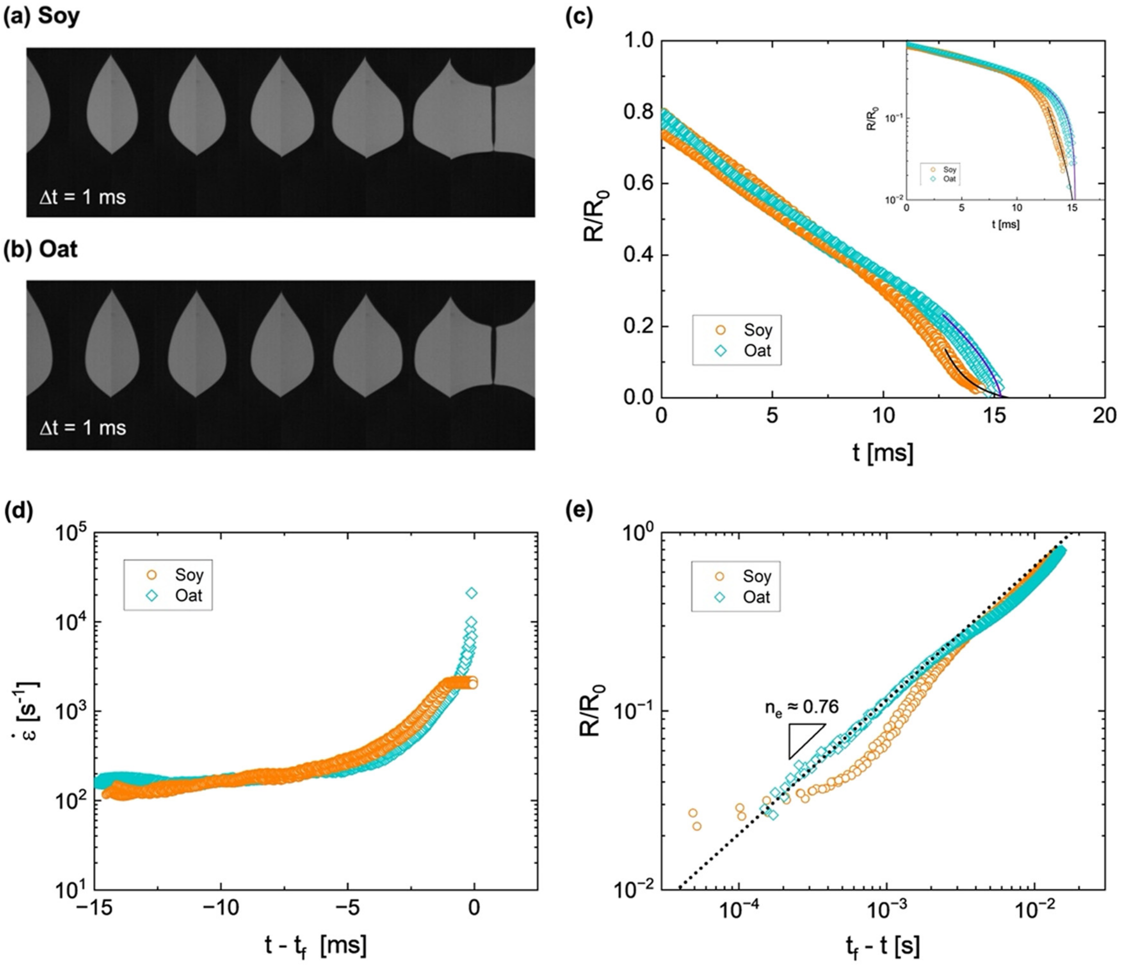


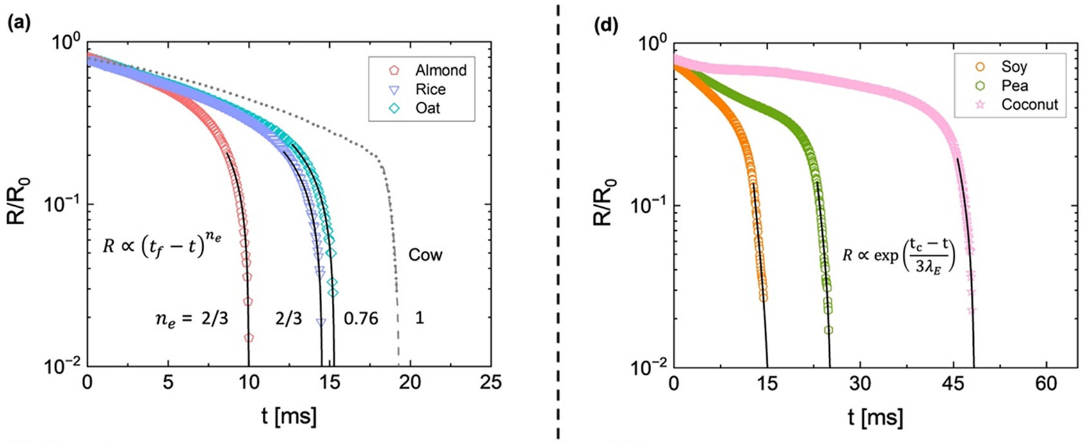
FIG. 3. Pinching dynamics of soy and oat milk. (a) and (b) Image sequences obtained using DoS protocols show slender cylindrical neck forms for soy and oat milk. All snapshots are 1 ms apart. (c) Radius evolution data for soy and oat milk, with several representative trials plotted on a linear scale (and in the inset, on a semi-log scale). (d) The extensional rate, $\dot{\varepsilon}$, in the thinning liquid necks plotted as a function of time to pinch-off $(t - t_f)$, where t_f is the filament lifespan. A constant $\dot{\varepsilon}$ regime observed for soy milk indicates an elastocapillary (EC) response. (e) Scaled radius R/R_0 plotted against the time to pinch-off on a log–log scale, illustrates that the power-law response for oat milk is similar in the initial and terminal regime. In contrast, the soy milk starts as a power-law response and then displays an unmistakable EC regime in its terminal stage.

(HEC) and sodium carboxymethylcellulose (NaCMC), and the radius evolution profiles are similar to semi-flexible polysaccharides, but distinct from solutions of flexible polymers.[49,53,87,88] Following the protocols developed for the polysaccharide solutions and formulations, we fit the radius evolution data for the viscoelastic plant milks using the Anna-Mckinley expression that incorporates an exponential decay for EC regime and linear decrease in radius associated with the terminal viscoelastocapillary (TVEC) regime governing the pinching dynamics for highly stretched polymers accounting for finite extensibility effects.

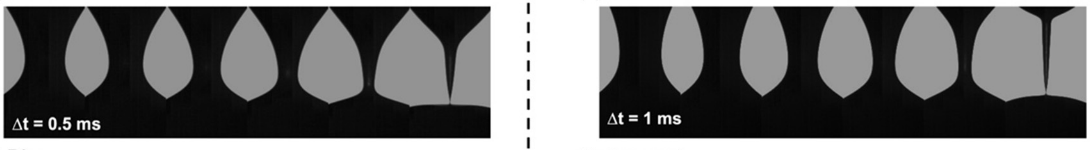
DISCUSSION

Non-Newtonian shear rheology of plant-based milks

The steady shear viscosity data for all plant milks except rice milk exhibited shear thinning behavior that can be fit using the empirical power-law model, $\Sigma_{12} = K\dot{\gamma}^n$ or alternatively $\eta = K\dot{\gamma}^{n-1}$. The model captures the shear stress or viscosity vs shear rate relationship using two parameters: an exponent called power-law index, n and a prefactor, called consistency, K. The K and n values obtained for all milks are included in Table II. All plant milks included in this study, except rice, contain gellan gum, a polysaccharide thickener or rheology modifier. Thus, even though the emulsions have similar low-volume fractions of dispersed fat globules or oil drops, cow, goat, and rice milk display no rate-dependence, whereas most of the plant milks are highly shear thinning, potentially due to the role played by the polysaccharide additives. A shear-thinning response is often considered more beneficial for designing formulations in the food, cosmetics, paints, personal care, and pharmaceutical industries.[44,52,60] Following the formulation industry practice, we list viscosity values at a low and a high shear rate (1 and 1000 s^{-1}). Higher



(b) Almond **(e) Pea**



(c) Rice **(f) Coconut**

FIG. 4. Plant milks display inertiocapillary (IC), power law (PL), and elastocapillary (EC) pinching. (a) Minimum neck radius evolution for almond and rice was obtained using DoS protocols, fitted with the IC scaling law characteristic of water-like fluids. Radius evolution for bovine milk is shown for contrast, and oat milk that displays a PL pinching is also included. (b) and (c) Image montages corresponding to the datasets in (a) display the conical neck shapes anticipated usually for low viscosity fluids like water. (d) Minimum neck radius evolution for pea and coconut milk, fit using a semiempirical expression from Anna and McKinley[81] which describes the EC and TVEC pinching behavior (and includes linear term for TVEC regime, in addition to the exponential decay expression included in the plot). (e) and (f) Image montages corresponding to the datasets in (d), displaying the cylindrical neck shapes expected of viscoelastic fluids.

viscosity at a low shear rate is conducive to limiting creaming, aggregation, or coalescence of the dispersed droplets (or particles) during storage and thus enhances emulsion or formulation stability and shelf life. A significant decrease in viscosity at higher rates is advantageous for typical processing operations and daily use, such as pumping, pouring, dispensing, and spreading.

Pinching dynamics and extensional viscosity of animal and plant milks

The DoS rheometry protocols facilitated the analysis of pinching dynamics and extensional rheology despite the relatively low viscosity of animal and plant milks. Such measurements would not be possible using conventional CaBER (capillary breakup extensional rheometer)

TABLE II. Measured values of shear viscosity at a nominal low and a high shear rate, consistency, and power-law exponent, surface tension, mean dispersed drop/particle size, and terminal pinching behavior of animal and plant milks.

Milks (25 °C)	η_1(mPa·s)	η_{1000}(mPa·s)	K (mPa·s^n)	n(-)	Γ(mN m^{-1})	$d_{3,2}$(μm)	Pinching behavior
Cow	1.83	1.83	1.83	1	47.1	1.00	IC to VC
Goat	1.74	1.74	1.74	1	47.6	0.66	IC to VC
Soy	17.4	4.06	17	0.78	46.1	0.63	PL to EC-TVEC
Pea	46	5.48	44	0.66	43.0	0.64	PL to EC-TVEC
Oat	19.3	3.16	18.7	0.72	38.9	1.18	PL to PL
Coconut	13.3	2.39	13	0.74	45.1	1.10	PL to EC-TVEC
Almond	15.7	2.27	14	0.70	48.5	0.86	IC
Rice	1.52	1.52	1.52	1	X	0.82	IC

protocols, which rely on step strain to create a stretched liquid bridge.[56,82] As the liquid bridge breaks before the step strain is completed (in 40–50 ms), CaBER is unsuitable for $\eta < 50\,\mathrm{mPa\cdot s}$ or $\lambda < 1$ ms unless slow retraction mode is employed.[56,82] Numerical and computational studies for generalized Newtonian fluids, including those governed by power law $\eta = K\dot{\gamma}^{n-1}$ prescribe a power-law response in radius evolution data with the same index.[56,89–91] Even though the plant milks (except rice) exhibited a power-law shear rheology response (see Table II), power-law pinching with a similar exponent capturing the radius evolution data is observed only for oat milk. In contrast, almond and rice milk exhibited IC response in neck shape and R vs t data, as summarized in Table II. A limited few studies of pinching dynamics of power-law fluids that used CaBER or DoS show that radius evolution exhibits $R(t) \propto (t_f - t)^{n_e}$ with the extensional exponent $n_e \neq n$ often distinct for polymeric complex fluids.[49,52,55,60,86] Observation of a slender neck for oat milk is consistent with previous experimental and computational studies that study the pinching of power-law fluids with $n > 0.6$. Table II shows that all the milks have comparable surface tension values and comparable mean drop/particle sizes. Previous studies report a power-law response in both shear and extensional flow for solutions and formulations with polysaccharide rheology modifiers.[49,52,53,55,56,86,90,91] Here, all plant milks except rice contain polysaccharides and all display shear thinning. However, even though the pinching dynamics response shows an initial power-law regime for all plant milks, three kinds of milk (soy, pea and coconut) exhibit EC response, and two (rice and almond) only the IC pinching regime.

Oat milk displays apparent extensional viscosity that decreases with increased strain, as shown in Fig. 5(a). The extensional rate in pinching necks for oat milk [plotted in Fig. 3(c)] can exceed $1000\,\mathrm{s}^{-1}$, but as η_E values also decrease with increased strain rate, implying oat milk displays extensional thinning behavior. Almond and rice display IC response, where the power law of 2/3 is associated with interplay of inertia and capillarity, and the viscous stresses are considered negligible. In contrast with almond, rice, and oat, three plant milks—soy, pea, and coconut—showed elastocapillary pinching, a characteristic of viscoelastic flow behavior. The apparent extensional viscosity vs Hencky strain plots in Fig. 5(a) for the three plant milks (soy, pea, and coconut) with EC pinching show strain hardening. Their apparent Tr ratio values are at least an order of magnitude higher than those observed for cow and goat milk. The EC regime in the radius evolution plot [see Fig. 4(b)] culminating in TVEC regime, and strain hardening in the apparent extensional viscosity vs strain plot [see Fig. 5(a)] are strong evidence for the influence of the polysaccharide rheology modifier. Similar neck shapes and EC-TVEC behavior in the terminal regime arises for aqueous solutions and formulations containing polysaccharides like cellulose gum, guar gum, hydroxyethyl cellulose (HEC), and ethyl hydroxyethyl cellulose (EHEC), among others.[41,46,49,51,88,92] For comparable solvent quality and M_w, polysaccharides have a weaker influence on pinching dynamics in dilute solutions than flexible polymers like poly(ethylene oxide) (PEO) or poly (acrylamide) (PAM) due to a trifecta of effects: lower flexibility (longer Kuhn length, fewer Kuhn segments), lower extensibility (ratio of fully stretched to unperturbed coil size), and higher segmental dissymmetry (ratio of Kuhn length to packing length).[88] However, polysaccharides entangled at lower concentrations for semi-dilute solutions, displaying power-law shear thinning response and PL followed by EC-TVEC in pinching dynamics.[49,53,87,88,93] The formation of complexes between polysaccharides and amphiphilic molecules or macromolecules (proteins) can also result in extensional thinning or thickening, depending upon the energetics or kinetics of association–disassociation.[47,53,54,94,95]

The animal milks display a transition from IC ($n_e = 2/3$) to TVEC ($n_e = 1$) beyond a critical extensional rate. For drops in emulsions, the dimensionless measure of the deformation rate, Capillary number, *Ca* is defined as the ratio of shape relaxation time, and (τ_σ) to the deformation time (τ_d) (inverse of deformation rate). If the suspending fluid viscosity (μ) exceeds drop viscosity ($\lambda\mu$) or the viscosity

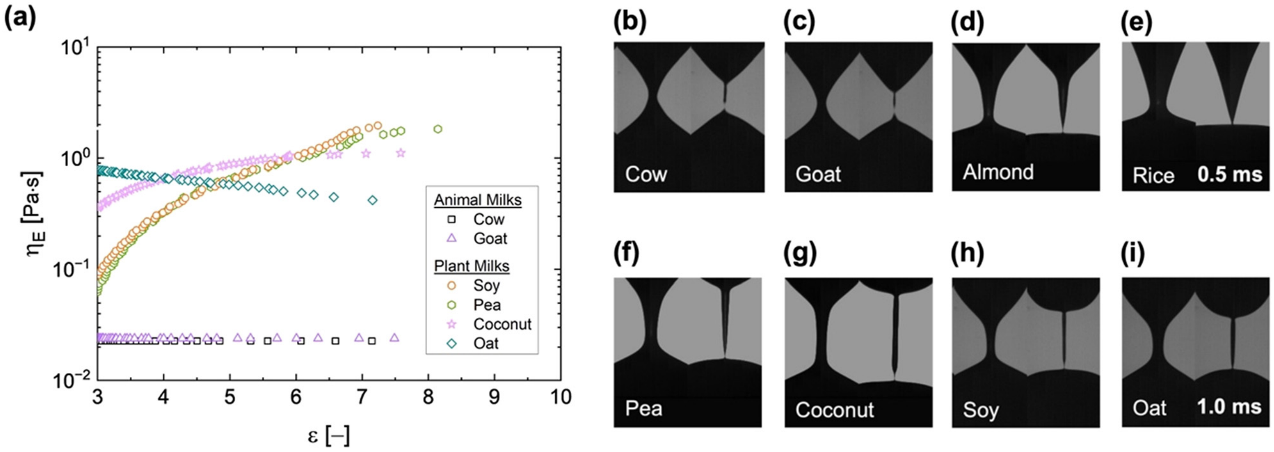


FIG. 5. Extensional viscosity and neck shapes of animal and plant milk. (a) Transient extensional viscosity plotted as a function of dimensionless strain for four plant milk—soy, pea, oat and coconut—contrasted with the terminal, steady extensional viscosity of the two animal milk (cow and goat). The radius evolution data extracted from the DoS protocols is used for computing extensional viscosity (equals the ratio of capillary stress and extensional rate) and Hencky strain $\varepsilon = -2\ln R/R_0$. The animal milk exhibits a terminal viscoelastocapillary pinching response, yielding a constant η_E significantly smaller magnitude than η_E for the three plant milks (soy, pea, and coconut), which exhibit strain-hardening and elastocapillary response, and oat milk which displays extensional thinning. (b)–(i) Two snapshots each are shown for eight milks to highlight the differences in pinching dynamics and material response. The first row shows images separated by $\Delta t = 0.5$ ms and the second row shows $\Delta t = 1.0$ ms.

ratio, $\lambda \leq O(1)$, shape relaxation time is computed as $\tau_\sigma = \mu a/\sigma_{eq}$, where a is the average droplet size and σ_{eq} is the equilibrium interfacial tension, giving the following expression:

$$Ca = \frac{\tau_\sigma}{\tau_d} = \frac{\mu \dot{\gamma} a}{\sigma_{eq}}. \qquad (5)$$

For a highly viscous droplet with $\lambda \gg 1$, the characteristic shape relaxation time, $\tau_\sigma = \lambda \mu a/\sigma_{eq}$. Assuming liquid-like properties of bovine milk fat, with mean Sauter diameter of globules as $a = 1\,\mu\text{m}$, $\lambda\mu = 80\,\text{mPa}\cdot\text{s}$, $\sigma_{eq} = 5\,\text{mN}\,\text{m}^{-1}$, and $\dot{\varepsilon}$ or $\dot{\gamma} = 10^3\,\text{s}^{-1}$ gives $Ca \approx 0.04$. This implies substantial drop deformation could occur for $\dot{\varepsilon} > 10^3\,\text{s}^{-1}$, especially for drops much larger than the mean size. Even though Mulder and Walstra[96,97] had argued that drops remained undeformed in the comparable shear rate range, we noticed they incorrectly estimated τ_σ using suspending fluid viscosity. Using the higher viscosity of the droplet phase provides the correct and higher estimate for Ca, possibly in the last stage, $Ca > Ca_c$. The classical studies of flow-induced drop deformation for clean or surfactant-coated interfaces show drops can deform from ellipsoids to longer slender shapes at high Ca and even undergo breakup under fast and fast-evolving flows.[57,98–105] The value Ca_c is lower for extensional flows. However, an even better estimate would require careful accounting for the membrane properties and mechanics, the influence of other dispersed drops and particles, and the effect of confinement in rapidly pinching necks. We estimated $Ca \sim 10^{-3}$ using Eq. (5) for plant milk like soy, using $\lambda \leq O(1)$, $a = 0.6\,\mu\text{m}$, $\mu = 10\,\text{mPa}\cdot\text{s}$, $\sigma_{eq} = 30\,\text{mN}\,\text{m}^{-1}$, and $\dot{\gamma} = 10^3\,\text{s}^{-1}$. Due to the combination of higher viscosity of suspending fluid and higher interfacial tension (and interfacial rheology) than animal milks, the capillary number for plant milk droplets could be consistently smaller and considered as $Ca \ll 1$ in all cases. Furthermore, due to the contribution to stresses generated by polysaccharides (and possibly proteins and particles interacting with polysaccharides), the rate of growth of Ca is significantly slower for plant milks that display elastocapillary pinching.

Milks as emulsions with extra dispersed particles and macromolecules

The experimentally obtained rate-dependent shear viscosity of animal and plant milks can be compared to the theoretical estimates for emulsions detailed in many reviews[57,106,107] and textbooks.[61,108] As cow milk has a low droplet or fat globule volume fraction (ϕ), Taylor's formula for dilute emulsion appears appropriate. Still, studies reported severe underestimates for shear viscosity if only the effect of fat globules is considered.[8,96,97,109] Practically, the shear viscosity of animal milk shows the combined influence of milk fat globules, lactose, whey proteins, casein micelles, charge interactions, and possibly, the complex properties of MFGM are accounted for. The physicochemical properties of the suspending phase, an aqueous solution containing lactose and minerals, such that milk pH at 25 °C is between 6.5 and 6.7, also influence interactions and viscosity.[96,97] The viscosity of whole and skim animal milk is said to be captured by Eilers empirical equation written as $\eta_r = \left(1 + \frac{1.25 \sum(\phi_i)}{1 - \sum(\phi_i)/\phi_{\text{max}}}\right)^2$ which incorporates the influence of any dispersed ingredient i with the volume fraction, ϕ_i and a size, an order of magnitude greater than a water molecule.[96,97] Here, a relatively high value $\phi_{max} \sim 0.9$ is used for fluid milk products as consistent with size/interaction polydispersity and deformability of dispersed species.[96,97]

A quantitative analysis of different plant milks accounting for ϕ_i is warranted but not available. Unlike animal milks, plant milks contain polymer additives that make their continuous phase non-Newtonian, and this influences every aspect of drop deformation, coalescence, breakup, and interactions.[57,67] The viscosity ratio of plant milks can be approximated as $\lambda \sim \frac{50\,\text{mPa}\cdot\text{s}}{10\,\text{mPa}\cdot\text{s}} = 5$ by considering the significant effect of the addition of gellan gum on the continuous phase viscosity, and assuming room temperature oil viscosity of the dispersed phase as $\eta_d = 50\,\text{mPa}\cdot\text{s}$ from an approximation of known viscosities of canola, soybean, and sunflower oils.[70] In contrast, the viscosity ratio of bovine milk can be approximated as $\lambda \sim \frac{80\,\text{mPa}\cdot\text{s}}{1\,\text{mPa}\cdot\text{s}} = 80$ by assuming a water-like viscosity for the continuous phase. The dispersed phase in animal milk typically consists of fat globules, solid-like at refrigeration temperature, and solubilized and liquified at room temperature. Though a higher viscosity ratio of animal milks could indicate less drop deformation,[61] a thorough assessment should be made only after shape relaxation time and deformation rates are considered.

We anticipate revisiting the question of drop deformation in plant milks using a model system with known and fewer components to make a better comparison with theories, experiments, and numerical computations of drop deformation in shear and extensional flows, summarized in our recent review of drop deformation and emulsion rheology,[57] and classic papers by Leal, Mason, and coworkers.[67,98–102,104,105,110,111] Of great relevance for plant milks are more recent studies of deformable particles or drops in non-Newtonian fluids with attention to the influence of interfacial viscoelasticity.[57,67,74,112] We anticipate that by outlining suitable measurement protocols for characterizing shear and extensional rheology, illustrating contrast in the flow behavior of animal and plant milks, and discussing the role of dispersed colloidal particles, drops, and macromolecules, this contribution will facilitate the formulation and design of more nutritious and sustainable future milks.

CONCLUSIONS

We characterized the shear viscosity as a function of the shear rate of two animal-based milks (cow and goat) and six plant-based milks (soy, pea, oat, coconut, almond, and rice) using standard torsional rheometry. Animal milks display rate-independent constant shear viscosity for the nominal shear rate range (1–$10^3\,\text{s}^{-1}$). In contrast, plant milks, except rice milk, display shear thinning. We report the first detailed comparisons of pinching dynamics and extensional rheology response of animal and plant milks, carried out using dripping-onto-substrate (DoS) rheometry protocols. Despite a constant shear viscosity twice the viscosity of the water-like suspending fluid, the animal milks displayed a distinctive terminal viscoelastocapillary (TVEC) response governed by extensional viscosity ten times the shear viscosity. We estimated that the IC-TVEC transition occurs for animal milks as the critical capillary number is likely exceeded for bovine milk. The Capillary number, Ca, i.e., the product of deformation rate and shape relaxation number, captures the magnitude of flow strength or deformation rate. In contrast with animal milks, rice and almond milk display inertiocapillary pinching, including characteristic conical neck shape and power law with an exponent of 2/3. Oat milk appears to be a power-law fluid in both shear and extensional rheology response and displays shear-thinning and extensional-thinning. Three highly shear-thinning plant milks—soy, pea, and coconut—display an elastocapillary pinching followed by terminal viscoelastocapillary regime in radius evolution and manifest strain hardening in apparent extensional viscosity vs strain plot: these responses appear consistent with rheology modification by polysaccharide additives. We find that rheologically, plant milks are distinct from animal milks in both shear and extensional rheology, arguably due to changes in suspending fluid and overall emulsion rheology caused by the influence of polysaccharide additives. Animal milks show constant shear and extensional viscosity. In contrast, except rice, plant milks show shear thinning and strain hardening (soy, pea, and coconut), leading to distinct mouthfeel, texture, foamability, and foam stability compared to animal milks. We anticipate carrying out visualization of milk drops in controlled flow fields would enable us to see if drop deformation shows an analogy with $Ca > Ca_c$ behavior leading to slender drop shapes, the formation of satellite drops, tip-streaming, and filament stretching. Looking forward, we expect that our ability to reformulate plant milks will benefit from recognizing the role of many types of dispersed colloidal phases (oil drops, particles, proteins, and polysaccharides), colloidal forces, viscoelastic interfaces, and non-Newtonian suspending fluids.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Lena Hassan: Formal analysis (equal); Investigation (equal); Methodology (equal); Validation (equal); Visualization (supporting); Writing – original draft (equal); Writing – review & editing (supporting). **Karim Al Zahabi:** Formal analysis (equal); Methodology (equal); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal). **Nadia N. Nikolova:** Formal analysis (supporting); Validation (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Michael W. Boehm:** Conceptualization (supporting); Investigation (supporting); Project administration (supporting); Resources (equal); Writing – review & editing (supporting). **Stefan K. Baier:** Conceptualization (equal); Investigation (equal); Methodology (supporting); Project administration (equal); Supervision (supporting); Validation (equal); Writing – review & editing (supporting). **Vivek Sharma:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Resources (equal); Software (equal); Supervision (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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