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Crunchiness Loss and Moisture Toughening in Puffed Cereals and Snacks

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Abstract: Upon moisture uptake, dry cellular cereals and snacks loose their brittleness and become soggy. This familiar phenomenon is manifested in smoothing their compressive force–displacement curves. These curves' degree of jaggedness, expressed by their apparent fractal dimension, can serve as an instrumental measure of the particles' crunchiness. The relationship between the apparent fractal dimension and moisture content or water activity has a characteristic sigmoid shape. The relationship between the sensorily perceived crunchiness and moisture also has a sigmoid shape whose inflection point lies at about the same location. The transition between the brittle and soggy states, however, appears sharper in the apparent fractal dimension compared with moisture plot. Less familiar is the observation that at moderate levels of moisture content, while the particles' crunchiness is being lost, their stiffness actually rises, a phenomenon that can be dubbed "moisture toughening." We show this phenomenon in commercial Peanut Butter Crunch® (sweet starch-based cereal), Cheese Balls (salty starch-based snack), and Pork Rind also known as "Chicharon" (salty deep-fried pork skin), 3 crunchy foods that have very different chemical composition. We also show that in the first 2 foods, moisture toughening was perceived sensorily as increased "hardness." We have concluded that the partial plasticization, which caused the brittleness loss, also inhibited failure propagation, which allowed the solid matrix to sustain higher stresses. This can explain other published reports of the phenomenon in different foods and model systems.

Keywords: brittleness, crispness, failure propagation, glass transition, moisture sorption, plasticization, water activity

Introduction

Brittle breakfast cereals and snacks owe much of their universal appeal to their crunchiness, the generation of satisfying acoustic and other pleasant sensations in the mouth. (The word "crunch," frequently associated with "crush," probably originated in an attempt to imitate the produced sound.) When a dry cereal or snack absorbs moisture it loses its crunchiness, or crispness, and becomes soggy. This familiar phenomenon has been documented in the celebrated paper of Katz and Labuza (1981) who reported an inverse linear relation between sensorily perceived crispness and water activity. Later works have shown that when the entire range of water activities is taken into account, the relationship between crunchiness or crispness and water activity or moisture contents has a characteristic sigmoid shape (for example, Peleg 1994; Roudaut and others 2002).

Since the early 1990s, the plasticization of brittle dry foods by moisture sorption, or its inverse the hardening of plasticized food by moisture loss during drying, was explained in terms of glass-transition theories originally developed for synthetic rubbery polymers (Slade and others 1991; Roos 1995). According to these theories, still in vogue, water being an effective plasticizer of hydrophilic biopolymers lowers these polymers' otherwise high glass-transition temperature, "T_g," causing them to undergo a transition from a glassy to rubbery state at ambient temperature. Obviously, wet cereals and snacks are not "rubbery" and do not bounce back when hitting a hard surface. The term "soggy" seems to describe their wet state better than the one imported from the polymer science literature. It has also been claimed in several publications that the moisture effect on a food's T_g (and hence texture) could be described and perhaps even predicted by the Gordon–Taylor equation, originally used to estimate the T_g of compatible polymer blends from the T_g's of their 2 components. More complicated models such as the Couchman–Karasz equation, which also takes into account the 2 polymers' heat capacity, have also been proposed for this purpose. Although the term "T_g" is still widely used in polymer science, food science, and other fields, the concept that there exists a unique glass-transition temperature has been largely discredited even in the polymer literature (for example, Seyler 1994; Ryan 2001; Langer 2007; Brostow and others 2008). It is now well accepted that the transition occurs over a temperature range rather at a point (Peleg 1993; Roos 2001) and the same applies to the moisture content. It has also been recognized that the rate and method of monitoring the transition can have a substantial effect on the determined value of T_g. Moreover, the frequently cited glass-transition temperature of the highly unstable glassy water (~136 K), whose relevance to dry foods plasticization at ambient and higher temperatures is yet to be satisfactorily explained, might need a revision (for example, Velikov and others 2001). Despite the shortcomings of the so-called "polymer science approach" to food texture and rheology, the transition between brittle and soggy (not rubbery!) state in crunchy foods is obviously real and has important practical implications in their formulation, processing, and packaging.

A related but less familiar effect of moisture on brittle foods can be dubbed "*moisture toughening*," a counter-intuitive physical

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phenomenon where absorbed water, a most potent plasticizer, can actually *raise* the food's stiffness over a substantial range of moisture contents. This phenomenon was first reported in puffed cereals by Meg Harris (Harris and Peleg 1996) and is the topic of this work. Its focus will be on studies performed at the University of Massachusetts in the mid and late 1990s where we demonstrated that moisture toughening is not only detectible by mechanical instruments but can also be perceived sensorily by panelists.

Definitions

When it comes to textural attributes, there is frequently a mismatch between the sensory vocabulary and the terms used in mechanics and material science. We will use the following mechanical terms in this article.

Brittleness

The tendency of a material to fracture or shatter after very small deformation, frequently accompanied by audible acoustic emission. A dry food material failing in this mode is commonly perceived as crunchy or crispy.

Compressive mechanical signature

A record of the force–displacement or force–time relationship produced by a specimen subjected to uniaxial compression (pre- and postfailure).

Stiffness

A measure of a specimen's resistance to deformation. It is expressed as a modulus, the slope of stress–strain curve prior to reaching the failure region and has pressure units. Stiffness is frequently perceived as hardness in foods.

Toughness

The mechanical energy absorbed by a deforming specimen prior to its failure. It is manifested in the area under the stress–strain curve, has work per unit volume units, and is probably also perceived as hardness or perhaps chewiness in some foods.

Mechanical Characterization of Dry Puffed Cereals and Snacks Tested Intact

Stiffness and toughness

Solid foods are frequently tested in uniaxial compression using a Universal Testing Machine operated at a relatively low constant displacement rate (speed). In many solid foods, preparation of cylindrical specimens is rather simple, and testing them in compression eliminates the problem of providing proper grip that besets tensile tests, which are more common in polymer science. The 2 main ways to prepare a cylindrical specimen for a compression test are boring, as in cheeses or fruits flesh, or forming in a cylindrical mold, as in gels. Unfortunately, neither method is a practical option when it comes to fragile cellular cereals and snacks. Such particulates, therefore, have to be tested individually and *intact* if of sufficiently uniform size and shape, or *in bulk* when they are not—see below. When the compressed cellular specimen is spherical or has an irregular shape, conversion of the testing machine's force–displacement record into a meaningful stress–strain relationship is extremely difficult if not utterly impossible. To complicate matters further, the recorded compressive mechanical signatures of brittle puffed particulates, are both irregular and irreproducible as demonstrated in Figure 1. Therefore, instead of 'stiffness' properly expressed as a modulus, which is hard to determine, we will use 2 empirical "*stiffness measures*" instead. To determine them, we fitted the digital force–displacement data with a polynomial model as shown in the figure and calculate the force at 2 arbitrary displacement levels, for example, 15% and 25%, of the original diameter or 25% and 35% of the particles bed's depth. The reason for using 2 displacement levels instead of 1 is to assure that whatever conclusion we derive concerning the effect of moisture on a cereal's or snack's stiffness is independent of the chosen displacement

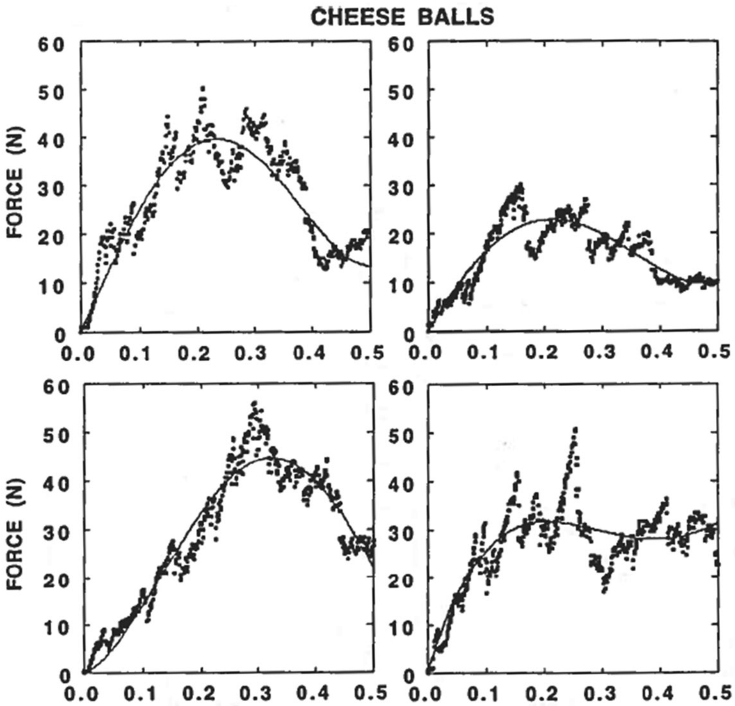

Figure 1–Examples of irregular and irreproducible mechanical signatures of Cheese Balls fitted with a fourth degree polynomial model to calculate the stiffness measures.

level. The area under the force–displacement curve, regardless of whether it is smooth or jagged, primarily depends on the forces level. Consequently an increase or decrease of the stiffness parameters can also be viewed as representing a corresponding increase or decrease in toughness.

Brittleness

When an individual particle of a brittle cellular cereal or snack is compressed, fracture of cell walls and structural collapse commences almost at once and continues even after the original structure is destroyed. The many uneven sharp force drops in the mechanical signature are most probably a visual record of fracture events of different magnitudes. The resulting discontinuous fluctuating force pattern can be characterized by its *apparent fractal dimension*, which serves as measure of its *jaggedness*. A curve's jaggedness scale is from 1.0 (perfectly smooth curve—the Euclidian dimension of a line is 1.0 to 2.0 (the upper theoretical limit of curves so convoluted and dense that they almost fill the area on which they are plotted (the Euclidian dimension of an area is 2.0. It should be made clear that a mechanical signature obtained from a testing machine is not a true fractal object. Because the force is recorded at fixed time intervals, the force–time or force–displacement curve should be considered as self-affine, that is, it exhibits self-similarity only in the force direction. Also, because the force is recorded at a finite resolution, self-similarity can only exist over a limited number of length scales. The visual

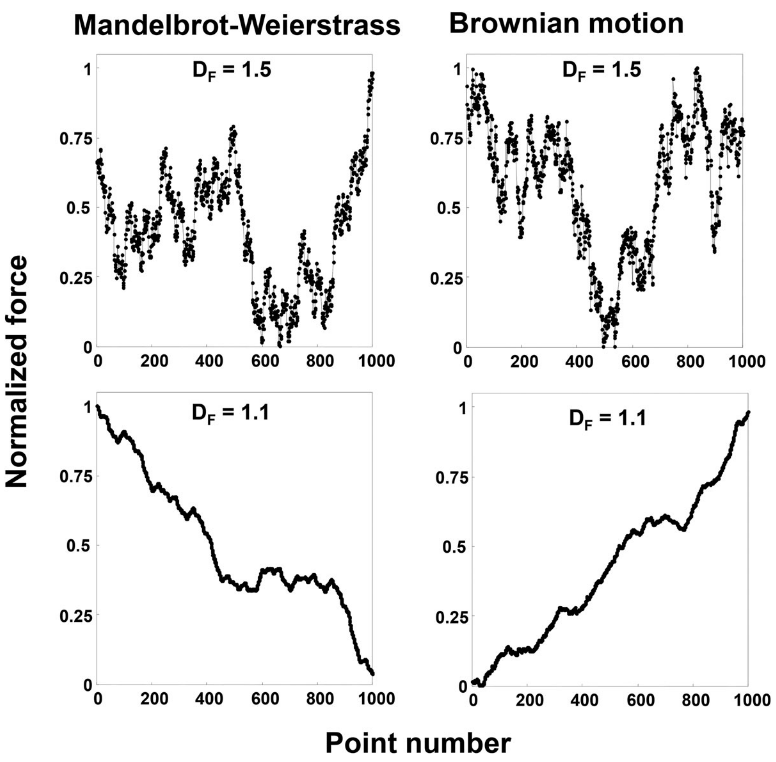

Figure 2–Simulated irregular mechanical signatures curves having 2 degrees of jaggedness produced with 2 different algorithms (left and right). Notice that the apparent fractal dimension is insensitive to the curve's general shape.

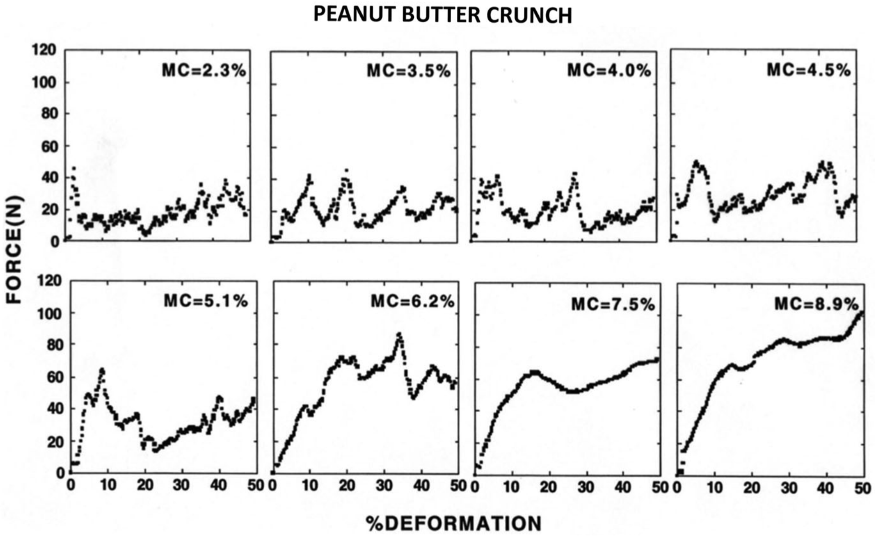

Figure 3–Examples of the mechanical signatures of Peanut Butter Crunch particles having different moisture contents. Notice that the curves where the moisture content was 6.2% to 7.5% are is higher than those where the moisture content was 2.3% to 4.5%. Adapted from Suwonsichon and Peleg (1998).

jaggedness of a machine-created signature depends on both the fluctuating force amplitude and the sampling rate. For a demonstration of the interrelation between fractal dimension, resolution and the visual appearance of jagged curves open http://demonstrations.wolfram.com/LineJaggednessVisualizationWithTheMandelbrotWeierstrassFunct/ and move the control sliders to modify the image. (To download the free Wolfram CDF Player that runs the Demonstration, and over 10000 others to date, follow instructions on the screen.) Despite the above-mentioned limitations, the apparent fractal is a most convenient measure of jaggedness for comparing signatures of similar length recorded at the same resolution (Barrett and others 1992). This is because the determined apparent fractal dimension is insensitive to the signature's overall shape and morphological details, which can

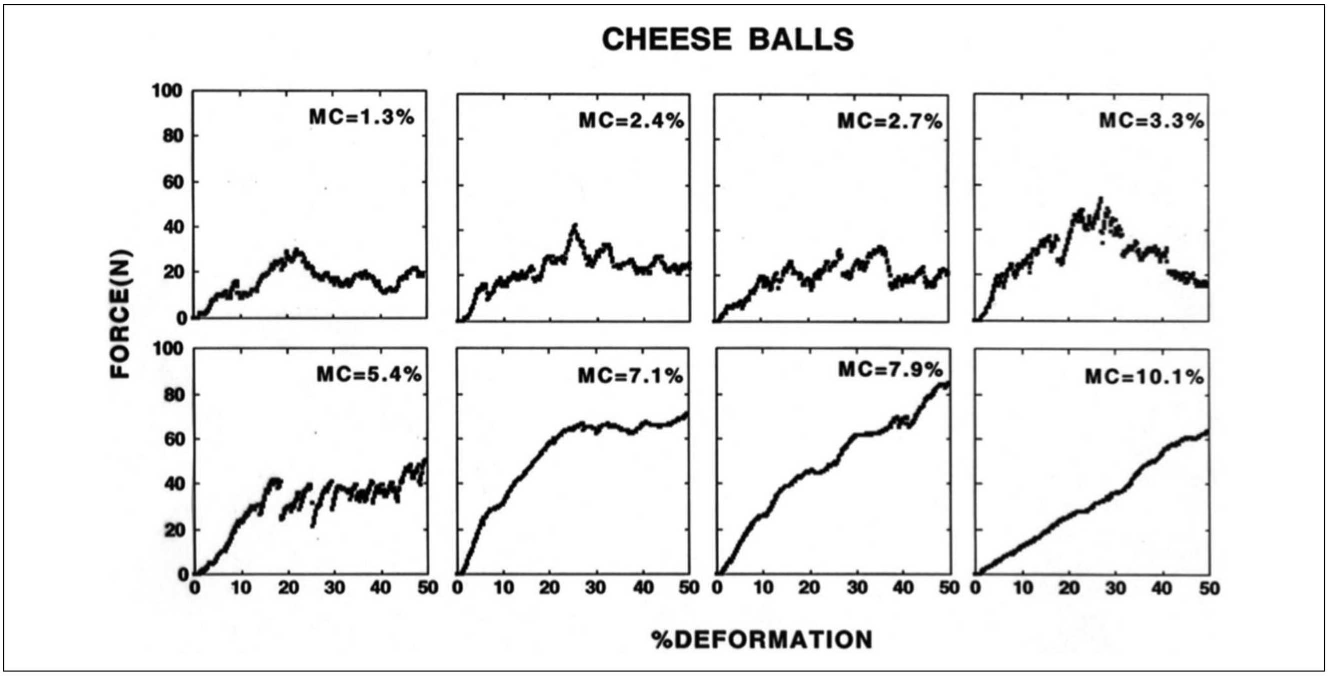

Figure 4–Examples of the mechanical signatures of Cheese Balls having different moisture contents. Notice that the curves where the moisture content was 7.1% to 7.9% are higher than those where the moisture content was 1.3% to 2.7%. Adapted from Suwonsichon and Peleg (1998).

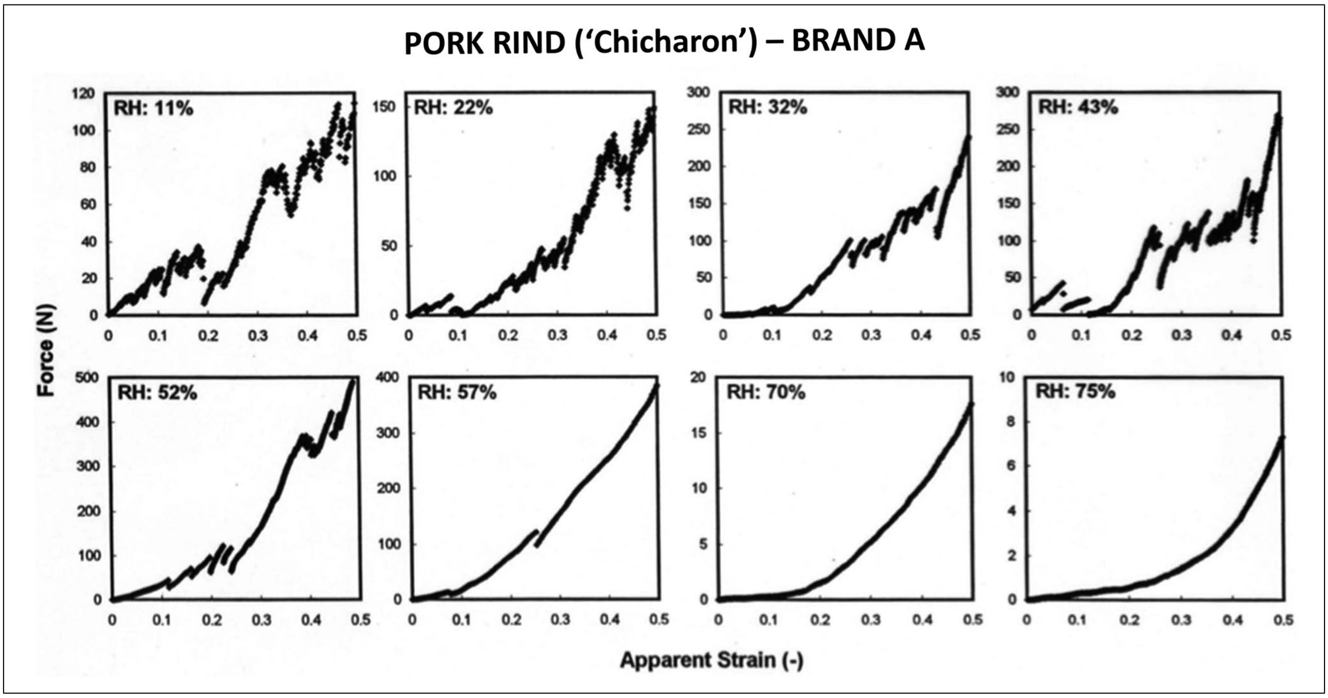

Figure 5–Examples of the mechanical signatures of Pork Rinds ("Chicharon") at different relative humidity levels tested in bulk. Notice that despite the fluctuations' amplitude suppression by the averaging effect, the loss of jaggedness is still clearly visible. Also notice that the curves of the particles stored at 32% to 52% RH are higher than those of the particles stored at 11% and 22% RH. Adapted from Gonzalez Martinez and others (2003).

vary dramatically among the signatures of same cereal and snack at the same moisture contents. For a demonstration of the fractal dimension's insensitivity to morphological details see Figure 2. It shows that jagged curves of very different overall shapes and produced by different algorithms can have the same fractal dimension in agreement with visual appearance. Another advantage of the apparent fractal dimension as a jaggedness measure is that it is easy to determine directly from digital data with suitable software such as that of Russ (1994). As with the stiffness measures, we have used 2 algorithms to determine the apparent fractal dimensions, Richardson's (the "compass method") and Kolmogorov's (the "box counting method"), for mutual verification. (For visual comparison of the apparent fractal dimensions with jaggedness measures based on the scatter's standard deviation open http://demonstrations.wolfram.com/ComparingMeasuresOfLineJaggedness/).

Testing particulates in bulk

For obvious reasons, when the individual particles vary dramatically in size and shape as in pork rind ("Chicharon") – see below – testing them singly is not a practical option. The alternative is to compress a shallow layer of them in a cell or several cells of different diameters. The particles' stiffness in this case would still be manifested in the fitted force at a chosen displacement, or 2 displacements, with or without adjustment for the cell's cross-sectional area (Gonzalez Martinez and others 2003). In this form of testing, the mechanical signature's jaggedness would be smaller than that of the individual particles because of an averaging effect (Ulbricht and others 1994). In principle and in practice, one can estimate the original particle signature's jaggedness from bulk measurements (Suwonsichon and others 1997). This, however, was unnecessary in our case because we were only interested in relative changes caused by moisture sorption. (The averaging effect and the principle of the jaggedness restoration can be viewed in http://demonstrations.wolfram.com/NoiseRetrievalFromAveragedSequences/.)

Effect of Moisture

Many studies of the effect of moisture contents or water activity on the texture of dry brittle foods (for example, Vickers and

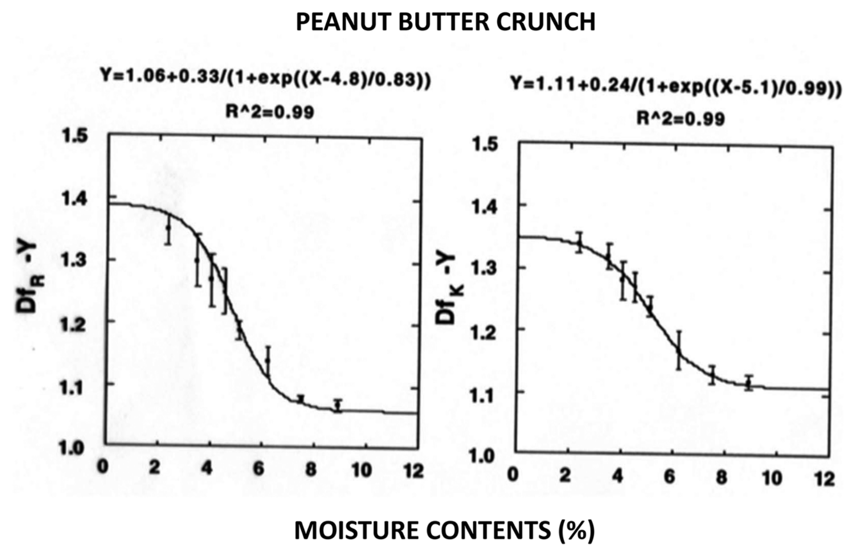

Figure 6–Plots of the apparent fractal dimension (Df) of the mechanical signatures of Peanut Butter Crunch particles at different moisture contents. Df_R (left) determined with the Richardson ('compass') algorithm, Df_K (right) with the Kolmogorov's box counting algorithm. Adapted from Suwonsichon and Peleg (1998).

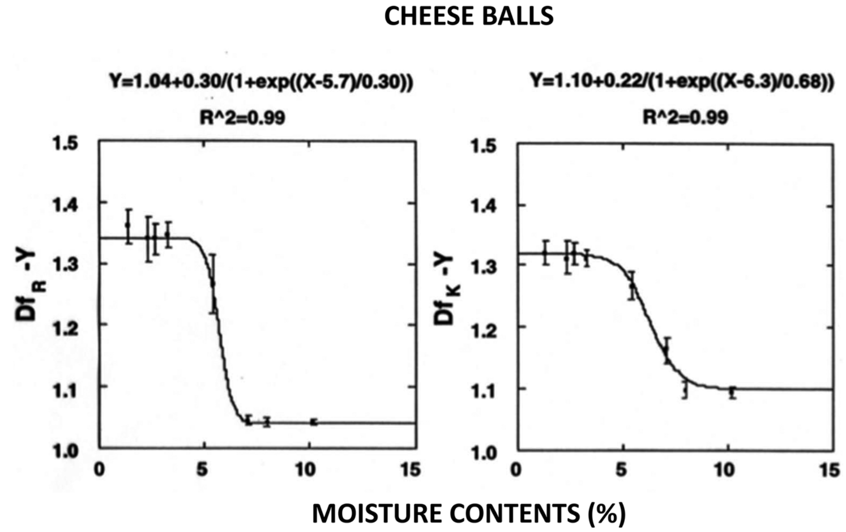

Figure 7–Plots of the apparent fractal dimension (Df) of the mechanical signatures of Cheese Balls at different moisture contents. Df_R (left) determined with the Richardson ('compass') algorithm, Df_K (right) with the Kolmogorov's box counting algorithm. Adapted from Suwonsichon and Peleg (1998).

Bourne 1976; Tesch and others 1995; Norton and others 1998; Valera and others 2009) indicate that the complexity of the acoustic signature, jaggedness of the mechanical signature and perceived crunchiness are closely related. This strongly suggests, albeit does not prove, that the fracture events recorded as force drops in the mechanical signature are those that produce the acoustic emissions that produce the crunchiness sensation. In other words, it is not unreasonable to treat the mechanical signature's degree of jaggedness as representing the specimen's brittleness and crunchiness, a notion to which we'll adhere.

Examples of the mechanical signatures (force–displacement curves) of individual particles of the starchy sweet cereal Peanut

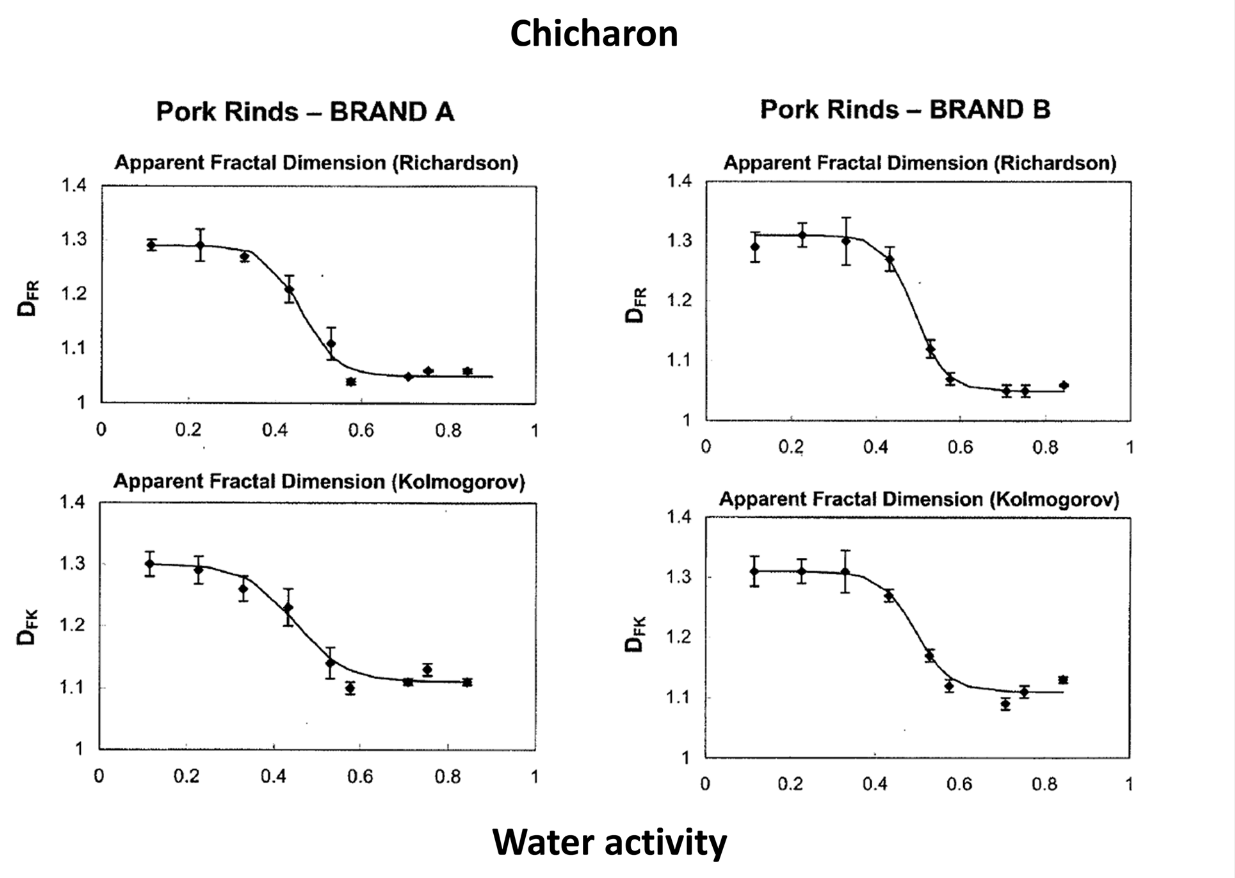

Figure 8–Plots of the apparent fractal dimension (D_F) of the mechanical signatures of Pork Rinds of 2 brands at different water activity levels tested in bulk. D_{FR} (top) determined with the Richardson ("compass") algorithm, D_{FK} (bottom) with Kolmogorov's box counting algorithm. Adapted from Gonzalez Martinez and others (2003).

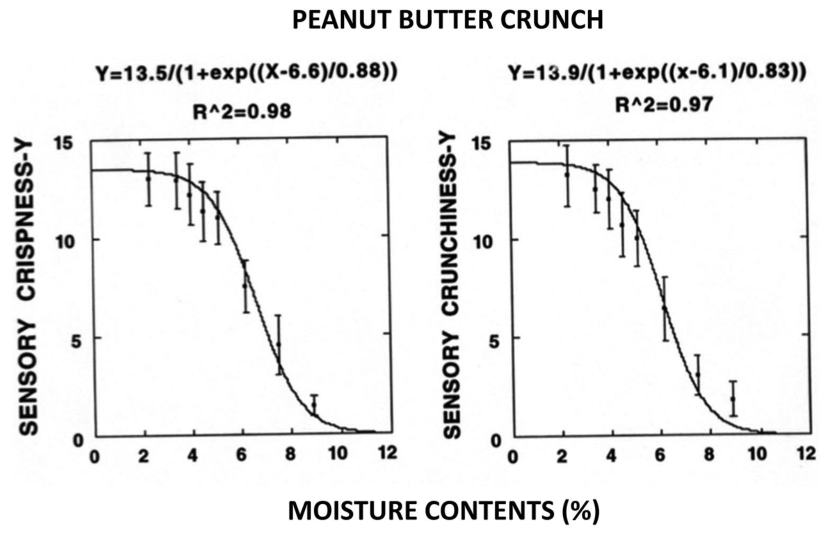

Figure 9–Plots of the sensory crunchiness/crispness evaluations of Peanut Butter Crunch at different moisture contents. Compare to Figure 6. Adapted from Suwonsichon and Peleg (1998).

Butter Crunch®, recorded at different moisture content are shown in Figure 3. Mechanical signatures of the puffed salty, starchy cheese flavored snack known as Cheese Balls, also tested individually at various moisture contents levels, are shown in Figure 4. Since the particles of both the Peanut Butter Crunch and Cheese Balls are approximately spherical and their size is relatively large and uniform, they could be compressed individually without any special mounting or preparation. Details of the equilibration and testing procedures can be found in Suwonsichon and Peleg (1998). Pork Rind ("Chicharon") is a curly salty snack made of deep-fried seasoned pork skin. The pieces are about 3 to 5 cm in length and 1 to 2 cm thick and their shape vary dramatically. Examples of the force–displacement curves of these particles tested in bulk at different relative humidity/water activity levels are shown in

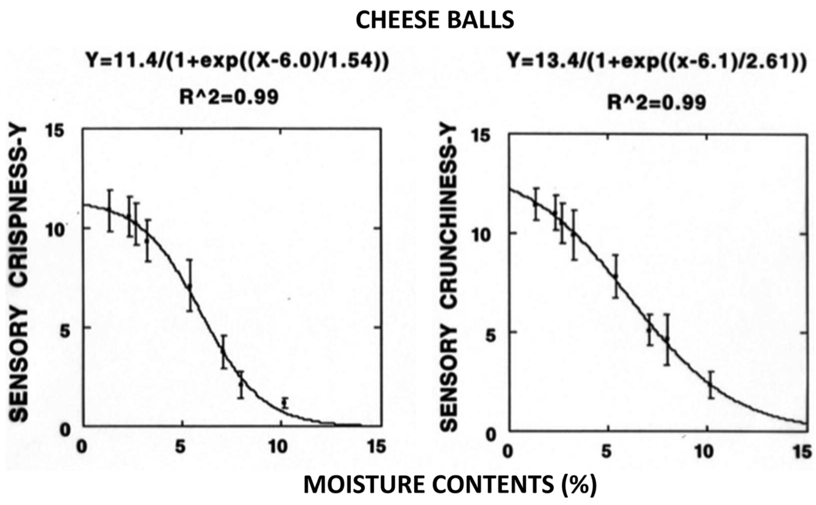

Figure 10–Plots of the sensory crunchiness/crispness evaluations of Cheese Balls at different moisture contents. Compare to Figure 7. Adapted from Suwonsichon and Peleg (1998).

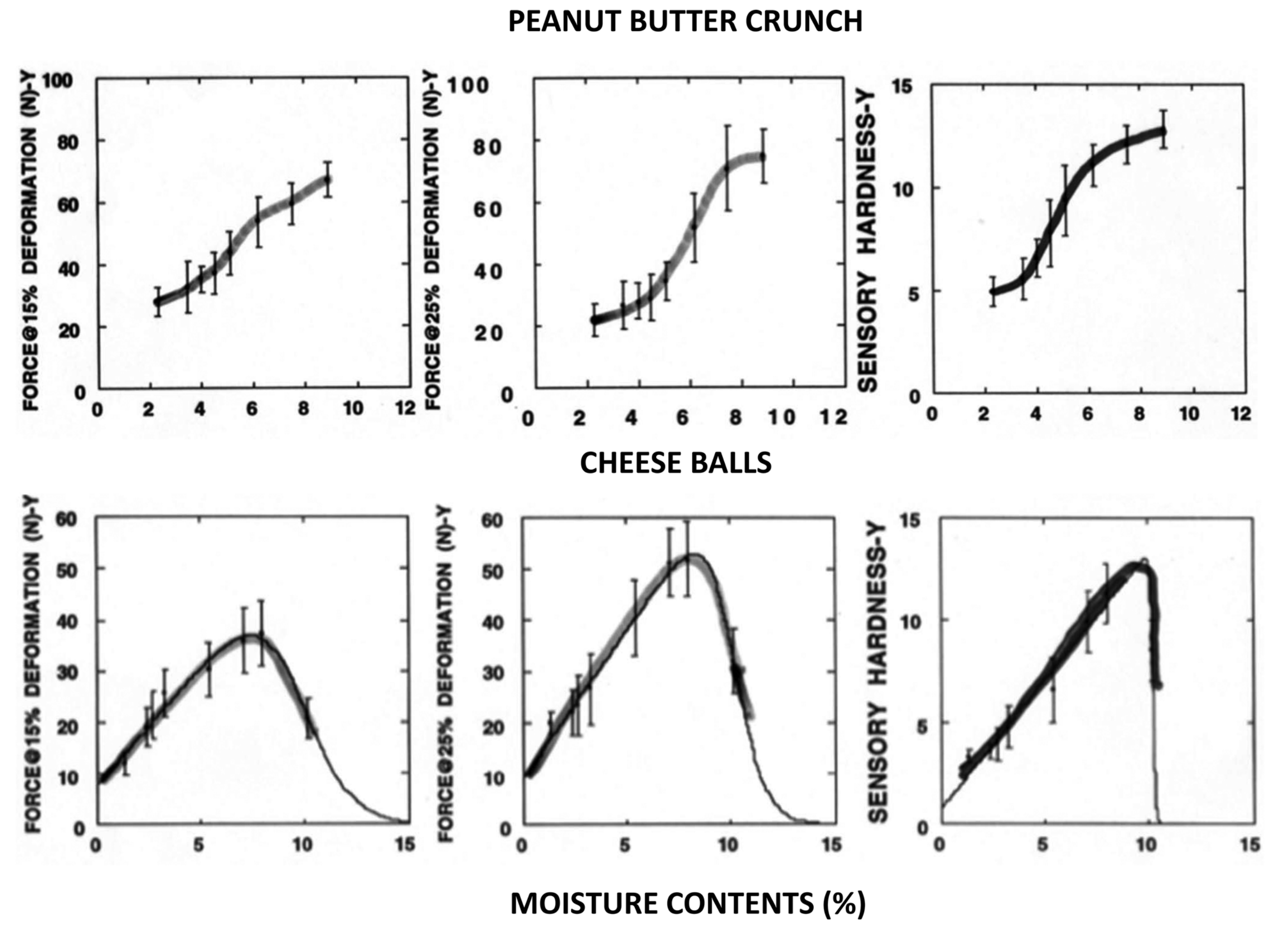

Figure 11–Moisture toughening of Peanut Butter Crunch particles (top) and Cheese Balls (bottom) monitored instrumentally (left and middle), and evaluated sensorily as increase followed by decrease of hardness (right). Adapted from Suwonsichon and Peleg (1998).

Figure 5. Having grossly non-uniform shape and size, pieces were placed as a shallow layer in a wide metal cell in which they were compressed together as a single specimen. Details of the procedure can be found in Gonzalez Martinez and others (2003).

Effect of moisture on the mechanical signature's jaggedness

The 3 examples given in Figure 3 to 5 all show the same trend: As the moisture contents or water activity increased, the mechanical signature became visibly smoother and vice versa. Plots of the mechanical signatures apparent fractal dimension compared with moisture contents or relative humidity are shown in Figure 6 to 8. In all 3 cases, and despite the individual signatures' irreproducibility, their mean apparent fractal dimension, regardless of whether determined by the Richardson (Df_R) or Kolomogorov (Df_K) algorithm, describes a remarkably discernible sigmoid curve when plotted against moisture content or relative humidity (Wollny and Peleg 1994). Very similar sigmoid curves describe the sensory evaluations of the Peanut Butter Crunch's and cheese ball's crunchiness and crispness as shown in Figure 9 and 10, in agreement with those reported by Roudaut and others (2002).

The repeated observation of a sigmoid relation between the mechanical signature's jaggedness and moisture contents and a corresponding sigmoid relationship between sensorily perceived crunchiness and moisture lends support to the notion that the apparent fractal dimension can indeed serve as an instrumental indicator of crunchiness. The inflection point of the sensory crunchiness–moisture relationship, marking the loss of half of the dry samples' rating, and that of the mechanical signature's jaggedness compared with moisture relationships are very close. Nevertheless, the instrumental measure implies a sharper transition between the brittle and soggy states than that perceived sensorily. Whether this is merely a sensitivity issue or evidence of a non-mechanical sensory stimulus's or stimuli's interference is yet to be revealed. As far as glass-transition theories are concerned, there is nothing surprising in the sigmoid brittleness loss pattern, except perhaps that it provides another demonstration that the transition in relatively dry foods occurs over a moisture range rather than at a point, and that the resulting textural changes need not be of several orders of magnitude as claimed when the 'polymer science approach' was introduced.

Effect of moisture on stiffness and toughness

Figure 3 to 5 show that increased moisture contents or water activity level can result in a force–displacement curve that is *well above* that of a drier specimen. The effect is highlighted in Figure 11 and 12 where the stiffness parameters (fitted force at 2 chosen displacement levels) are plotted compared with moisture content or water activity. No doubt that in all 3 foods moisture sorption had been accompanied by stiffness and toughness *rise* prior to the specimen's collapse or dissolution. The 3 foods, however, have very different composition, the first starchy (several grains and modified starch) and sugar rich, the second primarily made of puffed corn and is salt rich, and the third highly fatty "low carb" and salty. It is therefore difficult to explain the observation of moisture toughening as stemming for the 3 foods' composition and chemical characteristics. More likely the observed phenomenon in puffed cereals, snacks and other foods is associated with *failure propagation*. When the cell-wall material is partially plasticized by moisture, fracture propagation is inhibited to at least some extent. Consequently, certain elements of the structure do not shatter immediately, which allows the specimen to absorb more mechanical energy during its deformation. The overall result is higher measurable force at the same displacement, which in our terminology translates to increased stiffness and toughness. The blocking of fracture propagation, however, becomes irrelevant when the solid material in the structure is fully plasticized and collapses. Beyond this moisture content, the material is so weak and compliant that all structural integrity is lost. Support for this explanation comes from works of others who observed moisture toughening in wheat gluten films (Gontard and others 1993), Gluten and Starches (Nichols and others 1995), and cereal flakes (Georget and others 1995).

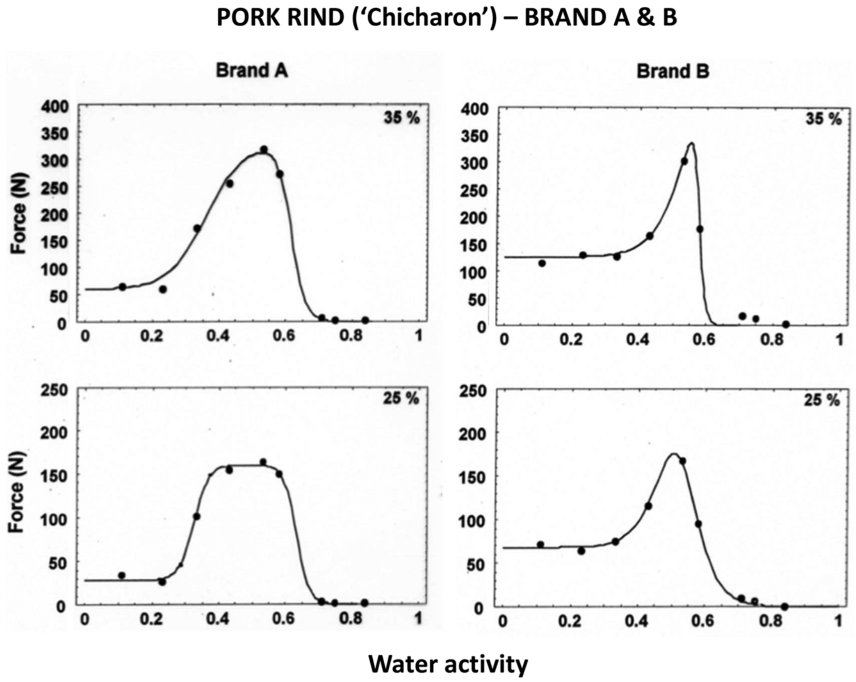

Figure 12–Moisture toughening in 2 commercial brands of Pork Rind ('Chicharon') monitored instrumentally. Top plots: The fitted force at 35% displacement, bottom plots at 25% displacement. Adapted from Gonzalez Martinez and others (2003).

As seen in Figure 11 moisture toughening can be sensed sensorily, albeit identified as increased "hardness." Roudaut and others (2002) reported similar increase in the perceived "hardness" of extruded flat bread at intermediate water activities. Both suggest that people can simultaneously sense and grade crunchiness and toughness separately from the same set of mechanical, acoustic and perhaps other stimuli in the mouth.

Concluding Remarks

No doubt many physical properties of foods originate from their chemical composition and microstructure. Hence it is tempting to explain them in terms of changes in molecular states and the formation or breakage of chemical bonds. Glass transition in synthetic and food polymers has been explained in terms of "molecular mobility" which increases with temperature and/or in the presence of water. Although consistent with this notion, moisture toughening is regulated by the inhibition of failure propagation, a primarily macroscopic phenomenon which follows rules that at least at the current stage of knowledge cannot be derived directly from the food's chemical composition alone. This is evident in that the moisture toughening phenomenon has been observed in foods and systems that bear hardly any similarity as far as chemical composition is concerned and who differ dramatically in their structure and microstructure too.

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