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Novel Technological Paper

Soft Matter Science Leads to Healthier and Tasteful Chocolate

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Chocolate is one of the most popular food types and flavors in the world [1-3]. Unfortunately, at present, chocolate products contain too much fat, leading to obesity. While this issue was called into attention decades ago and led to some patent application, no solution was actually found. In order to bypass this issue, two manufacturers, Proctor and Gamble and Nabisco, introduced some low calorie fats to substitute for cocoa butter. Somehow, their fat substitutions can only be used in a limited number of countries. Chocolate lovers deny their products as chocolate. Here we show that this issue is deeply related to the basic science of soft matters, especially to their viscosity and maximally random jammed (MRJ) density. With application of an electric field pulse to liquid chocolate, we aggregate the suspended spherical cocoa particles into prolate spheroids in micro-meters. This microstructure change increases the MRJ density significantly, breaks the rotational symmetry, making the viscosity anisotropic and especially, reducing the viscosity along the flow direction substantially. Hence the fat level in chocolate can be effectively reduced. We are looking forward to a new class of healthier and tasteful chocolate coming to the market soon.

Chocolate is a healthy food. Research has found that chocolate, eaten in moderation, can lower blood pressure and positively affect the circulatory system. Cocoa solids are also one of the richest sources of flavanol antioxidants [1-3]. Unfortunately, present chocolate products contain too much fat, leading to obesity. For example, a typical molding chocolate has fats up to 40% in total, including cocoa butter, milk fat, and other vegetable fat. Chocolate for covering ice cream has fat 50 - 60%. Especially, as children are the leading chocolate consumers, reducing the fat level in chocolate products to make them healthier is important and urgent. This issue was called into attention and elaborated in articles and books decades ago and led to some patent application, but all previous attempts were failed [3,4]. To bypass this issue, Proctor and Gamble and Nabisco introduced some low calorie fats to substitute for cocoa butter in their chocolate flavor products. Somehow, their fat substitutions are not allowed in Canada and Europe, etc.

Why is reducing the fat level in chocolate so difficult? What is the underlying science? In this letter, we will show that the issue is deeply related to the basic science of soft matter. Most people think of chocolate as a solid. To chocolate makers, however, chocolate is a liquid and only solidified just before it is packed for warehouse or shop.

Liquid chocolate is a suspension of solid particles consisting of cocoa, sugar, milk solids etc. in a base liquid of melted fats, mainly cocoa butter. There are two important quantities playing key roles here: maximally random jammed (MRJ) density [5], ϕ_x , and particle's intrinsic viscosity, ν , which depends on the particle shape.

Einstein first studied the viscosity of dilute liquid suspensions and found $\nu = 2.5$ for spheres [6,7]. Also ϕ_x is about 64% for suspensions with mono-disperse spherical particles. The following Krieger-Dougherty formula was generalized for liquid suspensions made of different shape particles with volume fraction ϕ [8],

$$\eta = \eta_0 \left(1 - \phi / \phi_x\right)^{-\nu\phi_x} \qquad (1)$$

where η_0 is the viscosity of base liquid.

Under microscope, the cocoa solids are confirmed to be spherical (Figure 1a). The size is around 2 μm. Therefore, for such chocolate, the total fats cannot be lower than $1-\phi_x = 36\%$. Otherwise, the liquid chocolate is jammed and the production becomes impossible. According to equation 1, even if ϕ is smaller than but close to ϕ_x, removal of a small amount of fat from liquid chocolate would sharply increase its viscosity to interrupt the production. This is why all previous attempts, such as heating and shearing, were failed. The key to reduce the fat level in chocolate is to discover some unconventional method to accomplish the following two tasks simultaneously:

1) Reducing the viscosity of liquid chocolate effectively at high ϕ; 2) Increasing ϕ_x, the MRJ density.

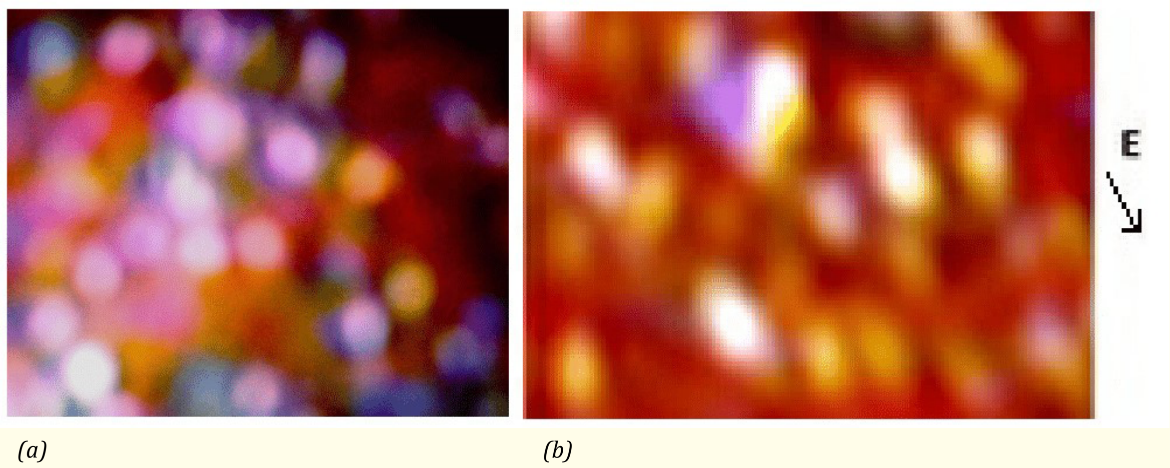

Figure 1: *(a) The original cocoa solids in liquid chocolate are spherical, around 2 μm. (b) Electric field aggregates these particles into short chains, which are similar to prolate spheroids.*

The failure of previous efforts clearly indicates that conventional methods cannot accomplish these two tasks; unconventional technology is critically needed.

In this work, we report that electric field pulse provides an excellent solution for the issue. The approach is illustrated in figure 2. Liquid chocolate flows along a production pipe. A short electric field pulse is applied to the chocolate as it passes the local electric field. Since the dielectric constant of cocoa solid $\varepsilon_p \approx 2.5$, higher than the dielectric constant of melted fats $\varepsilon_f \approx 1.8$, the particles are polarized inside the electric field [9,10],

$$\vec{p} = 4\pi\varepsilon_f \vec{E}_{loc} a^3 (\varepsilon_p - \varepsilon_f)/(\varepsilon_p + 2\varepsilon_f) \qquad (2)$$

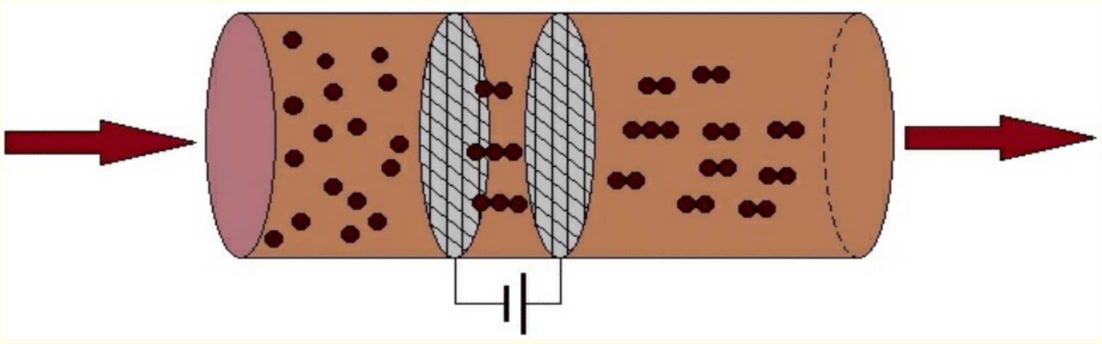

Figure 2: *As liquid chocolate passes a strong local electric field, the solid particles aggregate along the field direction to form streamline aggregates and the viscosity along the flow direction is reduced.*

where $\vec{E}_{loc}$ is the local electric field acting on the particles, stronger than the applied electric field. The interaction between two dipoles is given by

$$U = p^2 (1 - 3\cos^2\theta)/(4\pi\varepsilon_f r^3) \qquad (3)$$

Here r is the distance between the two particle centers and θ is the angle between the field and the line joining the two dipoles. When $\theta = 0$ and $r = 2a$, U has the minimum $U_{min} = -p^2/(16\pi\varepsilon_f a^3)$. Therefore, the dipolar interaction forces the particles to aggregate into short chains along the field direction quickly. The images under microscope confirm it. The aggregated short chains are similar to prolate spheroids (Figure 1b). This microstructure change immediately brings two significant changes in macroscopic properties of liquid chocolate.

First, the MRJ density, ϕ_x , strongly depends on the particle shape. As shown in figure 3, spheroids have a higher MRJ density than spheres. In fact, for spheroids $\phi_x \geq 0.72$, while sphere have MRJ density 0.64 [11,12]. Therefore, the minimum amount of base liquid is down from 36% to 1 - 0.72=28%, reduced by 22.2%, In fact, because electric field induced aggregation also makes the particle size more poly-dispersed, the poly-disparity further increases the MRJ density ϕ_x . Second, the aggregation of short chains along the field direction breaks the rotational symmetry, making the viscosity of liquid chocolate anisotropic [13-15]. Along the field direction, the viscosity is significantly reduced. As the electric field is in the flow direction, the liquid chocolate flow is improved by the viscosity reduction [15]. The amount of fat can thus be significantly reduced.

Let us denote prolate spheroid with its rotational z-axis along the flow direction as

$$(x^2 + y^2)/b^2 + z^2/a^2 = 1 \qquad (4)$$

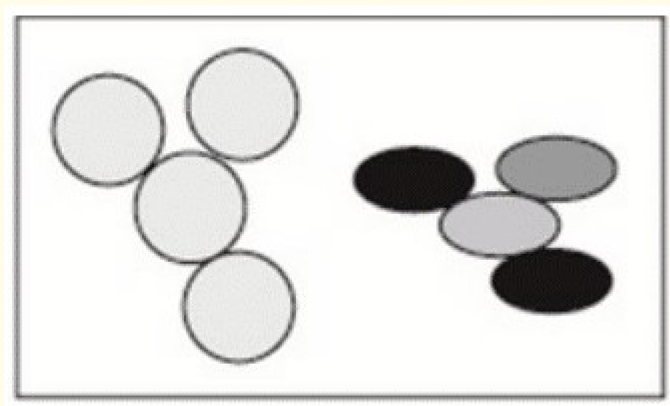

Figure 3: *Spheroids have a higher MRJ density than spheres.*

For such spheroid, the intrinsic viscosity along the z-axis $\nu_{||}$ is smaller than the intrinsic viscosity of spheres 2.5, while the intrinsic viscosity along the directions perpendicular to the z-axis, $\nu_{\perp}$, is higher than 2.5 [16]. If $b/a = 1/2$, for example, we have $\nu_{||} = 2.174$ and $\nu_{\perp} = 2.819$.

To estimate the viscosity, we consider a liquid chocolate model, consisting of 60% cocoa solid and 40% of melted fats. Before the electric field treatment, the relative viscosity of liquid chocolate is given by

$$\eta/\eta_0 = (1 - 0.6/0.64)^{-2.5*0.64} = 84.45 \qquad (5)$$

After the electric field treatment, if all particles are aggregated into the prolate spheroids of the same size, the relative viscosity along the flow direction is reduced to

$$\eta_{||}/\eta_0 = (1 - 0.6/0.72)^{-2.174*0.72} = 16.52 \qquad (6)$$

down 80.4%. The relative viscosity along the directions perpendicular to the flow is given by

$$\eta_{\perp}/\eta_0 = (1 - 0.6/0.72)^{-2.819*0.72} = 37.82 \qquad (7)$$

The higher viscosity in the directions perpendicular to the flow does not provide any resistance to the flow, but helps to suppress vortex formation and turbulence [15].

Because $\eta_{||}$ is significantly reduced, we can reduce the fat level inside the liquid chocolate. For example, because, $(1-0.6777/0.72)^{-2.174*0.72}=84.45$ we can increase the particle volume fraction to 67.77% and reduce the total fat from 40% to 32.23% while keeping the treated liquid chocolate with the same viscosity as the original one. The new chocolate has the fat level reduced by 19.4%, very significant.

If we just want to reduce the total fat by 10%, from 40% down to 36%, we make the particle volume fraction 64%. The relative viscosity of electric field treated chocolate along the flow direction is $(1-0.64/0.72)^{-2.174*0.72}=31.16$, 62.9% down from the original viscosity, much better for production.

The above theoretical prediction has been verified by our experiments. The key is to confirm that application of electric field can effectively reduce the viscosity of liquid chocolate. Traditionally, measuring chocolate's viscosity utilizes rotational viscometers. However, rotational viscometers cannot measure anisotropic viscosity properly. Therefore, we have to invent our own device. As shown in figure 4, chocolate fills the top container and is maintained at pre-specified temperature. In addition to gravity, nitrogen gas is also used to apply additional pressure to control the chocolate flow, which passes the electrodes, metal meshes, and a capillary tube to a cup on a microbalance. On the electrodes, we can apply a voltage to produce an electric field in the flow direction. The microbalance measures the collected chocolate mass as a function of time, monitoring the flow rate, *Q*. The capillary tube serves as a viscometer, providing the chocolate's viscosity along the flow direction as

$$\eta=\frac{\pi\rho^2 gR^4}{8Q}\left(1+\frac{h}{L}+\frac{P}{L\rho g}-\frac{v^2}{2gL}\right) \qquad (8)$$

Figure 4: *Our test device.*

Here *R* and *L* are the capillary tube's radius and length respectively, h is the height of chocolate level above the capillary tube, ρ is the liquid chocolate's density, g is the acceleration of earth gravity, and P is the pressure of nitrogen gas. The average flow velocity is given by $v=Q/(\rho\pi R^2)$.

The typical results are in figure 5a, which recorded the collected chocolate mass as a function of time. The curve's slope is the flow rate. This chocolate sample was from Mars Chocolate and the test was at 40^{0}C. In addition to gravity, we applied 3 psi pressure. Without electric field applied, the flow rate was initially 0.155 g/s, indicating the viscosity was 1088.59 poise. After an electric field of 1600V/cm was applied, the flow rate was increased to 0.2705 g/s, up 74.3%, indicating that the viscosity along the flow direction was reduced to 614.65 poise, down 43.5% (Figure 5b, 5c). With such viscosity reduction, we can easily reduce the fat level by 10% or more.

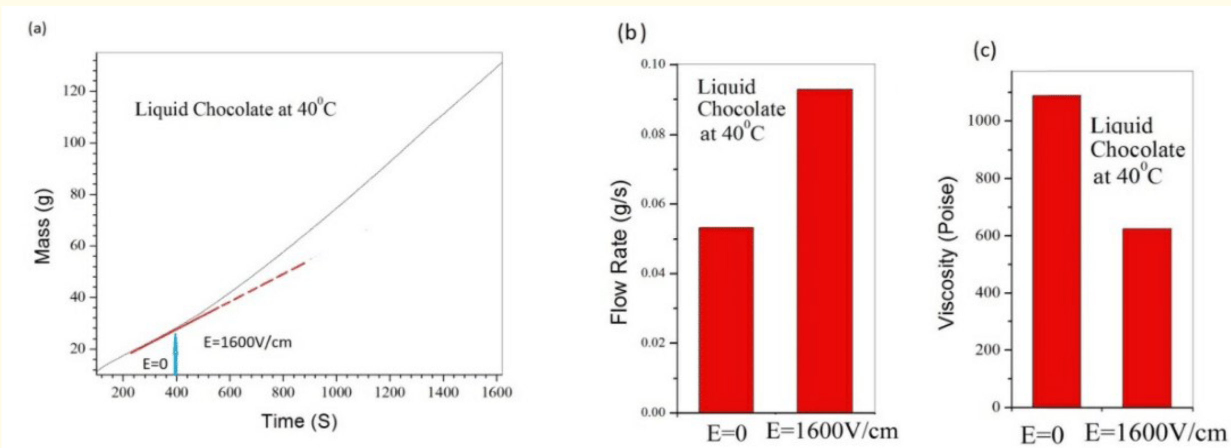

Figure 5: *(a) Application of electric field 1600V/cm significantly increases the flow rate. (b) The flow rate was increased by 74.3%. (c) The viscosity was reduced by 43.5%.*

Recently, we conducted tests with samples from all major chocolate manufacturers, such as Mars, Hershy, and Blommer. The results are quite similar as in figure 5. With application of electric field, we can effectively reduce the viscosity of all kinds of chocolate by 40 - 50%. Therefore, we can easily reduce the fat level by 10% or more for all these samples. It is clear that this method is universal, applicable to all kinds of chocolate [17].

The above results fully confirm the theoretical analysis. On the other hand, in comparison with the theoretical prediction, there is still some room for improvement. There are two important parameters: The electric field strength and pulse duration, i.e. the time for chocolate inside electric field. If we find the optimal range for these two parameters, we can further reduce the chocolate's viscosity.

We note that for liquid chocolate, the relationship between the shear stress and shear rate can be approximated by Casson model. Here is a shear thinning effect, which was used to reduce its viscosity [4]. In our tests, when the electric field is applied, there is no additional shear force applied. The viscosity reduction by the electric field also well exceeds the reduction from the shear thinning effect.

Naturally, there is a question: How long can such reduced viscosity last after the electric-field treatment? To answer the question, we conducted a number of tests and found that the reduced viscosity kept more than 48 hours. The above results are understandable. The aggregated short chains make the suspension as a viscoelastic fluid. To dissemble such viscoelastic chains is very slow [13]. The viscosity is kept reduced as long as the short chains remain.

The electric field treated chocolate has wonderful taste. Some people even claim that the electric field treated chocolate has a slightly stronger cocoa solid flavor, better than the original chocolate. We are thus looking forward to a new class of healthier and tasteful chocolate products to market soon.

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