Journal of Oleo Science
Copyright ©2022 by Japan Oil Chemists' Society
doi : 10.5650/jos.ess22148
J. Oleo Sci. **71***, (11) 1577-1589 (2022)*

JOURNAL OF OLEO SCIENCE
Jos

Effect of Sugar and Milk Powder Addition on the Mechanical Properties and Texture of Chocolate

Yu-yao Chen[1,2], Xing-yu Zhou[1,2], Shan-hua Qian[1,2], and Jing-hu Yu[1,2]*

[1] *School of Mechanical Engineering, Jiangnan University, Wuxi, Jiangsu, 214122, CHINA*
[2] *Jiangsu Province Key Laboratory of Advanced Food Manufacturing Equipment & Technology, Wuxi, Jiangsu 214122, CHINA*

Abstract: In order to investigate how the addition of two common ingredients in chocolate, sugar and milk powder, affects the mechanical properties of solid chocolate, uniaxial compression tests and wedge fracture tests were carried out using four ratios of chocolate as the experimental material. Mechanical properties such as Young's modulus and fracture toughness were directly correlated with textural properties such as hardness, elasticity, and brittleness. The results show that adding sugar increases Young's modulus and fracture toughness of chocolate, while milk powder is the opposite. In equal amounts of both, sugar played a more substantial role. In combination with the properties exhibited by chocolate in the above tests, data from creep tests were collected to improve the classical Bingham model and develop a new constitutive model for predicting the mechanical behaviour of solid chocolate with different ratios of added sugar and milk powder. The new four-component constitutive model allows for a more accurate fit to the creep test data and this work provides some suggestions for making different tasting chocolates by adjusting the addition of ingredients.

Key words: chocolate, mechanical properties, sugar, milk powder, constitutive model

1 Introduction

Chocolate is one of the most popular confectionery products globally[1)], and high-quality chocolate requires a glossy surface, a soft texture, and a certain chewiness. Chocolate's unique mechanical behaviour during oral processing has unique sensory properties[2)]. The physical properties, rheological behaviour, and sensory perception of chocolate are largely influenced by its processing, particle size distribution, morphology and composition. To enhance the texture of chocolate, the solid particle size distribution and composition can be controlled to modify its physical properties, rheological characteristics, and sensory attributes. In recent years, several improvements in chocolate quality have been made through changes in processing strategies. However, using a composition as a tool to modify the texture of chocolate still requires a deeper understanding of the principles and factors that influence the mechanical properties[3)].

Chocolate can be defined as a suspension of solid particles. These solid particles are derived from sugar, milk powder, and cocoa solids in the continuous fat phase, which determines the final form of chocolate and influences its flavour, colour, and texture.

Sugar is mainly used as sweetener and filler in chocolate, which influences taste, texture, and flavour of chocolate. Sugar and fats are not mutually soluble, and this system is relatively stable but complex in phase change. Some sugars can participate in non-enzymatic browning reactions, such as the maillard reaction, which gives chocolate unique flavours and colours. It has been shown that sugar particles can provide non-uniform nucleation sites for the crystallization of cocoa butter and tend to accelerate the crystal growth rate[4, 5)].

Milk powder is recognized as a food with high nutritional value. Its protein content is high and contains various amino acids needed by the human body, with a nutritional absorption rate of 90%-100%. The high free fat content in full-fat dairy powder reduces the amount of cocoa butter in chocolate products[6)]. The addition of whole milk powder reduces cocoa butter's crystallization and melting temperature. It is well known that the microstructure of chocolate significantly affects the rheological behaviour of the material and textural properties such as hardness[7−9)].

Therefore, studying the effect of sugar, milk powder, and other ingredients on the quality of chocolate is an essential guideline for adjusting the formulation of chocolate and

---

*Correspondence to: Jing-hu Yu, School of Mechanical Engineering, Jiangnan University, Wuxi, Jiangsu, 214122, CHINA
E-mail: jhyu@jiangnan.edu.cn
Accepted July 25, 2022 (received for review April 20, 2022)
Journal of Oleo Science ISSN 1345-8957 print / ISSN 1347-3352 online
http://www.jstage.jst.go.jp/browse/jos/ http://mc.manusriptcentral.com/jjocs

products and improving the quality of products.

At present, much research has been conducted on the rheological properties of chocolate to determine how the taste of chocolate can be changed by adding different ingredients.

Belščak[10] studied the sensory properties of chocolate with the addition of different sweeteners, and although the use of low-calorie sugars was able to achieve the same level of sweetness as conventional chocolate, the overall acceptability was lower. It was also measured that the addition of sweeteners such as xylitol increased the hardness of the chocolate because of the larger particle size of these compounds. Sokmen[11] studied the effect of l different particle size sweeteners on the rheological properties of molten chocolate and found that the addition of maltitol resulted in similar rheological properties compared to sucrose. Yield stress of chocolate samples with maltitol was significantly higher than that with isomalt. In contrast, isomalt increased plastic viscosity due to its number of particles. All these studies give a theoretical basis for the production of lower-calorie chocolate.

In contrast, the mechanical behaviour of solid foods can usually be characterized by material properties, such as Young's modulus, yield strength, and fracture stress[12]. Bikos[13] investigated the effect of micro aeration on the mechanical properties of chocolate products and found that micro aeration enhanced the brittleness of chocolate and had significant effects on the material properties such as Young's modulus, yield strength, and fracture stress. And these material properties are directly related to the hardness, brittleness, and other textural properties of the chocolate that determine the deformation and rupture of the material under complex loading conditions observed during the first bite.

Extensive literature has been devoted to the rheology of molten chocolate. However, the mechanical behaviour of solid chocolate has been less explored in depth. The sugar and milk powder content of chocolate significantly affects its taste and flavour release, but the existing literature does not explore in-depth the effect of sugar and milk powder addition on the mechanical and fracture properties of solid chocolate. A more accurate food propriety model is needed to predict the mechanical response and fracture behaviour of solid chocolate at the first bite.

This study investigated the mechanical behaviour of four chocolate products (dark chocolate made from 70 g of cocoa liquor and 20 g of cocoa butter, three kinds of dark chocolate with 10 g of sugar added, 10 g of milk powder added, 5 g of cane sugar and 5 g of milk powder added) with different formulations under mechanical action to explore this issue. Uniaxial compression and wedge fracture experiments were performed on all chocolate products to highlight the effect of ingredient addition on the mechanical and fracture properties, respectively. The mechanical parameters and stress-strain data were obtained by testing four different formulations of chocolate products and calibrating the constitutive model of chocolate to predict the changes in the mechanical behaviour of different ratios of chocolate in oral processing.

In this study, an attempt was made to investigate the effect of sugar and milk powder addition on solid mechanical properties of chocolate and relate it to the internal structure of chocolate at different ratios. Certain recommendations are provided for the creation of chocolate with a specific taste.

2 Materials and Methods

2.1 Materials

The basic chocolate sample is made of cocoa liquor block and cocoa butter. The raw materials, sugar, and milk powder originated from Shaoxing, Zhejiang Province. The recipe for the chocolate specimen production is shown in Table 1.

The cocoa liquor blocks and cocoa butter are heated in a water bath at 60℃ underwater for 1 hour. Powdered sugar and milk powder from the ingredient list above was gradually added to the melted cocoa ingredients. The mixture was continuously stirred at 200 rpm for 1 hour using a mechanical stirrer to ensure that all the added ingredients were evenly distributed in the molten chocolate. The mixture was then cooled to 27.2℃. Finally, the molten solution was reheated to 32.2℃. Untempered chocolate is soft and not effectively demoulded. The tempered chocolate solution was poured into cylindrical moulds (24 mm diameter, 13 mm depth) for uniaxial compression experiments and rectangular moulds (20 mm depth, 15 mm width, 50 mm length) for wedge fracture tests. Before demolding, the moulds were placed in a food-grade constant temperature refrigerator (4℃, two hours). In this study, four sample formulations of chocolate with different functional ingredients were made. These were dark chocolate (DC), chocolate samples with added powdered sugar (SC), chocolate samples with added milk powder (MC), and chocolate with added powdered sugar and milk powder mixture (SMC). The fat content in chocolate products affects the final sensory perception of the product[3]. Therefore, the mass of cocoa butter added in all samples

Table 1 Chocolate sample ingredient list.

Chocolate sample	DC	SC	MC	SMC
Cocoa liquor	70 g	70 g	70 g	70 g
Cocoa butter	20 g	20 g	20 g	20 g
Sugar powder	–	10 g	–	5 g
Milk powder	–	–	10 g	5 g

was the same.

2.2 Uniaxial compression tests

Uniaxial compression tests were performed at room temperature of 23℃ on a mass spectrometer (TMS-Pro) sensor with a maximum range of 1 kN. (The maximum load observed during compression was 750 N.) The data obtained from uniaxial compression experiments were used to obtain values of true stress and true strain data through the relationship given by Eqn (1):

$$\left.\begin{aligned}\sigma &= F/A_i\\ \varepsilon &= ln(H_i/H_0)\end{aligned}\right\} \quad (1)$$

Where F is the force, H_0 is the original reference size of the sample, H_i is the current sample height, A_i is the instantaneous cross-sectional area of the sample, assuming that all samples are entirely incompressible, Poisson's ratio$\upsilon = 0.5$. The current cross-sectional area of the sample is calculated from the original cross-sectional area of the sample:

$$H_i A_i = H_0 A_0 \quad (2)$$

Compression tests were performed at three constant compression rates of 0.1 mm/s, 0.5 mm/s, and 1 mm/s by applying load in a straight line to 90% of the sample height and repeating each case three times. These rates were chosen because the compression rate also affects the results of the compression experiments, and 0.5 mm/s is the compression rate commonly used for testing the mechanical properties of food[14].

Loading and unloading tests were performed on all chocolate products in compression mode to highlight any inelastic effects. Cyclic loading tests were performed on four samples with initial loading strain at three levels of 0.025, 0.08, and 0.16, followed by unloading to return the specimens to the zero-force position. The procedure was repeated three times for each condition and each type of chocolate, with the loading phase at a rate of 0.1 mm/s. After converting the force-time curves to true values Young's modulus could be calculated as the slope of the initial linear region. The region for calculating Young's modulus should not contain any significant fracture events.

Creep tests were performed on all chocolate samples with a constant load to obtain strain-time curves to calibrate the constitutive model. Creep tests were performed on four chocolates at a loading rate of 0.1 mm/s at constant stress. The experiments were performed at constant stress lower than the compressive fracture stress of the chocolate to avoid the fracture behaviour affecting the accuracy of the constitutive model.

2.3 Cutting fracture tests

In the wedge test, the wedge angle is 15°, the radius of curvature of the wedge tip is 5 mm, and the width of the wedge is 40 mm (wider than the sample, to make the sample split completely so that the area of the fracture surface becomes a known value[15]), is mounted on a mass spectrometer so that the wedge probe divides the food sample. The wedge separates the sample by internal tension, thus testing the resistance of the food to type I fracture. During the experiment, the wedge was passed through four different ratios of chocolate samples at the rate of 0.1 mm/s and 0.5 mm/s. The wedges were then removed, and the split samples were gently pushed back together and then subjected to this test again[16]. The data obtained from the wedge fracture experiments were used to calculate the fracture energy and fracture toughness by the following procedure.

The wedge test assumes that the plastic dissipation or elastic stored energy in the sample edges is negligible in both processes ($U_e = U_p = 0$). In the second process, the recorded force is caused only by the friction between the wedge and the sampling process ($U_{f,pass2} = 0$). The assumption allows the energy balance of each process to be expressed as Eqn (3) and by simplifying to obtain Eqn (4), (5).

$$U_s = U_f + U_p + U_d + U_e \quad (3)$$

$$U_{s,pass1} = U_f + U_d \quad (4)$$

$$U_{s,pass2} = U_d \quad (5)$$

Where U_s is the total system energy, U_f is fracture energy, U_p is the energy dissipated by plastic deformation, U_e is the energy dissipated by other processes, and U_d is the elastic energy storage. The units of all energies are [J]. U_p and U_d are zero in the line elastic condition. The total energy applied to the system can be expressed as $U_s = \int_0^h Fdx$, where F is the force applied to the sample, x is the displacement, and h is the net displacement obtained during the whole deformation.

Where $U_{s,pass1}$ is the energy applied to the sample during the first test (leading to fracture), $U_{s,pass2}$ is the energy applied to the sample during the second test (empty process). The fracture energy U_f can be found by subtraction. The fracture toughness is given by Eqn (6).

$$Gc = \frac{U_f}{wh} \quad (6)$$

Where G_c is the fracture toughness, w is the sample width, and h is the sample height. The fracture energy of the sample can be calculated from the integration of the force-displacement curve in the stable crack expansion region[17].

The bionic chewing experiment was performed on a bionic chewing platform (made in our laboratory, see Fig. 1), where four different types of chocolate were placed under the cutting teeth of the platform so that the two incisors could completely cut the rectangular chocolate. Gently close the separated chocolate after lifting the inci-

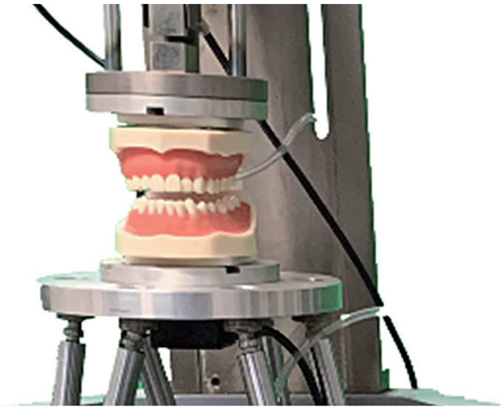


Fig. 1 Bionic chewing platform.

sors by program control and repeat the above steps.

3 Experimental Results

3.1 Uniaxial compression tests

Figure 2 shows the stress-strain diagrams for four different ratios of homemade chocolate at uniaxial compression rates of 0.1 mm/s, 0.5 mm/s and 1 mm/s. The mechanical response of all the chocolate samples exhibited small variations during the three replicate tests. It is observed in Fig. 2 that the stress-strain response of all types of chocolate tested showed a significant dependence on the compression rate. Different mechanical behaviours were produced at different compression rates. MC chocolate had the lowest stress-strain response among the four chocolates, with up to 50% difference compared to SC chocolate, which had the highest stress-strain response.

The Young's modulus of the four chocolates can be obtained from the slope of the linear phase of the stress-strain curve. It has been established in the literature that Young's modulus and fracture stress are directly related to the perceived hardness of the material[18−21]. Chocolate SC containing powdered sugar is harder than chocolate DC containing only cocoa butter and cocoa liquor mass. According to the study by Afoakwa *et al.*[22], the hardness of chocolate is related to the strength of the interactions between the particles. The addition of powdered sugar as a large particle molecule increased the solid volume content of the fatty base of the chocolate and an increase in the intermolecular forces, increasing the hardness of the chocolate. At the same time, it was observed that the chocolate MC containing milk powder had a minor hardness and softer texture. Liang and Hartel[23] found that an increase in free milk fat concentration and a decrease in solid fat

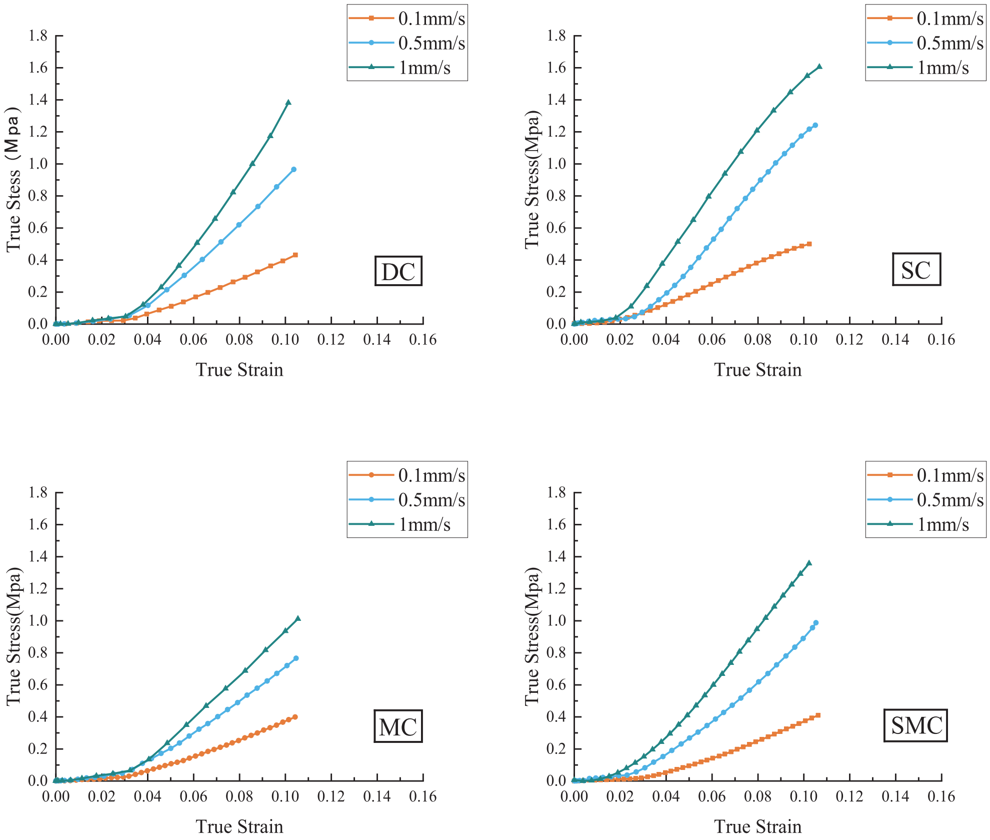


Fig. 2 Stress-strain diagrams of four chocolate samples in monotonic uniaxial compression.

content led to a decrease in chocolate hardness, which is consistent with the results obtained in this paper. MC chocolate contains a proportion of butter fat that causes an eutectic effect, which preventsbloom formation, results in a lower melting point, softeningof texture[24]. The hardness of SMC chocolate was observed to be less than SC chocolate but greater than DC chocolate when both powdered sugar and milk powder were added to the chocolate. The two different component in equal proportions reacted in a complex way in the fat matrix. Therefore, the experimental results showed that the addition of powdered sugar had a more significant effect on the hardness, which enhanced the overall hardness of the chocolate.

The curves of the loading-unloading paths obtained from the loading-unloading cycle experiments for the four chocolates at a strain level of 0.025 overlaps almost exactly, and the stress values are too small hard to plot. So only the loading-unloading cycle curves for the remaining two strain levels of 0.08, 0.16 are shown in Fig. 3. For minor elastic strains of 0.025 strain applied and removed in all tested materials, the loading and unloading paths can be well matched to the predicted linear elastic material. At more minor strain levels, all chocolates exhibit good elasticity. However, for samples at strain levels above 0.08 and 0.16, deformation was not recovered for a more extended period after the cyclic experiments were completed with the removal of the load, implying that permanent inelastic strain occurred in the chocolate during the initial stress-strain response phase of compression.

According to Gonzalez's study[25] on the mechanical properties of fat-based materials, the inelastic deformation during the loading phase of compression experiments is caused by the structural rearrangement resulting from the compression of the fat crystals. It is suggested to determine the elasticity of material by measuring the degree of recovery of deformation of the sample during unloading. The percentage of deformation recovery for the four types of chocolate at two strain levels obtained from the above curves and data calculations are plotted in Fig. 4(a), where the elasticity decreases in the order of SC, DC, SMC, MC for the four different types of chocolate.

In Fig. 3, Young's modulus calculated from the linear unloading part of the curve is summarized for both strain levels. Figure 4(b) summarizes the highest differences that can be observed for Young's modulus values obtained separately for the loading-unloading phase of the cyclic loading test. In the case of the chocolate with only milk powder added, Young's modulus of the loading path obtained in the loading-unloading test is 8 MPa, while the slope of the unloading path is about 16 MPa, with a difference of almost 100% between the two. It was also observed that for SC chocolate, the difference between Young's modulus values

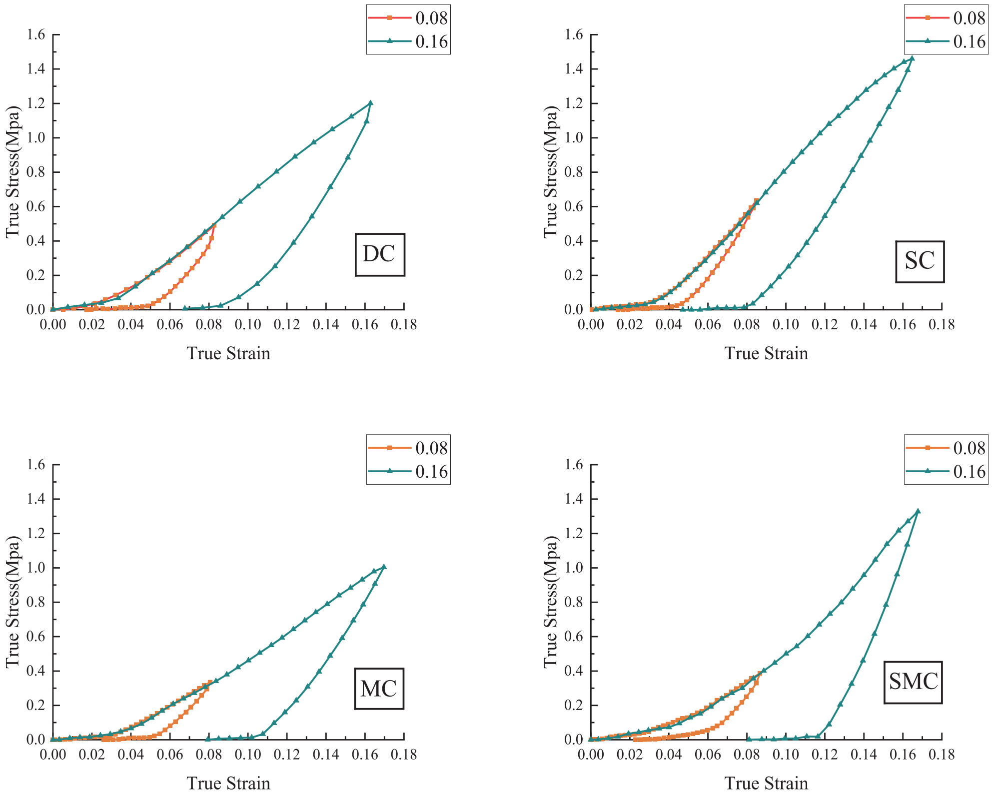
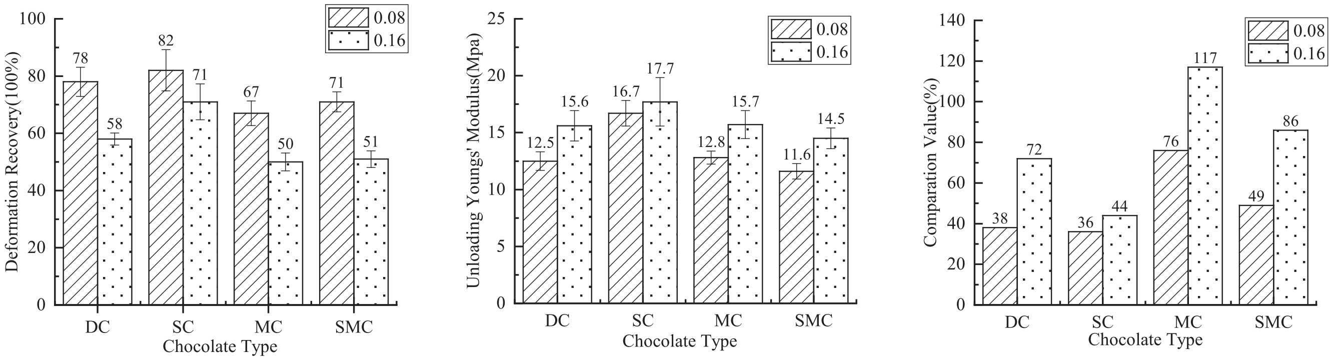


Fig. 3 Stress-strain diagrams of loading-unloading tests for all chocolate samples at different strain levels of 0.08, 0.16 at a compression rate of 0.1 mm/s.



(a) (b) (c)

Fig. 4 Loading-unloading tests results.
(a) Recovery of deformation of chocolate after unloading in the loading-unloading test for all chocolate samples at two different strain levels of 0.08,0.16
(b) Young's modulus values extracted from the unloaded portion of the stress-strain curve in the loading-unloading test at two strain levels of 0.08,0.16 for all chocolates
(c) Comparative values of Young's modulus extracted from the loaded part and Young's modulus extracted from the unloading part in the loading-unloading test for all chocolates at two strain levels of 0.08, 0.16

in the unloading path and those in the loading path was smaller compared to DC chocolate, which showed better elasticity. In contrast, the slope of the MC chocolate unloading path is much larger than that of the loading path, and the stress-strain response is closer to that of visco-plasticity. These inelastic effects seem to come from the early stage of material deformation, where the higher the free fat content, the higher the destruction of the fat crystal structure during compression, leading to this plastic deformation. The deformation recovery of SMC chocolate is close to that of DC chocolate. At the same time, the difference value of Young's modulus in the loading-unloading stage is larger than that of DC chocolate. The elasticity of SMC chocolate was smaller than that of DC chocolate. Under the conditions of this test, the equal proportion of milk powder had a more significant effect on the elasticity of chocolate than sugar powder.

3.2 Cutting fracture tests

Figure 5 shows the load-displacement plots of wedges cutting four types of chocolate samples with different cutting rates, illustrating the significant rate-dependence of fracture for all types of chocolate materials. In the load-displacement plots for the four types of chocolate, a smaller load-displacement response is found to cut at a cutting rate of 0.5 mm/s than a cutting rate of 0.1 mm/s. Based on the above curves, it can be observed that the calculated parameters such as fracture energy and fracture toughness are listed in Table 2.

The fracture load of SC chocolate with only powdered sugar added was significantly higher than that of DC chocolate. In contrast, the fracture load of MC chocolate with only milk powder added was reduced. The fracture load of SMC chocolate with both powdered sugar and milk powder was similar to that of DC chocolate. When the wedge overcomes the yield stress and enters the chocolate, the curve appears a slight "turn". Through the 'turn', the stress rises continuously before a fracture occurs.

All four chocolates showed significant stress hardening effects before fracture at slower cutting rates. After the yielding stage, the forces between the macromolecules increase, causing the polymer viscosity to rise and the polymer to tend to "harden"26). For chocolate, the continuous rise in stress is due to the nonlinear elastic behaviour associated with structural changes in the fat microcrystals after yielding24). It is observed in Figure 5 that for different ratios of chocolate, the onset of the unstable crack expansion in MC chocolate (the point marked by the arrow in the figure) is less than the wedge penetration in DC chocolate at the same cutting rate, and the less penetration force required, the more brittle the specimen. Also, MC chocolate has lower fracture toughness and fractures energy relative to DC chocolate. The free fat in the milk powder effectively dilutes the dispersed phase volume, thereby reducing yield stress and plastic viscosity27).

The fracture toughness and fracture energy of SC chocolate are greater than those of DC chocolate. The addition of powdered sugar increases the viscosity of chocolate, and the ability of chocolate to resist crack expansion is enhanced; the addition of powdered sugar reduces the brittleness of SC samples. Moreover, milk powder increases milk fat content in the fat base, and milk fat can react with cocoa butter. Milk fat will inhibit the polymorphic transformation of cocoa butter to its most stable state28). Unstable

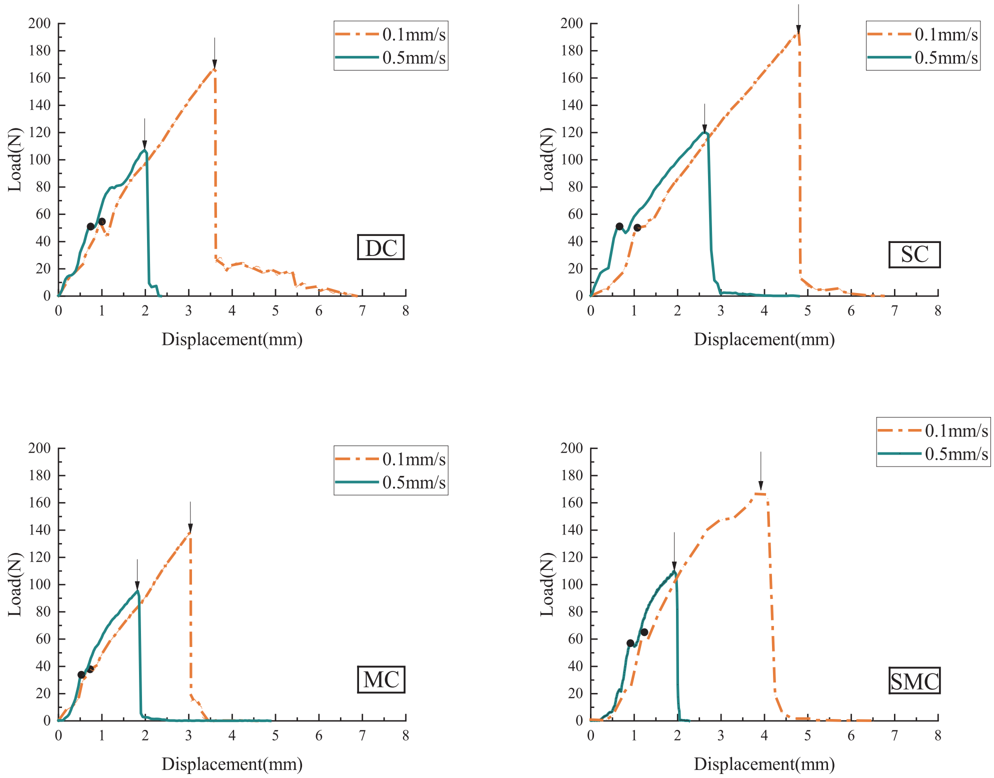


Fig. 5 Displacement-load diagrams from the first cut in the wedge fracture test at cutting rates of 0.1 mm/s and 0.5 mm/s for all chocolate samples (the second cut with a smaller load is not plotted but is included in the calculation).

Table 2 Result of Fracture Test.

Chocolate Sample	Cutting Speed (mm/s)	Fracture Load (N)	Fracture Displacemen (mm)	Fracture Energy (MJ)	Fracture Toughness (J/m^2)
DC	0.1	166 ± 25	3.59 ± 0.32	356 ± 32	1189 ± 83
	0.5	109 ± 12	1.98 ± 0.12	126 ± 16	420 ± 40
SC	0.1	193 ± 14	5.23 ± 0.28	531 ± 38	1170 ± 126
	0.5	120 ± 8	2.63 ± 0.14	199 ± 18	663 ± 60
MC	0.1	129 ± 17	3.04 ± 0.08	219 ± 27	730 ± 90
	0.5	95 ± 7	1.83 ± 0.15	99 ± 13	330 ± 43
SMC	0.1	169 ± 27	3.93 ± 0.12	403 ± 25	1343 ± 90
	0.5	107 ± 9	1.92 ± 0.08	137 ± 20	457 ± 67

cracks will expand rapidly through the unstable structure, and MC chocolates exhibit higher brittleness less fracture toughness. SMC chocolates with both powdered sugar and milk powder added showed higher fracture energy and fracture toughness than DC chocolates, and powdered sugar had a greater effect on enhancing the chocolate's ability to resist unstable cracking.

Fracture mechanics is based on the fact that every item has inherent microcracks that compromise its strength. Once a microcrack absorbs enough energy through an external force, it propagates through the material and causes a fracture. For the crack to expand, the energy must be concentrated around the crack's tip. The energy required for crack expansion depends on the way the material is loaded the yield stress of the material and is related to the geometry of the crack itself. In the experiments of this paper, the material was loaded in a way that the geometry of the crack itself was kept consistent. Therefore the fracture energy required for different types of chocolate directly reflected its yield stress, with the stress values in the order of SC, SMC, DC, MC from largest to smallest.

The observation of the fracture surfaces showed that the chocolate DC and MC without any additives and with only milk powder had flatter fracture surfaces. The chocolate

SC and MC with added powdered sugar had rougher fracture surfaces and more curved cracks, probably due to the different dispersion of powdered sugar in the production process[29]. The inhomogeneity of food particle size significantly affects the distribution and dissipation of strain energy at fracture, and heterogeneity is an essential appendage of brittleness that can produce brittle cracks in different directions[30].

In mastication, the wedge test helps determine the fracture characteristics of chocolate under loading geometry, such as incisor splitting[31]. By linking the fracture characteristics of chocolate measured by the wedge test to the fracture behaviour of chocolate in the first bite of the masticatory test, the results of the wedge test were predicted to be directly related to the masticatory variable.

The displacement-load diagrams of the four types of chocolates obtained from the masticatory experiments based on the bionic chewing platform are plotted in Figure 6. The fracture toughness and fracture energy obtained in the wedge experiments can still predict the brittleness of the four types of chocolates to some extent, and it can still be verified that the addition of powdered sugar reduces the brittleness of the chocolates. At the same time, the milk powder increases the brittleness of the chocolates.

However, fracture displacement and fracture load of SMC chocolate were smaller than those of DC chocolate, only with the same results obtained from the wedge test at a loading rate of 0.5 mm/s. This shows that the loading rate has a significant influence on the chocolate fracture test. The effect of milk powder on the brittleness of chocolate was greater in the masticatory loading mode with equal proportions of milk powder.

In the wedge loading test, the chocolate had a longer time for stable cutting after the wedge was cut in, and the chocolate viscosity had a more significant effect on the test results. It can also be observed that the energy imparted to the sample by friction during the secondary bite break in the imitation chewing experiment is more compared to that in the wedge block test. The shape of the wedge is similar to the shape of the incised tooth and both fracture types are the same. However, the wedge material is not the same as the tooth material, and all parameters obtained in the masticatory-like test differ from the wedge test.

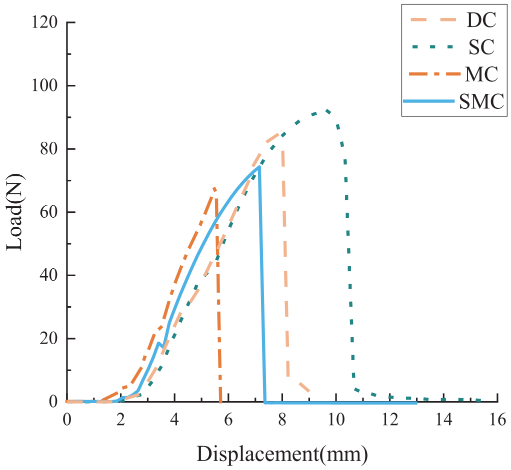


Fig. 6 Displacement-load diagram of all chocolate samples in the first cut in the bionic chewing test (the second cut load is smaller and not plotted in the figure but is involved in the calculation).

The fracture displacements of all four types of chocolate were greater than those of the wedge test performed at the same rate. The radius of curvature of the wedge chosen for the wedge test was similar to that of the tangent, but the fracture curve obtained from the regular shape of the wedge was smoother than that of the masticatory test and the section of the chocolate was smoother at fracture. Therefore, the wedge test can predict to some extent the difference in fracture behaviour of the additive on the chocolate during chewing, but it cannot truly reflect the possible deformation and fracture of the chocolate during chewing.

3.3 Constructive model

Based on the characteristics exhibited by the uniaxial compressive stress-strain diagrams of the four chocolates, suitable constitutive models were developed. A more accurate mathematical model to predict the addition of sugar and milk powder on the material properties, such as Young's modulus of chocolate, was used to evaluate the texture and taste of chocolate with different ratios.

By observing the results of the compression experiments, it can be seen that the stress-strain response of the samples has an obvious rate-dependence, which means that the samples produce a time-dependent irrecoverable deformation during the compression process and exhibit a more obvious visco-plasticity[18].

The unloading phase also shows a certain recovery deformation in the cyclic loading experiments, so the material has a certain elasticity. By the above properties, a suitable mathematical model is established to fit the experimental results to characterize the above material properties.

Creep tests allow for the classification of materials based on their creep behaviour[32], the ideal elastic solid will exhibit constant strain over time due to lack of mobility and will fully recover strain after removal of the load. The ideal plastic solid will also exhibit continuous plastic deformation after exceeding the yield stress with constant stress. On the other hand, the ideal viscous liquid exhibits a linear change in strain over time due to stable flow and zero recovery after unloading[33]. Due to its composition and structure, chocolate exhibits viscous, elastic and plastic behaviour at the same time. Therefore creep tests provide valuable information on the viscoelastic-plastic behaviour.

The rate of deformation of a visco-plastic body under load, when the stress reaches some critical value, yielding and flow phenomena occur, is related to the viscosity of the object. The visco-plasticity of the material can be described by the combination of damping elements and sliding elements. Moreover, the chocolate studied in this paper has the above three properties, so the common basic elastic visco-plastic Bingham model is chosen to describe the strain-time response of chocolate in this paper.

For this model, the total strain is the sum of the strain ε_0 in the spring element and the strain ε_1 in the parallel damping element and sliding element, and the stress in the spring is equal to the total stress, as described by the following Eqn (7) (8).

$$\varepsilon(t)=\varepsilon_0+\varepsilon_1 \tag{7}$$

$$\sigma(t)=\sigma_0=\sigma_1=E_0\,\sigma_0 \tag{8}$$

where σ_0 is the stress in the spring element that exhibits elasticity, the modulus of elasticity is E_0, and the sliding element that exhibits plasticity σ_p depends on whether the yield stress σ_s has been reached, and only when the stress value reaches the yield limit, deformation begins, which can be expressed in Eqn (9) (10).

$$\left.\begin{aligned}&\sigma_p=\sigma_0=\sigma(t)\ (\sigma_p<\sigma_s)\\&\sigma_p=\sigma_s+B\varepsilon_1(\sigma_p\geq\sigma_s)\end{aligned}\right\} \tag{9}$$

$$B=\frac{d\sigma(t)}{d\varepsilon_1}=\frac{d\sigma(t)}{(d(\varepsilon(t))-d\varepsilon_0)}=E_1/(1-\frac{E_1}{E_0}) \tag{10}$$

where B is the reinforcement parameter for stress hardening when the stress in the sliding element σ_p exceeds the yield stress, and E_1 is the tangential modulus in the sliding element.

At the same time, the damping element is connected in parallel with the sliding element so that the strain in both elements is the same and the stress in the two parallel elements σ_1 is the sum of the stress in the sliding element σ_p and the stress in the damping element σ_v, expressed as the following Eqn (11).

$$\sigma_1=\sigma_p+\sigma_v=\sigma_p+\phi\,\frac{\partial\varepsilon_1}{\partial t} \tag{11}$$

where ϕ is the parameter of the damping element, combined with the Eqn (7) (8) (9) (11) can obtain the relationship between $\sigma(t)$ and $\varepsilon(t)$ the Eqn (12).

$$\left.\begin{aligned}&\varepsilon(t)=\frac{\sigma(t)}{E_0}(\sigma_p<\sigma_s)\\&BE_0\,\varepsilon(t)+\phi E_0\frac{\partial\varepsilon(t)}{\partial t}=B\sigma(t)+E_0(\sigma(t)-\sigma_s)+\phi\frac{\partial\sigma}{\partial t}(\sigma_p\geq\sigma_s)\end{aligned}\right\} \tag{12}$$

When the stress applied to the sample is constant value σ_A, the above equation can be rewritten as follows:

$$\varepsilon=\frac{\sigma_A}{E_0}+\frac{\sigma_A-\sigma_s}{B}\left[1-\exp(-\frac{B}{\phi}t)\right](\sigma\geq\sigma_s) \tag{13}$$

Observing the stress-strain curves in the cyclic loading test, it can be observed that the mechanical behaviour before overcoming the yield stress does not precisely match the idealized elastic response. The non-linear phase at the beginning of the loading was observed to be due to some slippage when the probe comes in contact with the chocolate due to the smoother surface of the chocolate. Therefore, the above constitutive model was corrected based on the data obtained from creep tests after a period of loading, keeping the static force constant, and the fitted curves obtained from the data are plotted in the Fig. 7. It was observed that the fit was relatively accurate for all types of chocolate materials.

Based on the fitted constitutive model parameters, the same results as in the previous tests can be obtained, and the fitted values of the modulus of elasticity show a trend from SC, SMC, DC and MC decreasing. The strengthening parameters reflect the degree of stress hardening of chocolate after overcoming the yield stress, and it is easy to see from the results that they are consistent with the conclusions obtained in the fracture tests. Therefore, the model is more consistent with the material properties of chocolate, where the time-dependent parameters are less sensitive to the final strain response, which is not consistent with the experimental stress-strain response of chocolate with a more obvious rate dependence. Based on the above findings, the above equations are adjusted to construct a new constitutive model that is more consistent with the properties of chocolate.

This constitutive model consists of a visco-elastic model consisting of a damping element and a spring element connected in parallel and then in series with an ideal visco-plastic model, where the stresses σ_0 in the visco-elastic model are equal to those in the visco-plastic model, and their stresses can be calculated by the Eqn (14).

$$\left.\begin{aligned}&\sigma_0=E_0\,\varepsilon_0+\phi_0\frac{\partial\varepsilon_0}{\partial t}\\&\sigma_1=\sigma_s+\phi_1\frac{\partial\varepsilon_1}{\partial t}\\&\sigma=\sigma_0+\sigma_1\end{aligned}\right\} \tag{14}$$

Where, ϕ_0, ϕ_1 is the parameter of the damping element, ε_0 is the strain of the viscoelastic model, ε_1 is the strain of the viscoplastic model, and the sum of the two is the strain of the model when the stress used in the constant value σ_A, which can be expressed as Eqn (15).

$$\varepsilon=\varepsilon_0+\varepsilon_1=\frac{\sigma_A}{E_0}(1-e^{-\frac{E}{\phi_0}t})+\frac{\sigma_A-\sigma_s}{\phi_1}t+A \tag{15}$$

In Tables 3 and 4, it can be observed that the newly established constitutive model 2 has a higher degree of fit. Model 2 solves the problem that model 1 exhibits a time-independent fully elastic response before the stress reaches the yield stress, which is not consistent with the experimental results. The increased sensitivity of the parameters of the damping element to the degree of equation fit is more consistent with the results in the previous ex-

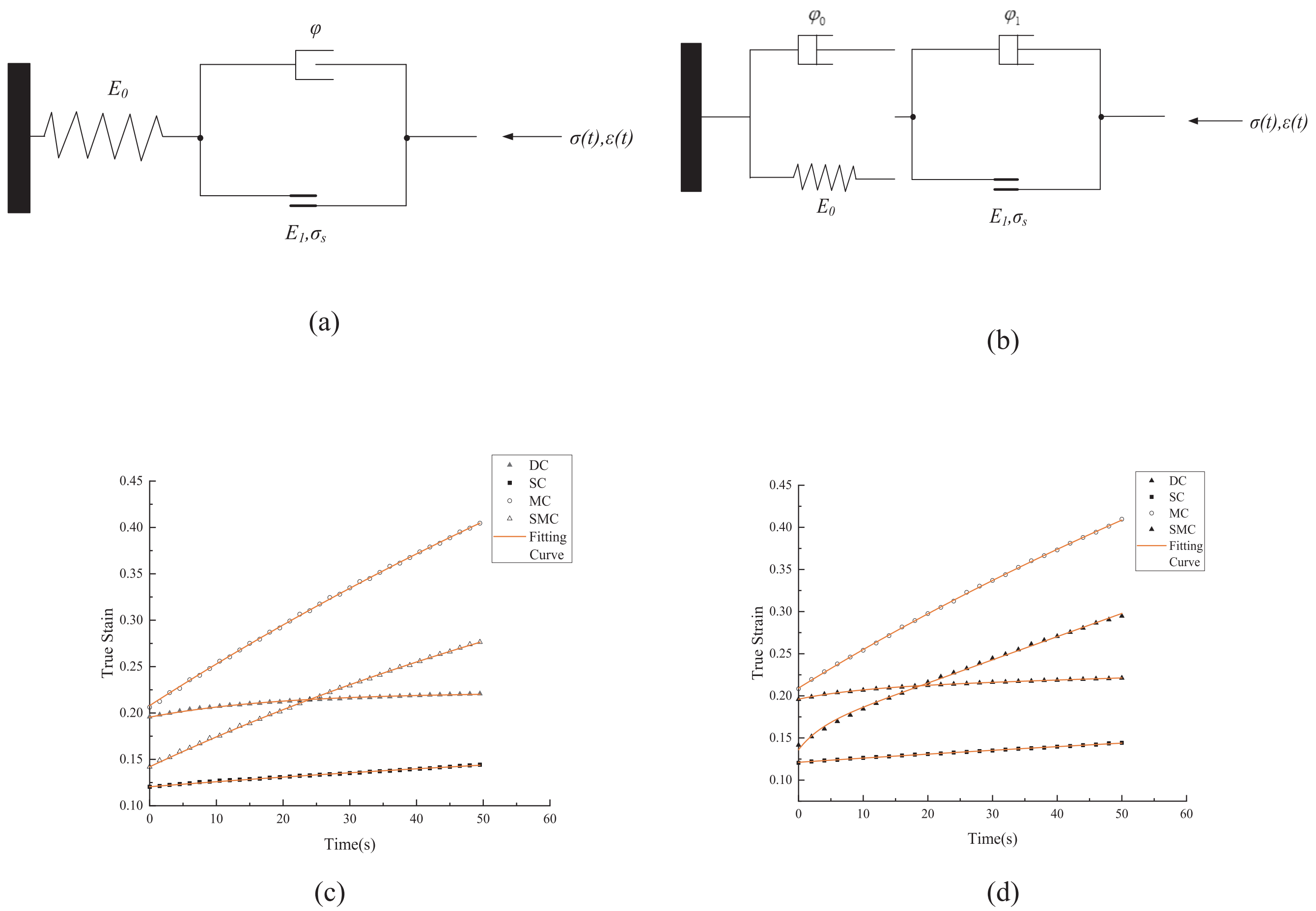


Fig. 7　Building a constitutive structure model.
(a) Constitutive model 1-Bingham model
(b) Constitutive model 2
(c) Fitting curve of instantiation model 1 -Bingham model
(d) Fitting curve of instantiation model 2

periments. This model can simulate the mechanical properties of chocolate well, but more sophisticated models are needed to provide predictive power beyond a given set of mechanical test conditions.

The above experiments and the results of the fitting of the constitutive model were used to construct functions between the necessary parameters and the amount of sugar and milk powder added in order to predict the mechanical behaviour of the chocolate in different ratios, which can be expressed as Eqn (16) (17).

$$\left.\begin{aligned} E_0 &= 0.35x_1 - 0.25x_2 + 3.10 \\ \phi_0 &= -2.11x_1 - 48.11x_2 + 390.93 \\ \phi_1 &= 19.75x_1 + 33.81x_2 + 26.82 \end{aligned}\right\} \quad (16)$$

$$\varepsilon = \frac{\sigma_A}{0.35x_1 - 0.25x_2 + 3.10}\left(1 - e^{-\frac{0.35x_1 - 0.25x_2 + 3.10}{-2.11x_1 - 48.11x_2 + 390.93}t}\right) + \frac{\sigma_A - \sigma_s}{19.75x_1 + 33.81x_2 + 26.82}t + A \quad (17)$$

Where, x_1 is amount of sugar added, x_2 is amount of milk powder added.

However, the test volumes in this trial were small and the predictions in this respect were poor, and subsequent trials with a large number of modifications to the sugar and dairy powder additions will be required to correct them. It is also designed to find the optimum amount of sugar and dairy powder to be added by the optimal shape method.

4 Conclusion

This study investigated the mechanical properties of chocolate as influenced by two common ingredients, sugar and milk powder, and attempted to relate the properties of different ratios of chocolate to its mechanical behaviour at the first bite. The effect of the addition of the two ingredients on the textural properties of the chocolate was determined.

Table 3 Material contents for constitutive model 1 for all chocolate materials.

Chocolate Sample	E_0 (Mpa)	σ_s (Mpa)	B	ϕ	R^2
DC	2.667	0.430	0.910	165.0	0.9934
SC	4.315	0.450	1.190	156.7	0.9970
MC	2.533	0.270	0.340	159.5	0.9997
SMC	3.662	0.320	0.540	153.2	0.9996

E_0 is the modulus of elasticity of the spring element in the intrinsic model 1, B is the reinforcement factor, σ_s is yield stress, ϕ is the parameter in the damping element and R^2 quantifies the goodness of fit between the values 0 and 1. The closer the value is to 1, the better the curve fit is.

Table 4 Material contents for constitutive model 2 for all chocolate materials.

Chocolate Sample	E_0 (Mpa)	σ_s (Mpa)	ϕ_0	ϕ_1	R^2
DC	3.147	0.500	379.9	77.63	0.9994
SC	4.808	0.540	391.4	74.76	0.9996
MC	1.808	0.300	161.4	145.1	0.9998
SMC	3.647	0.520	128.8	345.4	0.9993

E_0 is the modulus of elasticity of the spring element in the intrinsic model 2, σ_s is yield stress, ϕ_0, ϕ_1 is the parameter in the damping element and R^2 quantifies the goodness of fit between the values 0 and 1.The closer the value is to 1, the better the curve fit is.

The fracture behaviour of chocolate and mechanical properties changed when two common ingredients, sugar and milk powder, were added to the chocolate. In this study, the addition of powdered sugar resulted in enhanced material properties such as Young's modulus, yield stress, fracture toughness, and fracture load of chocolate. In contrast, milk powder had the opposite effect. Sugar played a more significant role in material properties in equal proportions of both together. The decrease in chocolate fracture stress may affect the perceived hardness of chocolate. In contrast, chocolate exhibited a high degree of rate dependence in the fracture experiments, indicating that different chewing rates significantly affect perceived hardness and brittleness of chocolate. All materials were subjected to cyclic loading tests to observe the elasticity changes of different chocolates by their deformation recovery. Although the addition of powdered sugar and porous structure increases the resistance of chocolate to deformation, the addition of milk powder destabilizes the fat crystals to a certain extent and makes the chocolate more susceptible to permanent deformation.

A four-component viscoelastic-plastic model based on Bingham's model can better explain the mechanical response of the chocolate in the test, which was corrected by the creep test data, and the parameters obtained confirmed the mechanical properties of the four different chocolates in the test. The model can be used to fit future test data for more ratios of chocolate to predict the texture properties of chocolate in different ratios, providing a suitable tool for the design and production of food products with specified sensory properties.

Funding

This study was supported by Jiangsu Key Laboratory of Advanced Food Manufacturing Equipment and Technology (FMZ201901), National Natural Science Foundation of China (51775244) and Jiangsu Key Laboratory of Advanced Food Manufacturing Equipment and Technology (FMZ201907).

Conflicts of Interest/Competing Interests

The authors declared that there is no conflict of interest.

References

1) Li, Y.; James, B. Oral processing preference affects flavor perception in dark chocolate with added ingredients. *J. Food Sci.* **86**, 177-183 (2021). doi:101111/1750-3841.15557

2) Bresson, S.; Lecuelle, A.; Bougrioua, F.; Hadri, M.; Baeten, V. Comparative structural and vibrational investigations between cocoa butter (CB) and cocoa butter equivalent (CBE) by ESI/MALDI-HRMS, XRD, DSC,

MIR and Raman spectroscopy. *Food Chem.* **363**, 130319(2021). doi:j.foodchem.2021.130319

3) Afoakwa, E.O.; Paterson, A.; Fowler, M. Factors influencing rheological and textural qualities in chocolate – a review. *Trends in Food Sci. Technol.* **18**, 290-298 (2007). doi:10.1016/j.tifs.2007.02.002

4) Dhonsi, D.; Stapley, A.G.F. The effect of shear rate, temperature, sugar and emulsifier on the tempering of cocoa butter. *J. Food Eng.* **77**, 936-942(2006). doi:10.1016/ j.jfoodeng.2005.08.022

5) Svanberg, L.; Ahrné, L.; Lorén, N.; Windhab, E. Effect of sugar, cocoa particles and lecithin on cocoa butter crystallisation in seeded and non-seeded chocolate model systems. *J. Food Eng.* **104**, 70-80(2011). doi:10.1016/j.jfoodeng.2010.09.023

6) Attaie, H.; Breitschuh, B.; Braun, P.; Windhab, E.J. The functionality of milk powder and its relationship to chocolate mass processing, in particular the effect of milk powder manufacturing and composition on the physical properties of chocolate masses. *Int. J. Food Sci. Technol.* **38**, 325-335(2003). doi:10.1046/j.1365-2621.2003.00678.x

7) Afoakwa, E.O.; Paterson, A.; Fowler, M.; Vieira, J. Microstructure and mechanical properties related to particle size distribution and composition in dark chocolate. *Int. J. Food Sci. Technol.* **44**, 111-119(2009). doi:10.1111/j.1365-2621.2007.01677.x

8) Glicerina, V.; Balestra, F.; Rosa, M.D.; Romani, S. Rheological, textural and calorimetric modifications of dark chocolate during process. *J. Food Eng.* **119**, 173-179 (2013). doi:10.1016/j.jfoodeng.2013.05.012

9) Stewart, A.; Grandison, A.S.; Ryan, A.; Festring, D.; Methven, L. Impact of the skim milk powder manufacturing process on the flavor of model white chocolate. *J. Agric. Food Chem.* **65**, 1186-1195(2017). doi:10.1021/acs.jafc.6b04489

10) Belščak-Cvitanović, A.; Komes, D.; Dujmović, M.; Karlović, S.; Biškić, M. Physical, bioactive and sensory quality parameters of reduced sugar chocolates formulated with natural sweeteners as sucrose alternatives. *Food Chem.* **167**, 61-70(2015). doi:10.1016/ j.foodchem.2014.06.064

11) Sokmen, A.; Gunes, G. Influence of some bulk sweeteners on rheological properties of chocolate. *LWT-Food Sci. Technol.* **39**, 1053-1058(2006). doi:10.1016/ j.lwt.2006.03.002

12) Le Révérend, B.; Saucy, F.; Moser, M.; Loret, C. Adaptation of mastication mechanics and eating behaviour to small differences in food texture. *Physiol. Behav.* **165**, 136-145(2016). doi:10.1016/j.physbeh.2016.07.010

13) Bikos, D.; Samaras, G.; Cann, P.; Masen, M.; Hardalupas, Y. Effect of micro-aeration on the mechanical behaviour of chocolates and implications for oral processing. *Food Funct.* **12**, 4864-4886(2021). doi:10.1039/D1FO00045D

14) Marie, K.; Hiroshi, S. Changes in mandibular movement during chewing of different hardness foods. *Odontology* **105**, 418-425(2017). doi:10.1007/S10266-016-0292-z

15) Lucas, P.W.; Copes, L.; Constantino, P.J.; Vogel, E.R.; Chalk, J. Measuring the toughness of primate foods and its ecological value. *Int. J. Primatol.* **33**, 598-610 (2012). doi:10.1007/s10764-011-9540-9

16) Zink, K.D.; Lieberman, D.E.; Lucas, P.W. Food material properties and early hominin processing techniques. *J. Hum. Evol.* **77**, 155-166(2014). doi:10.1016/j.jhevol.2014.06.012

17) McCarthy, C.T.; Hussey, M.; Gilchrist, M.D. On the sharpness of straight edge blades in cutting soft solids: Part I – indentation experiments. *Eng. Fract. Mech.* **74**, 2205-2224(2007). doi:10.1016/j.engfracmech.2006.10.015

18) Kim, E.H.-J.; Corrigan, V.K.; Wilson, A.J.; Waters, I.R.; Hedderley, D.I. Fundamental fracture properties associated with sensory hardness of brittle solid foods: fracture and texture of brittle solid foods. *J. Texture Stud.* **43**, 49-62(2012). doi:10.1111/j.1745-4603.2011.00316.x

19) Pascua, Y.; Koç, H.; Foegeding, E.A. Food structure: Roles of mechanical properties and oral processing in determining sensory texture of soft materials. *Curr. Opin. Colloid Interface Sci.* **18**, 324-333(2013). doi:10.1016/j.cocis.2013.03.009

20) Tunick, M.H.; Onwulata, C.I.; Thomas, A.E.; Phillips, J.G.; Mukhopadhyay, S. Critical evaluation of crispy and crunchy textures: A review. *Int. J. Food Prop.* **16**, 949-963(2013). doi:10.1080./10942912.2011.573116

21) Witt, T.; Stokes, J.R. Physics of food structure breakdown and bolus formation during oral processing of hard and soft solids. *Curr. Opin. Food Sci.* **3**, 110-117 (2015). doi:10.1016/j.cofs.2015.06.011

22) Afoakwa, E.O.; Paterson, A.; Fowler, M.; Vieira, J. Relationship between rheological, textural and melting properties of dark chocolate as influenced by particle size distribution and composition. *Eur. Food Res. Technol.* **227**, 1215-1223(2008). doi:10.1007/s00217-008-0839-5

23) Liang, B.; Hartel, R.W. Effects of milk powders in milk chocolate. *J. Dairy Sci.* **87**, 20-31(2004). doi:10.3168/jds.S0022-0302(04)73137-9

24) Afoakwa, E.O.; Paterson, A.; Fowler, M. Factors influencing rheological and textural qualities in chocolate-a review. *Trends Food Sci. Technol.* **18**, 290-298 (2007). doi:10.1016/j.tifs.2007.02.002

25) Gonzalez-Gutierrez, J.; Scanlon, M.G. Rheology and Mechanical Properties of Fats. in *Structure-Function Analysis of Edible Fats*. AOCS Press, pp. 19-168

(2018). doi:10.1016/B978-0-12-814041-3.00005-8

26) Dasgupta, A.; Hu, J.M. Failure mechanism models for plastic deformation. *IEEE Trans. Reliab.* **41**, 168-174 (1992). doi:10.1109/24.257775

27) Beckett, B.Sc.; Stephen, T. *Industrial Chocolate Manufacture and Use*. Wiley-Blackwell, Oxford, UK (2008). doi:10.1002/9781444301588

28) Lohman, M.H.; Hartel, R.W. Effect of milk fat fractions on fat bloom in dark chocolate. *J. Am. Oil Chem. Soc.* **71**, 267-276 (1994). doi:10.1007/BF02638052

29) Bricknell, J.; Hartel, R.W. Relation of fat bloom in chocolate to polymorphic transition of cocoa butter. *J. Am. Oil Chem. Soc.* **75**, 1609-1615 (1998). doi:10.1007/s11746-998-0101-0

30) Vincent, J.F.V. Application of fracture mechanics to the texture of food. *Eng. Failure Anal.* **11**, 695-704 (2004). doi:10.1016/j.engfailanal.2003.11.003

31) Pilkey, W.D.; Pilkey, D.F. *Peterson's Stress Concentration Factors*. John Wiley & Sons, Inc. Hoboken, NJ, USA (2007). doi:10.1002/9780470211106

32) Lannes, S.C.da S. *Cheese rheology and texture. Rev. Bras. Cienc. Farm.* **40** (2) (2004). doi:10.1590/S1516-93322004000200018

33) Lazaro, L.M.; Aranda, D.A.G. Process temperature profile and rheological properties of greases from vegetable oils. *Green Sustainable Chem.* **04** (01), 38-43 (2014). doi:10.4236/gsc.2014.41007