Understanding the Origins of Stickiness in Wheat Flour Tortillas and Devising Strategies to Reduce It

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ABSTRACT OF THE THESIS

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This thesis aimed to determine the factors which affect stickiness in wheat flour tortilla products based on a phase/state-change approach and measurement of water activity and surface properties. Strategies were considered to reduce stickiness of flour tortillas by adding GRAS ingredients and modifying processing conditions.

Commercial wheat tortillas with a wide range of stickiness were selected and equilibrated to different water activity levels (0.12-0.97). Moisture sorption isotherms were developed. Differential scanning calorimetry and mechanical spectroscopy were used to characterize the phase behavior and freezable water, wide-angle x-ray scattering to understand the effect of crystallinity, contact angle measurements to determine the surface hydrophobicity. An objective instrumental test technique was developed using a texture analyzer to quantify the stickiness in tortilla samples. X-ray microtomography was used to measure tortilla cellularity. Tortillas were prepared with Xanthan gum, carboxymethylcellulose, glycerol and propylene glycol. To understand the effect of processing conditions on stickiness, tortillas were prepared using different combinations of dough resting times, baking temperatures and cooling times after baking.

Sticky tortilla showed lower glass transition temperature compared to non-sticky tortillas but both were in rubbery state at room temperature. Higher product Aw resulted in increase in surface energy which in turn caused an increase in instrumental stickiness scores as hypothesized. The polar component of surface energy was found to have a good correlation with stickiness. The sticky tortillas showed low crystallinity as compared to non-sticky tortillas. Tortillas containing 0.5 % gums and 4 % glycerol showed increased water retention, decreased water activity, reduction in surface free energy and lower freezable water. Addition of glycerol reduced the water activity from 0.94 to 0.91. Tortillas baked at 450°F were stickier than tortillas baked at 350°. Rupture force to extend tortillas increases with increase in storage time and temperature. Storage of tortillas at lower temperatures retains freshness as was shown by reduced rupture force values.

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I. INTRODUCTION

1.1 Tortilla History

In Mexico, Central America, the United States, and Canada, a tortilla is a thin, unleavened flat bread, made from finely ground maize (corn) or wheat flour. The first tortillas, which date back approximately 10,000 years before Christ, were made of native corn with a dried kernel. Today, corn tortillas are made from either corn cooked in a lime-based solution or by using corn flour, producing dough, forming it like a pancake and finally baking it in an oven[2]. Among native Mexicans, tortillas are also commonly used as eating utensils.

Figure 1.1 :Tortillas made in Dr. Kokini's laboratory, Food Science, Rutgers University, NJ

In the Old West, "cowpokes" realized the versatility of tortillas and used tortillas filled with meat or other foods as a convenient way to eat around the campfire. The Spanish word **tortilla** [tor´tiʝa] derived from the word *torta*, means a plain round cake. A wheat tortilla is a chemically leavened flat bread consisting of flour, water, shortening or oil, baking powder, and salt. Flour tortillas are flat, circular, light colored, 1-2 mm thick and 15-30 cm in diameter (Wang and Flores 1999). Tortilla products have been traditionally homemade and consumed in Mexico for centuries (Serna-Saldivar et al. 1988).

Flour tortillas are a favorite bread item in NASA Shuttle menus. Tortillas provide an easy and acceptable solution to the bread crumb and microgravity handling problem, and have been used on most Shuttle missions since 1985[1]. Depending on the region tortillas may vary in size from about 6 to over 30 cm (2.4 to over 12 in). Wheat tortillas are very similar to the unleavened bread popular in Arab, eastern Mediterranean and southern Asian countries. It is also similar to Laobing (in China) and Roti (India).

1.2 Tortilla Industry at Present

Tortillas constitute 32 percent of the sales for the U.S. Bread Industry. It trails white bread sales by only two percent - making tortilla the second most popular bread type in America. Its sales surpass those of whole wheat bread, bagels, rolls, English muffins and pita bread. In 2004, tortilla sales in the U.S. reached the $6.1 billion mark and are expected to reach $6.6 billion dollars in 2006[2]. Tortilla Industry Association (TIA) estimates that Americans consumed approximately 85 billion tortillas in 2000 (not including tortilla chips). Tortillas can be used as substitutes for traditional breads in products such as hot dogs, lasagna, pitas, sandwiches and pizza. It can be used to hold a variety of fillings, used as tasty food scoops, toasted and topped with salad, or served hot

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[1] (www.spaceflight.nasa.gov)
[2] (http://www.tortilla-info.com)

and plain. It can be warmed in the oven, steamed, grilled, fried, heated in a microwave or toaster or baked in the oven.

1.3 New Tortilla Classification

Economic Classification Policy Committee of the Office of Management and Budget has established a new classification for tortillas[2]. Under the new system, bakeries and tortillerias (dedicated tortilla manufacturers) are placed together in a category within "Food Manufacturing". Tortillas were previously listed in the category of "food preparations not classified elsewhere”. The reclassification confirms the significance of the tortilla as a major manufactured food and the fastest growing segment of the U.S. baking industry. Tortilla manufacturing is now established as a sub-category of food manufacturing, equivalent to that of bread and bakery product manufacturing.

1.4 Nutritional Information[3]

Flour tortilla contain relatively low-fat and is a source of iron along with B vitamins. They have about 146.37 calories with 3.06 grams of fat per serving.

Table 1 1:

Nutritional Chart for Tortillas

MISSION FOODS, MISSION Flour Tortillas, Soft Taco, 8 inch serving 51.00g					
Protein	4.44	g	Total lipid (fat)	3.06	g
Carbohydrate, by difference	25.3	g	Ash	1.07	g
Energy	146.37	kcal	Water	17.14	g
Energy	613.02	kj	Calcium, Ca	97.41	mg
Iron, Fe	1.01	mg	Sodium, Na	248.88	mg
Fatty acids, total saturated	0.35	g	8:00	0	g
10:00	0	g	12:00	0	g
14:00	0	g	16:00	0.04	g
18:00	0.28	g	20:00	0.01	g
18:1 undifferentiated	1.4	g	18:2 undifferentiated	0.44	g
18:3 undifferentiated	0.02	g	20:4 undifferentiated	0	g
22:00	0.01	g	14:01	0	g
16:1 undifferentiated	0	g	20:01	0.01	g
Fatty acids, total monounsaturated	1.41	g	Fatty acids, total polyunsaturated	0.47	g
15:00	0	g	17:00	0	g
17:01	0	g	20:3 undifferentiated	0	g

USDA Handbook 8. Based on the average-sized serving; serving sizes may vary depending on the brand.

[3] USDA Handbook 8

II. OBJECTIVES

This study has the following main objectives:

1. To find the physical and physico-chemical basis of stickiness in wheat flour tortillas.

2. Based on determination of the cause of stickiness try to improve stickiness in tortillas using added GRAS ingredients (gums and polyols).

3. Determine tortilla processing conditions which provide changes in the properties of tortillas that reduce stickiness.

III. LITERATURE REVIEW

3.1 Properties of Wheat Tortillas

Studies on wheat tortillas have mostly concentrated on improving dough functionality (Adams and Waniska 2002) and increasing shelf life (Kelekci 2003, Seetharaman et al. 2002). Good quality flour tortilla should be flexible without tearing and cracking when folded, soft without sticking together, light colored, and well puffed (Bello et al. 1991, Pascut et al. 2004).

3.2 Methods to Evaluate Tortilla Characteristics:

Waniska (1976) introduced a subjective rollability test to evaluate tortilla self life. Friend et al (1995) used a rollability test to monitor effect of the additves in wheat tortilla texture. Rollability of tortillas at 23° C was evaluated every other day by wrapping a tortilla around a dowel (1.0 cm diameter). Tortillas were rated for cracking and breakage. Rollability was rated on a scale of 1-5, where 1 = no cracking (best), 2 = signs of cracking but no breaking, 3 = cracking and breaking beginning on one surface, 4 = cracking and breaking imminent on both sides, and 5 = unrollable, breaks easily. He observed better rollability scores when tortillas were made using 11.8 % protein flour as compared to 10.7 % protein flour. Sensory evaluation was used to evaluate appearance, puffing and brown spots in tortillas (Suhendro et al. 1995) and dough characteristics and dough machinability measurements (Friend et al. 1995). Extensibility, puncture, and bending tests can be used to measure tortilla texture objectively (Bejosano et al. 2005).

3.2.1 Bending Test:

In the bending test a tortilla strip 30 × 35 mm was evaluated. The strip was placed horizontally. The lower grip was attached to the texture analyzer platform and an aluminum guillotine attached to the analyzer arm. The test was run with the same setup as the two-dimensional extensibility test which is thoroughly explained in the materials and methods section. The probe moved down for 5.0 mm at a test speed of 1.0 mm/sec until the guillotine bent the tortilla strip to a controlled 40° angle (Suhendro et al 1999a).

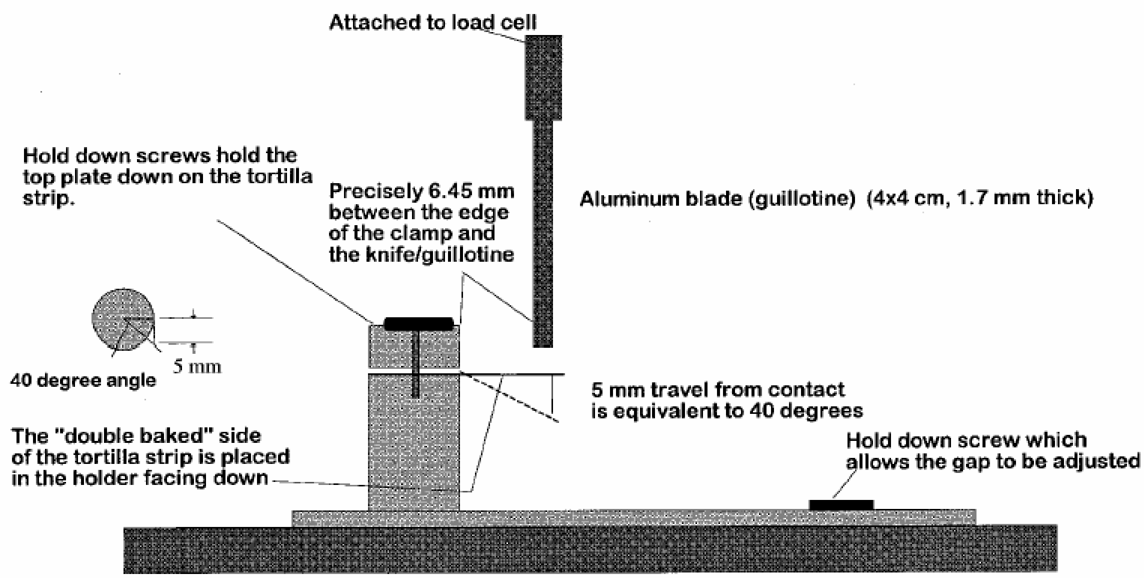


Figure 3 1: Bending instrument was used to objectively measure bending properties of corn tortillas **(Suhendro et al. 1998)**

The bending modulus of deformation (N/m), peak force (N), and bending work (Nm) were recorded for tortillas purchased from supermarket.

3.2.2 Puncture Test:

In the puncture test a metal probe 3.2 cm in length and 3.0 mm in diameter was used to puncture the tortillas. Tortillas were placed on an aluminum plate 1.3 cm thick with a hole 6.0 mm in diameter that accepted the metal probe as it is punched through the product. Force (N) and distance (mm) were recorded. The burst rig test, which is very similar to the puncture test, was used to evaluate tortilla texture in this thesis and is fully explained in the materials and methods section.

3.2.3 Tensile Test (Two dimension extensibility test)

Suhendro in 1999 used a tensile test to measure the extensibility of corn tortilla sample as shown in following figure. This method is thoroughly explained in the materials and methods section.

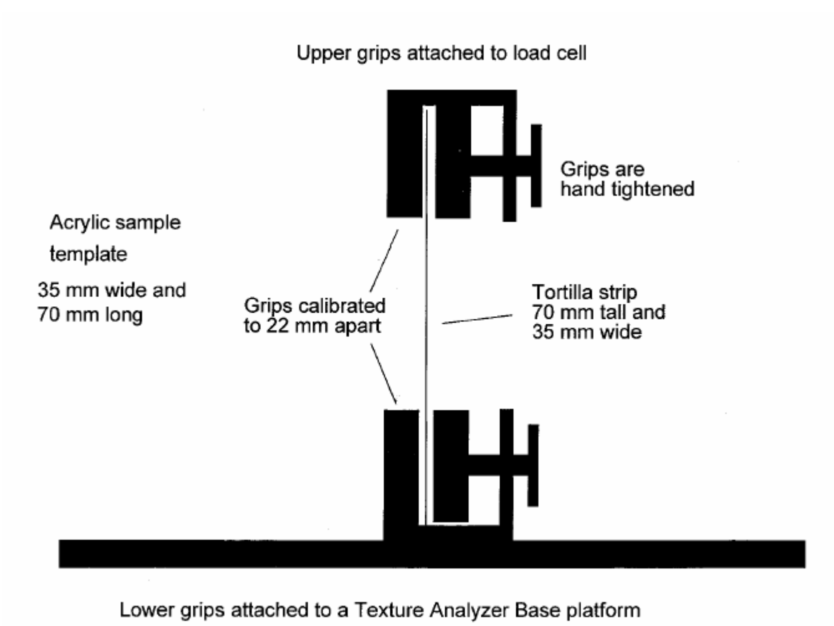


Figure 3 2 : Drawing of the apparatus used to objectively measure extensibility properties of corn tortillas (Suhendro 1999)

3.2.4 Foldability or Stretchability Measurement:

Wang and Flores measured the effect of addition of starch and gluten isolated from different wheat flour on tortilla stretchability or foldability (Wang and Flores 1999). The firmness of flour tortillas was determined with a texture analyzer equipped with a rounded-end probe 1.90 cm (0.75-in.) in diameter (TA- 108) as mentioned in the puncture test. Foldability of tortillas was subjectively measured. In this method one tortilla was folded firmly and then unfolded, and cracks on the surface of the tortilla were evaluated. A hedonic rating scale of 1–10 was used. Ten points were given to tortilla with no visible cracks on surface after they were folded. Wang and Flores concluded that flour reconstitution does not fully recover the properties of wheat flour. Foldability of tortillas made from reconstituted wheat flour was lower indicating a weak tortilla texture. (Serna-Saldivar 2004) substituted wheat flour with triticale and indicated that triticale could substitute for 50% of wheat flour without affecting texture, color, flavor, and overall acceptability of tortillas. (Mao 2001) analyzed the effect of mechanically damaged starch on tortilla texture. He noticed that stretchability values were lower in fresh tortillas made with damaged starch. He explained that damaged starch is finer in size, and its surface cannot be bound by protein. Damaged starch increases the starch surface by absorbing water and it swells more than sound starch. This results in insufficient formation of gluten covering the surface of the starch in the tortilla dough (Farrand 1969). Lower force of compression and shear was needed to break the tortillas made from damaged starch flour. He reasoned that the damaged flour had a higher degree of water absorption which led to a higher tender texture. Tortillas became stiff more quickly during refrigerated storage (3–10°C) (Bueso 2006). According to Waniska the main problems in tortillas making can be summarized as premature molding due to high pH, excessive adhesion (zippering) and sticking, inconsistent circular shape, diameter and thickness, translucency or incomplete opacity and difficulties in achieving extended shelf life and freshness characteristics (Waniska 1999).

3.2.5 X-ray diffraction analysis

X-Ray powder diffraction analysis is a powerful method by which X-Rays of a known wavelength are passed through a sample to be identified in order to identify the crystal structure of the material. The wave nature of the X-Rays means that they are diffracted by the lattice of the crystal to give a unique pattern of peaks of 'reflections' at differing angles and of different intensity, just as light can be diffracted by a grating of suitably spaced lines. The diffracted beams from atoms in successive planes cancel unless they are in phase, and the condition for this is given by the Bragg’s law.

$$n\lambda = 2d \sin \theta \text{ ....(1)}$$

Where , λ is the wavelength of the X-Rays

d is the distance between different plane of atoms in the crystal lattice.

θ is the angle of diffraction.

3.2.5.1 Bragg’s law[4]:

Bragg's Law can easily be derived by considering the conditions necessary to make the phases of the beams coincide when the incident angle equals the reflecting angle. The rays of the incident beam are always in phase and parallel up to the point at which the top beam strikes the top layer at atom z as shown in Figure 3.3. The second beam continues to the next layer where it is scattered by atom B. The second beam must travel the extra distance AB + BC if the two beams are to continue traveling adjacent and parallel. This extra distance must be an integral (n) multiple of the wavelength (λ) for the phases of the two beams to be the same:

$$n\lambda = AB + BC \qquad \text{.............................(2)}$$

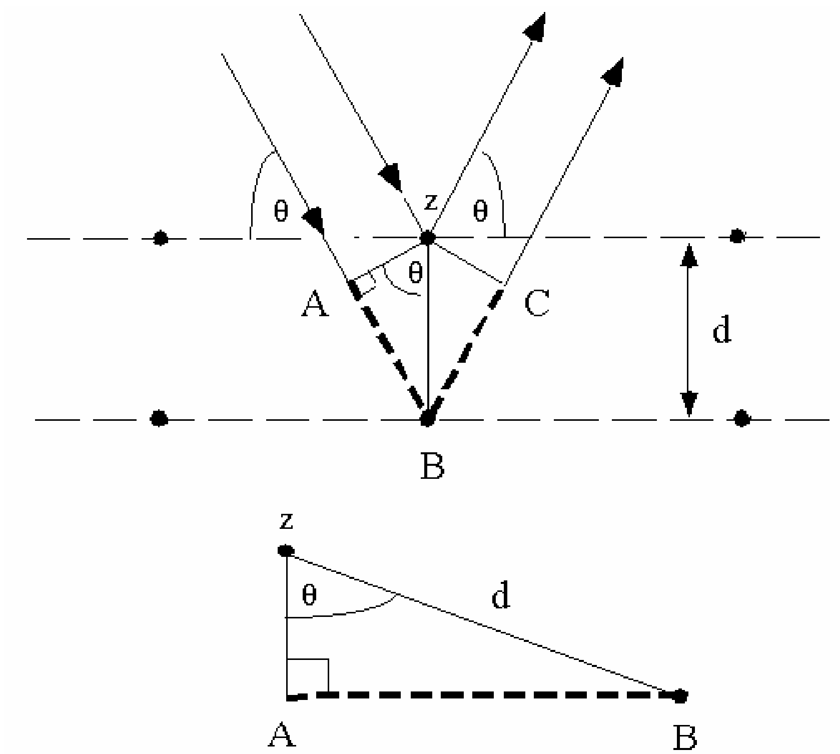


Figure 3.3 : Deriving Bragg's Law using the reflection geometry and applying trigonometry. The lower beam must travel the extra distance (AB + BC) to continue traveling parallel and adjacent to the top beam

[4] http://www.eserc.stonybrook.edu/ProjectJava/Bragg/

Recognizing d as the hypotenuse of the right triangle ABz, we can use trigonometry to relate d and θ to the distance (AB + BC). The distance AB is opposite θ so,

$$AB = d \sin \theta \text{ ....(3).}$$

Because AB = BC eq. (2) becomes,

$$n\lambda = 2AB \text{ ……………………….(4)}$$

Substituting eq. (3) in eq. (4) we have,

$$n\lambda = 2\, d \sin \theta\,,$$

The X-Ray detector moves around the sample and measures the intensity of these peaks and the position of these peaks [diffraction angle 2θ]. These results can be plotted as Intensity vs 2θ .

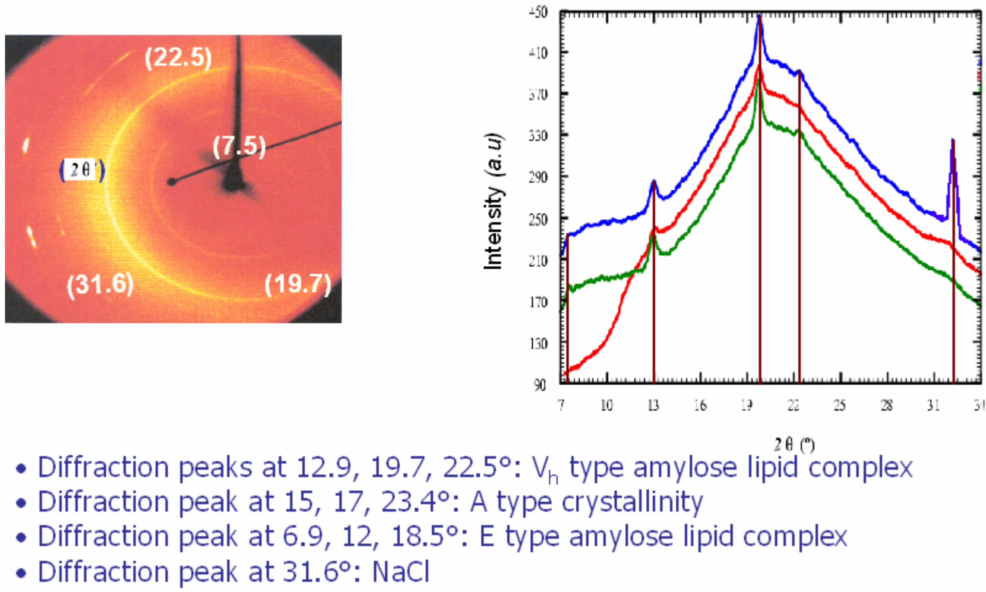


Figure 3 4 : Expected crystallinity patterns for a starch based product (Kokini lab), Food Science, Rutgers University, New Jersey

(Martínez-Bustos 1999) examined X-ray diffraction patterns (Figure 3.5) of wheat flour tortillas to understand the effect of infrared baking on tortilla relative crystallinity.

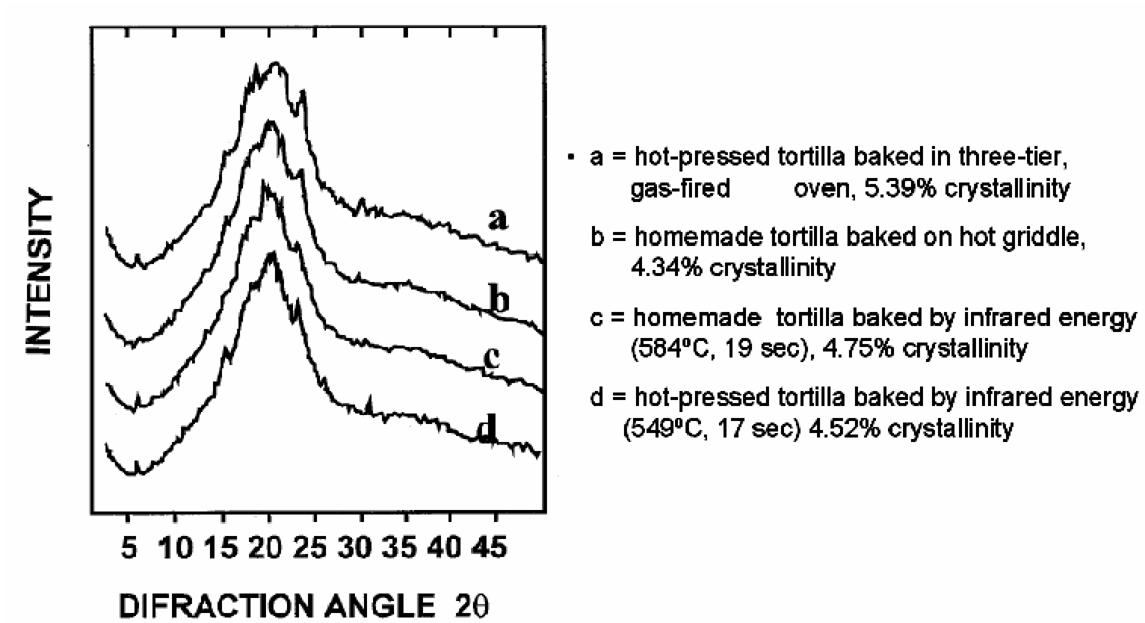


Figure 3.5 : X-ray diffraction patterns of wheat flour tortillas (Martínez-Bustos 1999)

As shown in Figure 3.5 X-ray diffraction has a similar pattern for tortillas prepared by the traditional process and baked by the IR method and for homemade tortillas baked on a hot griddle. Becker et al. 2001 observed melting of crystalline structure and formation of amylose-lipid complexes when maize grits were heated for 15 min or longer. It is Vh type crystallinity with a peak at 2 theta ~ 20˚.

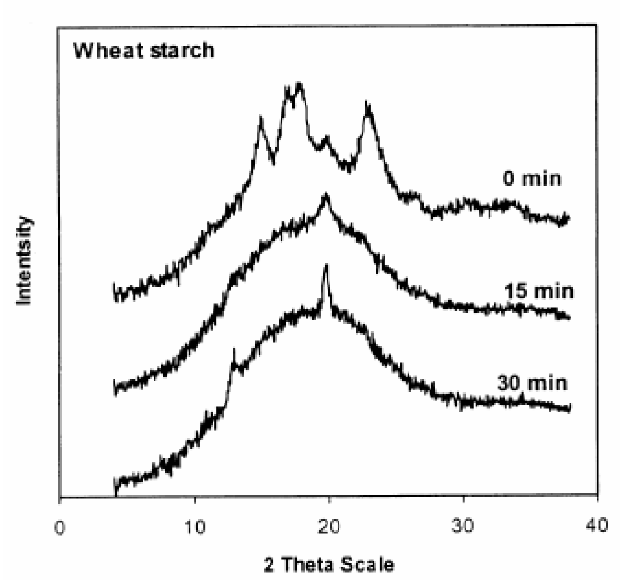


Figure 3 6 : X-ray diffraction pattern of wheat starch heated at 140 °C for 0, 15 or 30 min confirming the presence or absence of crystalline amylose-lipid complexes (Becker et al. 2001)

Zobel 1988 showed X-ray pattern for starch. The B structure is due to crystallization of an amorphous starch melt, which presumably is mainly amylopectin owing to complexing of the amylose with granule fatty acids. Thus, heating amylose-bearing starches that contain fatty acids can cause simultaneous crystallizing and melting effects to occur.

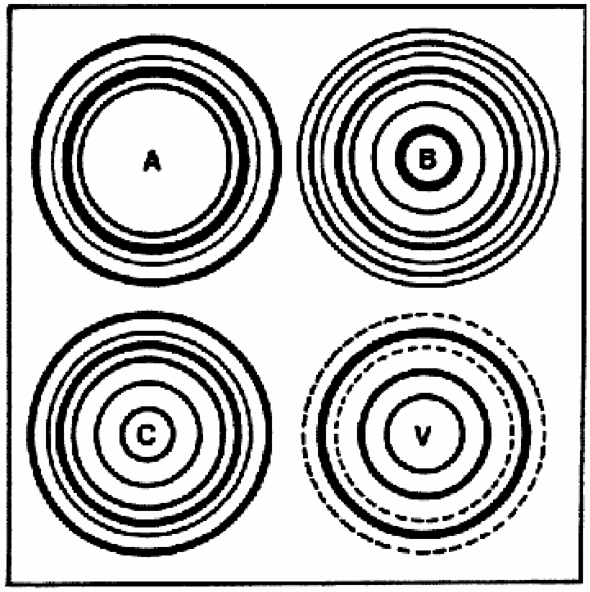


Figure 3 7 : Starch X-ray pattern designations **(Zobel 1988)**

Application of the term "melting" to starch gelatinization is based on X-ray and synthetic polymer technologies. Thus, from diffraction studies, crystalline specimens yield reflections from crystal planes. After melting, these reflections disappear and a broad halo appears, indicating a change from a crystalline to an amorphous (molten) state. Accordingly, the material is no longer a rigid solid but rather exhibits the properties of a liquid. In this state, polymeric materials may show low fluidity, and as such, fit into the classic definition of a melt. High molecular weight materials, however, impart high viscosity or rubber like qualities to the molten-liquid state (Mandelkern 1964). Waxy maize also has an A structure and lipids similar to those in maize, but amylose is absent for complex formation. Potato starch has a B structure and 22% amylose but lacks a lipid fraction for complex formation. The C form is native to certain root and tuber starches.

3.2.6 Water Activity Measurement[5]

Suhendro et al. 1995 observed that the addition of glycerol reduced water activity of tortillas from 0.93 to 0.9. Water activity is an important characteristic of tortillas which governs their stability. Water activity is derived from fundamental principles of thermodynamics and physical chemistry. In the equilibrium state:

$$\mu = \mu_o + RT \ln(f/f_o)$$

Where: μ (J mol^{-1}) is the chemical potential of the system; μ_o is the chemical potential of the pure material at the temperature T (°K); R is the gas constant (8.314 J mol^{-1} K^{-1}) ; f is the fugacity or the escaping tendency of a substance; and f_o is escaping tendency of pure material (van den Berg and Bruin, 1981). The activity of a species is defined as $a = f/f_o$. When dealing with water, a subscript is designated for the substance,

$$a_w = f/f_o$$

a_w is activity of water, or the escaping tendency of water in system divided by the escaping tendency of pure water without radius of curvature. For practical purposes, under most conditions in which foods are found, the fugacity is closely approximated by the vapor pressure ($f \sim p$) so;

$$a_w = f/f_o \sim p/p_o$$

[5] http://wateractivity.org/theory.html

Relative humidity of air is defined as the ratio of the vapor pressure of air to its saturation vapor pressure. Multiplication of water activity by 100 gives the equilibrium relative humidity (ERH) in percent.

$$a_w = p/p_o = \mathrm{ERH}\ (\%) / 100$$

Working Principle of Aqualab:

Aqualab uses the chilled-mirror dewpoint technique as shown above to measure the aw of a sample. The sample is equilibrated within the headspace of a sealed chamber that contains a mirror and a means of detecting condensation on the mirror. At equilibrium, the relative humidity of the air in the chamber is the same as the water activity of the sample. In the Aqualab, the mirror temperature is precisely controlled by a thermoelectric cooler.

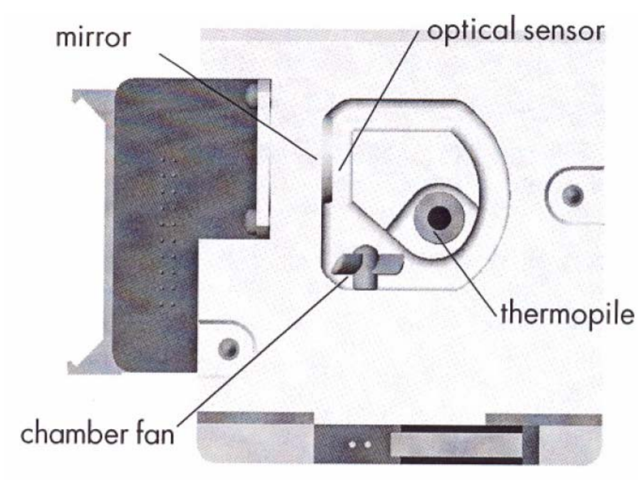


Figure 3 8: View of inside block chamber of AquaLab water activity meter[6] (Aqualab manual)

[6] AquaLab water activity meter manual

Detection of the exact point at which condensation first appears on the mirror is observed with a photoelectric cell. A beam of light is directed onto the mirror and reflected into a photodetector cell. The photodetector senses the change in reflectance when condensation occurs on the mirror. A thermocouple attached to the mirror then records the temperature at which condensation occurs. The final water activity and temperature of the sample is then displayed.

3.3 Tortilla Making:

Wheat flour tortillas are prepared by three processes

1. Hot-press
2. Hand stretch
3. Die-cut

Tortillas are mass produced using the hot-press method. In this method a hot press is used to press dough balls and make round disks of dough which are subsequently baked to in an oven. More than 90% of increase in the tortilla production is attributed to the hot press method (Waniska 1999). Hot-press method is not the most efficient method, but hot-press tortillas have the desired soft texture and retain more flexibility during storage. The die cut method is the most efficient method with lower cost, but the tortillas tend to be less soft, pasty and lose flexibility more quickly (Waniska, 1999). Die–cut tortillas used to prepare processed foods such as Burritos. Hand-stretch tortillas are large, thin and stronger (Waniska, 1999).

Tortilla making steps include dough making, dough ball making, proofing (dough resting), hot-pressing, baking, tortilla cooling and packaging/storage. Dough making involves mixing the ingredients in a blender. Water is added to the dry mixture and blended for 5 min.

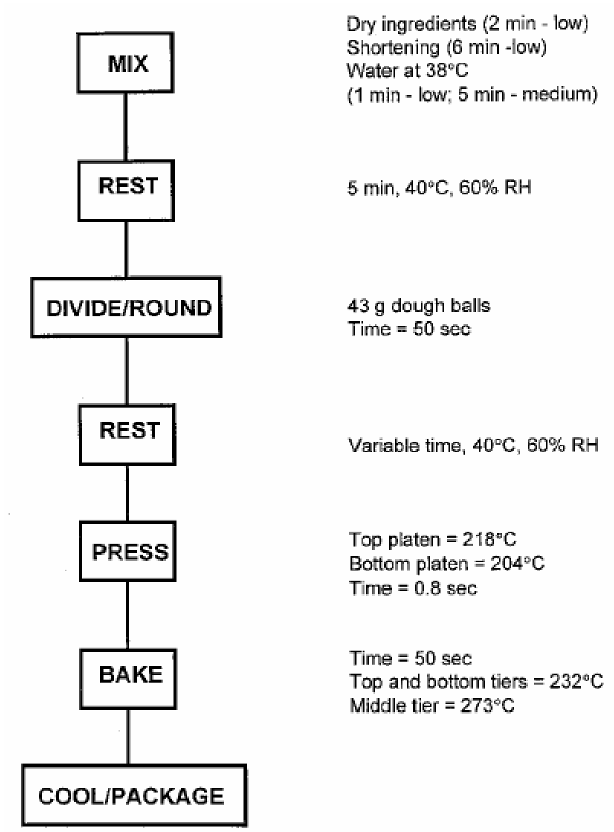


Figure 3.9 : Process flow chart for hot-press flour tortillas (Cepeda 2000)

The dough is rested for five minutes before turning it into balls. Dough balls are once again proofed for some time at a specific temperature and humidity before turning them into dough disks using hot-press. Dough disks are baked in the oven (~250˚C). Hot tortillas are cooled to room temperature before packaging and stored at lower temperatures. As shown in Figure 3.9 (Cepeda 2000) used the modified method of Bello et al (1991) to make tortillas.

3.4 Tortilla Ingredients:

The main tortilla ingredients are wheat flour, salt, baking powder, shortening and water. The other ingredients that might be used depending on need are gums, emulsifiers, preservatives, acidulants, polyols, fiber, sugar, and whey powder. Tortilla ingredients, percentage of addition, baking and hot pressing temperatures, dough resting and tortilla cooling time can be different depending on the product requirement. Wheat flour provides body to tortillas. Two main components of wheat flour are starch and protein.

Table 3.1

General Properties of Starch Granules

	Wheat starch
Granule size (major axis, μm)	2–55
Amylose (%)	28
Gelatinization/pasting temperature (°C)[a]	52–85
Relative viscosity	Low
Paste rheology[c]	Short
Paste clarity	Opaque
Tendency to gel/retrograde	High
Lipid (%DS)	0.9
Protein (%DS)	0.4
Phosphorus (%DS)	0.00
Flavor	Cereal (slight)

(Fennema 1996)

Starch granules are composed of amylose and amylopectin molecules arranged radially. They both contain crystalline and non crystalline regions in alternating layers. Crystallinity is produced by ordering of amylopectin chains. Starch molecules are semi-crystalline and hence insoluble in cold water. They require heat to gelatinize. Cereal lipids have been categorized into three groups: non-starch lipids, granule surface lipids and internal lipids. Monoacyl lipids that occur in all three categories can complex with the amylase. These complexes are believed to be present in native starch granules, next to free amylase and lipids. These complexes become mobile and readily crystallize from water as anhydrous crystals giving rise to a V-type X-ray diffraction pattern (Zobel, 1988). The melting temperature of these crystals is around 110 °C for endogenous cereal lipids. Lipid free amylose leaches out when heated with water and contributes toward crystallinity. Whereas lipid-complexed amylose restricts swelling and dispersion of granules and solubilization of amylose and hence reduces stickiness (Adhikari 2001) Wheat starch contains ~ 0.4% protein. Gluten forms a continuous network when blended with water in which hydrated starch granules get trapped. Salt gives taste and strength to tortillas. Baking powder is necessary to get leavening effect in tortillas. Shortening gives soft texture to tortillas. Preservatives such as potassium sorbate are added in tortillas to counter mold growth and thus prolonging shelf life of product. Formic acid is used to lower pH of product which enhances function of preservative. Anti-staling ingredients such as gums (Xanthan gum,guar, CMC), polyols (Glycerol, Propylene glycol), enzymes (amylase) and emulsifiers (mono/diglycerides) improve tortilla texture characteristics by reducing starch retrogradation.

3.4.1 Hydrocolloids

Hydrocolloids are a group of food ingredients that vary widely in form and function and are used at low levels. They are multifunctional and can work as water binders, texturizers and fat replaces. Hydrocolloids can help extend the shelf life of tortillas and improve freeze thaw stability. Some hydrocolloids (guar, xanthan, agar, pectin, etc.) absorb up to 6 times their weight in water(Rosell et al. 2001). The addition rate in tortillas can range from 0.01% to 0.5%. The most common hydrocolloids used in corn tortillas are CMC (Serna-Saldivar et al. 1990) and guar. Hydrocolloids increase water absorption many times their weight. Friend et al. 1993 observed an increase in water absorption percentage when dough was prepared using 1% CMC (46.2%) compared to control dough with 45% water absorption. Hydrocolloids are used in baked goods primarily to enhance finished product moistness (Heflich 1996). (Yau et al. 1994) evaluated the effect of CMC, guar gum, hydroxypropyl methylcellulose (HPMC) XG and other additives on storage stability of corn tortillas. Tortillas prepared using CMC retained more moisture (47.1 %) after 7 days of storage as compared to control tortillas (46.8 %). Addition of Carrageenan (0.3, 0.5, 1%), CMC (0.5, 1 %), guar gum (0.3, 0.5, 1%), HPMC (0.5, 1%), and XG (0.3, 0.5, 1 %) did not affect the stickiness of corn tortilla dough (Masa). Yau et al. observed improvement in storage stability of corn tortillas by 4 days when CMC (0.5, 1%) was incorporated.

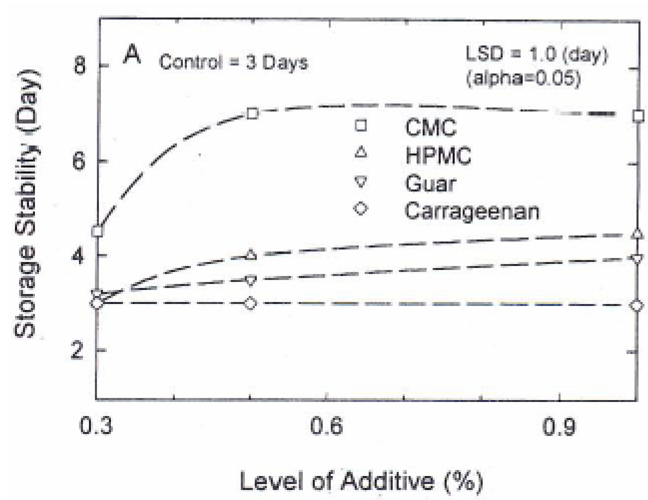


Figure 3 10 : Effect of gums on storage stability of corn tortillas (Yau et al. 1994)

They explained that CMC decreases starch gelatinization due to competition for water in dough and low retrogradation. The gums in masa during thermal processes compete with water retarding the degree of starch gelatinization (Bell 1990, Christianson 1982). Hydrocolloids also inhibit recrystallization of gelatinized starch which leads to decreased staling rate of baked foods (Christianson 1982).

Wheat tortillas containing natural gums, modified cellulose gums, or commercial blends were always consistently round, puffed slightly browned, and of good quality(Friend et al. 1993). Friend et al. also observed improvement in rollability scores of wheat tortillas when 0.5% guar and 1% CMC was incorporated as shown in the figure 3.11

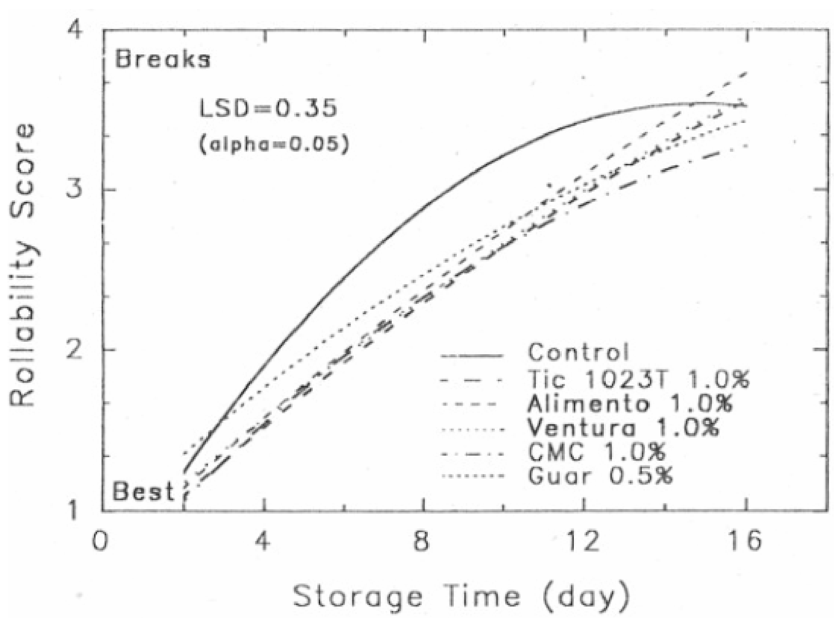


Figure 3.11 : Effect of addition of ingredients on tortilla rollability scores

. . (Friend et al. 1993)

3.4.2 Xanthan Gum

Xanthan gum (XG) is a polysaccharide. It is used as a food additive and can modify rheology of a food. Xanthan gum is produced by fermentation of glucose or sucrose by the *Xanthomonas campestris* bacterium.

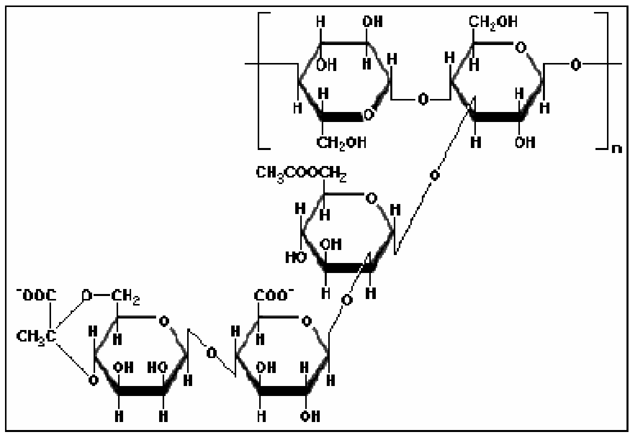


Figure 3 12 : Chemical structure of xanthan gum[7]

Xanthan gum is a polysaccharide with a β-D-glucose backbone like cellulose. Every second glucose unit is attached to a trisaccharide consisting of mannose, glucuronic acid, and mannose. The mannose closest to the backbone has an acetic acid ester on carbon 6, and the mannose at the end of the trisaccharide is linked through carbons 6 and 4 to the second carbon of pyruvic acid. XG keeps tortilla edges soft and pliable after freezing and it makes dough processing operations easier to achieve (Gurkin, 2002). Román-Brito et al. 2007 analyzed the effect of XG addition on corn tortilla moisture content. They observed that control tortillas had the lowest moisture content value as compared to tortilla containing 0.25, 0.5 and 1% XG. These results might be related to the

[7] http://www.scientificpsychic.com/fitness/carbohydrates2.html

retrogradation phenomenon, where some water is lost due to interaction among starch chains explained by Román-Brito et al. 2007. When the concentration of gum was increased in tortillas, the loss of water was lower. Another explanation could be that hydrocolloids produce a network that hinders water migration in tortillas, which retards the retrogradation phenomenon.

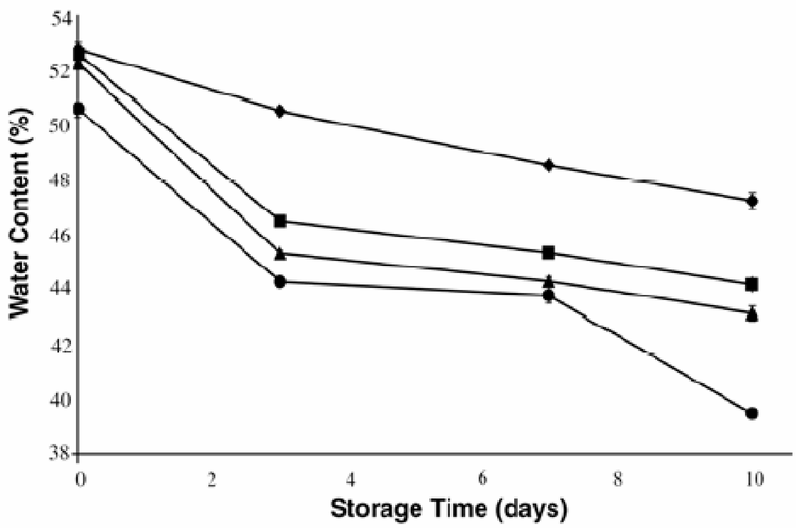


Figure 3 13 : Effect of xanthan gum on the moisture content (%) of corn tortillas stored for different times at 4°C. ●, Control; ▲, 0.25%; ■, 0.50%; ♦, 0.75% (w/w).

(Román-Brito et al. 2007)

XG hydrates faster and can act as a texturizing agent in tortilla making. XG was reported to uniformly distribute moisture throughout cake batters (Dziezak 1991). XG is an emulsion stabilizer; holds water; enhances freeze-thaw stability; inhibits starch retrogradation; improves shelf life and serves to bring about stabilization of dispersions, suspensions, and emulsions, thickener (Fennema 1996). XG can eliminate side ruptures in frozen burritos and enchiladas (Gurkin, 2002). Friend et al. 1995 observed increase in dough firmness and stickiness when XG was added at 0.5 % level. XG when added singly induced a desirable increase in dough resistance to extension (Collar 1999). Friend et al. 1995 observed an increase in water absorption of wheat tortilla dough when XG was incorporated. Dough incorporated with XG at 0.3 and 0.5% had water absorption of 47.5 and 48% respectively compared to 47% of control dough. Dough mixing time reduced from 6 to 5 min when 0.5% XG was incorporated.

3.4.3 CMC (Carboxymethyl cellulose)

Carboxymethyl cellulose, or CMC, is a cellulose derivative with carboxymethyl groups (-CH_2-COOH) bound to some of the hydroxyl groups of the glucopyranose monomers that make up the cellulose backbone. It is made by chemically altering the long chains of purified cellulose. CMC is made by treating cellulose with aqueous sodium hydroxide followed by an esterification reaction. CMC has the ability to increase moisture retention (Dziezak 1991). It controls rheological properties of cereal batters and dough (Sindhu and Bhawa 2000). (Bueso 2006) observed the addition of 0.25% CMC and 1,650 AU of amylase and 0.5 % CMC improved tortillas subjective pliability scores. However, as shown in the Figure 3.14 tortillas with 0.5% CMC and stored for 21 day at refrigeration temperature (3-10 ˚C) had a lower pliability score(>1.5). The authors reasoned that if tortillas age like a typical semicrystalline system, recrystallization should occur in the –23°C to 57°C range, showing a maximum rate around the middle of this range (17°C). It was found that freezing preserved tortilla pliability better than room and refrigeration temperatures after 21 days of storage, especially when 0.5% CMC was added. This could be because freezing temperatures close to or below tortilla *T*g limit the molecular mobility of compounds such as amylopectin and hence reduce their rate of crystallization.

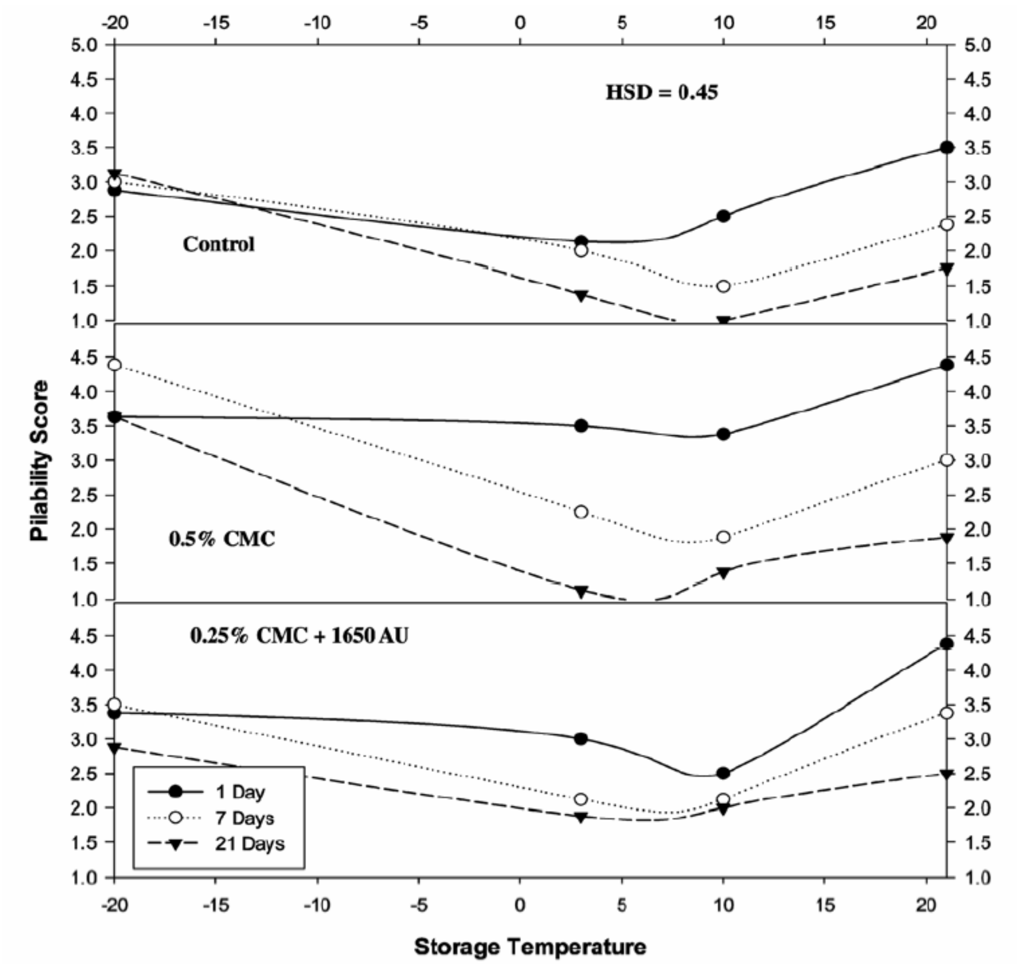


Figure 3 14 : Effect of storage temperature on pliability of tortillas with added carboxymethylcellulose (CMC) and maltogenic amylase after 1, 7, and 21 days **(Bueso 2006)**

Tortillas were evaluated in triplicate at 20 min, 1, 7, and 21 days after baking. Pliability was measure by squeezing a tortilla in the palm of one hand, holding it for 2 sec, and then releasing. A five-point scale was defined as 1= complete crumbling; 2= almost total crumbling; 3= a lot of cracking, no crumbling; 4= isolated cracks; and 5= completely pliable (no cracks).

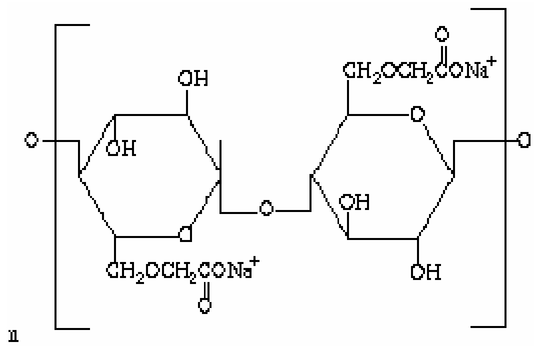


Figure 3 15 : Chemical structure of CMC[8]

(Friend et al, 1993) observed that when tortillas were prepared with 1% CMC water absorption increased from 47 to 48.5%. He also observed that resultant dough was ropey and slightly sticky which reduced the machinability (ease in handling of dough in dough ball maker, dough disk maker) of dough. Friend et al also found improvement in shelf stability of tortilla from 8.7 to 9.9 and 9.3 days when tortillas were prepared from 0.3 and 1% CMC respectively. Shelf stability was determined by wrapping a tortilla around a dowel and evaluating the extent of cracking and breaking. Two tortillas per treatment were wrapped around a 1.0-cm dowel every other day for 16 days and subjectively rated for cracking and breakage. The rollability scale was: 1= no cracking (best); 2= signs of cracking, but no breaking; 3=cracking and breaking beginning on one surface; 4= cracking and breaking imminent on both sides; and 5= breaks easily. Key functions of CMC in foods can be summarized as viscosity enhancer, water organizer, thickener, film former and ice/sugar crystal growth controller.

[8] class.fst.ohio-state.edu/.../lect20.html

3.4.4 Glycerol

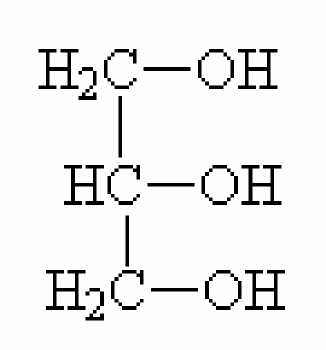


Figure 3 16 : Chemical structure of glycerol

Chemical name: Propane-1,2,3-triol

Chemical Formula: $C_3H_5(OH)_3$

Glycerol (glycerin or glycerine) is colorless, odorless, viscous liquid, widely used in food and pharmaceutical formulations. It is an alcohol with three hydrophilic alcoholic hydroxyl groups as shown in Figure 3.16. These groups are responsible for its solubility in water and its hygroscopic nature.

3,4.2.1 Glycerol in Foods and beverages

Glycerol, glycerin, glycerine,or 1,2,3-propanetriol, $CH_2OHCHOHCH_2OH$ is colorless, odorless and sweet-tasting liquid . Glycerol is a hygroscopic alcohol which melts at 17.8°C and is miscible with water and ethanol. The hygroscopic property of glycerol makes it valuable as a moistener. Glycerol is present as glycerides in all animal and vegetable fats and oils. Glycerol is synthesized on a commercial scale from petroleum. Glycerol can also be obtained during the fermentation of sugars if sodium bisulfite is added with the yeast. Glycerol serves as humectant, solvent and sweetener and may help preserve foods (such as anti-staling agent in cake). It works as solvent for flavors (such as vanilla) and food coloring. It is used in cosmetics, liquid soaps, candy, cakes, casings for meats and cheeses, inks, lubricants, production of antibiotics and medicine. Glycerol was reported to increase both shelf life and shelf stability (preservative free 90 days stable tortilla) of wheat and corn tortillas (Skarra et al 1988). According to Skarra glycerol acts as plasticizer, reduces water activity and maintains a higher nonfatty fluidity that is important for moistness, tenderness and flexibility. Water activity decreases with an increase in polyol level in tortillas (Suhendro et al, 1995) observed that addition of 4% glycerol reduced tortilla water activity from 0.93 to 0.9. It can be also seen from the Table 3.2 that propylene glycol (PG) and glycerol were more effective in decreasing water activity than were the other polyols.

Table 3.2

Water Activity of Tortillas Containing Polyols

Treatment	**Water Activity (%)**
Control[a]	0.93
Propylene glycol 4%	0.90
Glycerol 2%	0.91
Glycerol 4%	0.90
Glycerol 6%	0.90
Sorbitol 4%	0.92
Maltitol 4%	0.92
Least significant difference ($\alpha = 0.05$)	0.01

[a] Prepared from flour B (11.0% protein).

(Suhendro et al, 1995)

Suhendro et al. explained that PG and glycerol have lower molecular weights than sorbitol and maltitol; thus, they more effectively increased osmotic pressure and decreased water activity. As shown in the following table moisture content of tortillas decreased from 28.2 to 26.8% as when glycerol was incorporated at 6%. Darker brown spot were observed when tortillas were prepared with 6% of glycerol irrespective of protein content of wheat flour.

Table 3.3

Effect of Addition of Polyols on Tortilla Characteristics

Treatment	Moisture (%)	Appearance	Puffing	Brown Spots[a]
Flour A (10.2% protein				
Glycerol				
0% (control)	28.2	Rough, round	High	Light, few
2%	27.7	Rough, off-round	Medium	Light, few
4%	27.3	Rough, off-round	Medium	Light, more
6%	26.7	Rough, off-round	Medium	Darker, more
Flour B (11.0% protein)				
Glycerol				
0% (control)	30.5	Smooth, round	High	Light, few
2%	29.6	Smooth, round	High	Light, few
4%	29.3	Smooth, round	High	Light, few
6%	27.7	Smooth, round	High	Darker, more
Propylene glycol				
2%	29.8	Smooth, round	High	Light, few
4%	28.8	Smooth, round	High	Light, more
6%	27.3	Less smooth, off-round	Medium	Darker, more

(Suhendro et al, 1995)

(Vittadini et al, 2004) observed that addition of glycerol significantly decreases the 'freezable water' content of tortillas. In the Figure 3.17 the control tortillas had 44% "freezable" water (FW) and glycerol/salt added tortillas had 28% FW. An endothermic transition was observed at around 0˚C. It can be attributed to ice melting (Baik and Chinachoti, 2001). The transition was slightly shifted to lower temperatures in the glycerol/salt tortillas. Baik and Chinachoti, 2001 explained that it could be because of freezing point depression which was proportional to the amount of glycerol present. In Figure 3.17 it was in the order of 1–2˚C.

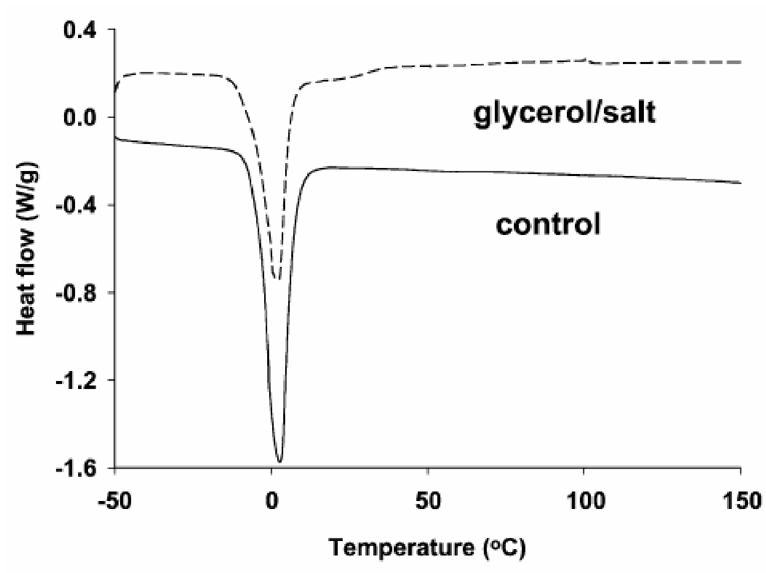


Figure 3 17 : Typical differential scanning calorimetry (DSC) thermograms for corn tortillas with and without glycerol or salt **(Vittadini et al, 2004)**

3.4.5 Propylene Glycol

Propylene glycol (propane-1,2-diol) is an organic molecule. It is generally a tasteless, odorless, and colorless clear oily liquid that is hygroscopic (hydrophilic) and miscible with water, acetone, and chloroform. It is manufactured by the hydration of propylene oxide. It can also be converted from glycerol, a biodiesel by-product.

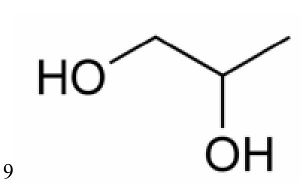
[9]


Figure 3 18 : Chemical structure of propylene glycol[14]

[9] http://en.wikipedia.org/wiki/Propylene_glycol

Propylene glycol is used as a moisturizer in, pharmaceuticals , cosmetics and food products, as an emulsification agent in Angostura and Orange bitters, as a solvent for relatively hydrophilic food colors and flavorings and as a humectant food additive.

3.5 Stickiness in Foods

Stickiness in tortillas can be defined as the difficulty in separating two or more tortillas. Stickiness is highly undesirable and reduces consumer's perception of the product. Increased mass production of tortillas has made it necessary to devise new ways to control stickiness in order to prevent manufacturing line shut-downs. Stickiness is an important issue both during processing (sticking of dough to press plates) and storage (sticking of packed products). Stickiness in dough reduces machinability (Ease in handling the dough in blender, dough ball divider, hot press). Sticky dough has low reduced MTI (mixing tolerance index), reduced dough stability and lower bread volume (Martin et al. 1986).

Stickiness is observed in cereal (dough and tortilla stickiness), confectionary (hygroscopic sugars turns sticky in moist environments), dairy (stickiness and caking of milk powder), and packaging (adhesion of food material in cans and packages) industries.

3.5.1 Factors Affecting Stickiness:

Table 3.4

Factors Responsible for Stickiness in Food Products and Their Relative Contribution

Factors	Relative Contribution to Stickiness
Protein	o
Polysaccharides	o
Fats	+
Low molecular sugars	++
Organic acids	++
Water/relative humidity	+++
Particle size distribution	+
Compression/pressure	++
Temperature	+++
Viscosity	+++

o, base point (negligible contribution); +, high contribution; ++, higher contribution; +++, highest contribution.

(Adhikari et al 2001)

3.5.2 Viscosity:

Viscosity of frozen tortilla samples were in the range of 10^8 -10^9 Pa.s as shown the following Figure 3.19. Upon heating the viscosity decreases sharply to 10^{5-6} Pa.s. This low viscosity of tortillas and hence increased molecular mobility could be one of the reasons responsible for stickiness. Kumar et al 1976 inversely correlated cooked rice stickiness with consistency.

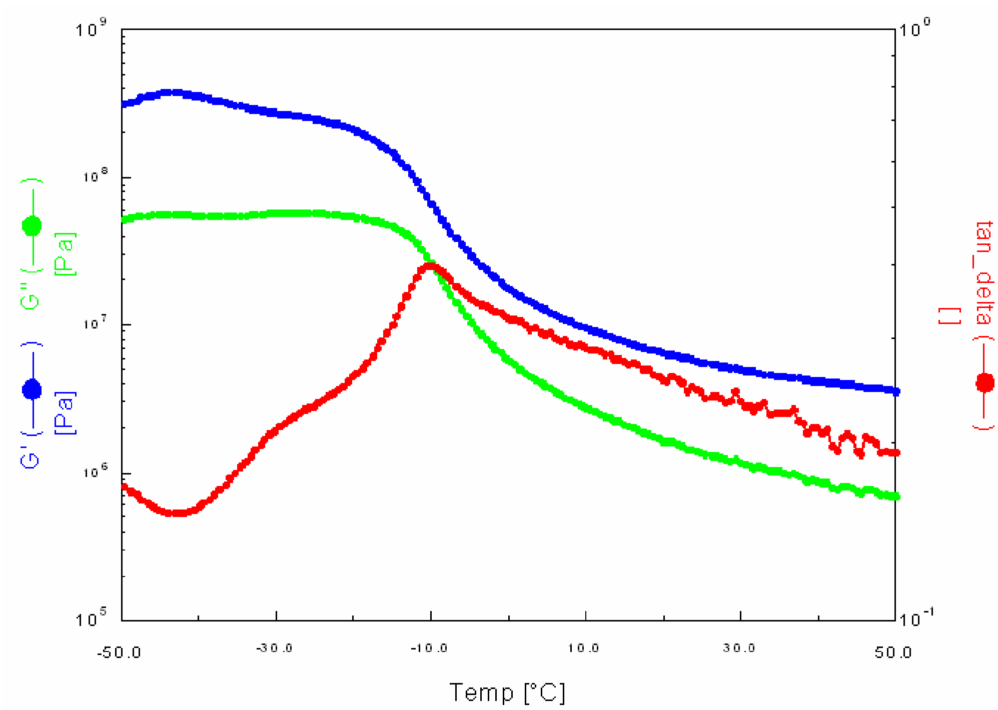


Figure 3 19 : Temperature sweep of a commercial tortilla with 0.94 water activity at frequency of 1 Hz and a heating rate of 5˚C/min

3.5.3 Water Content:

Water content is a catalyst for low moisture foods because it affects viscosity, surface tension and solvation of ingredients (Adhikari et al 2001). Water is a ubiquitous plasticizer and reduces Glass Transition Temperature (Tg) of low moisture food products (Slade et al 1991). Atkins, 1987 observed that *Tg* of food materials such as sugars, starch, gluten, gelatin, hemicellulose, and elastin decreases rapidly to about −10 ˚C or so when moisture content increases to 30% by mass. Slade et al., 1989 attributed, the rate of decrease of *Tg* to water plasticization as ~10˚C per 0.01g of water/g of material. Slade et al.1991 stated that plasticization of amorphous food powders and the subsequent depression of their glass transition temperature below ambient temperatures is related to stickiness. Water on food particle surfaces can reduce micro-roughness and allow them to come closer which leads to stickiness (Iveson, 1997).

Food product temperature determines phase/state and viscosity of food product. If product is stored at higher than its Tg, it shows liquid behavior and increased stickiness (Roos 1993). Griffith, 1991 observed that if powders are exposed to sufficient external compression, they are inclined to cake, meaning higher pressure or compression of a solid system stimulates stickiness and caking of food powders (Adhikari 2001).). Higher molecular weight (MW) water soluble ingredients have high Tg (Fox and Flory, 1950) that means lower MW ingredients with lower Tg values results in stickiness.

Adhikari et al. 2001 summarized the mechanism behind stickiness in food products as below.

3.5.4 Intermolecular forces:

Adhesion between two surfaces without material bridges is primarily attributable to *Fvdw* and electrostatic forces. The *Fvdw* can be calculated for two spheres of diameter *d*1 (*m*) and *d*2 (*m*) separated from each other by distance *x* (*m*), as:

$$F_{vdw} = \frac{E_p d_1 d_2}{16\pi(d_1 + d_2)x^2}$$

(Adhikari 2001)

Ep, the Van der Waals' interaction energy, is between 10^{-19} and 2×10^{-18} J. Van der Waals' force (N) is maximum when particles are in intimate contact with each other.

3.5.5 Electrostatic Forces:

Electrostatic forces occur where particles possess excess opposing charges. For an ideal electrical insulator the adhesion forces, *Fel,i,* (*N*) can be calculated using Coulomb's law as:

$$F_{el,i,} = \frac{\pi q_1 q_2 {d_1}^2 {d_2}^2}{\varepsilon_r \varepsilon (d_1 + d_2 + 2x)}$$

(Adhikari 2001)

where *q*1 and *q*2 are the electric charges per unit surface area of spheres (Coulomb.*m*−2) and εr (dimensionless) and ε (Coulomb2.*N*1.*m*−2) are the relative and absolute dielectric constants of the surrounding medium, *x* (*m*) is the distance of separation between the spheres.

3.5.6 Liquid Bridges :

Mobile liquid bridges are forces originating from mobile liquid between the particles. It is subdivided into three groups: pendular, funicular, and capillary. Pendular state is the one in which liquid has occupied only a part of the total voidage between the particles (Newitt et al 1958). In this state the liquid bridges have strength resulting from the pressure drop developed through curvature of the liquid meniscus and also from the interfacial tension exerted by the liquid along the wetted perimeter. Hence, liquid bridges result in a combined pull of the solid particles by these two forces (Rumpf, 1962; Newitt and Conway- Jones, 1958; Papadakis and Bahu, 1992). Assuming perfect contact between two rigid spheres, and perfect wetting, the tensile strength, *T* (Pa) of agglomerate coming from pendular water is given by (Newitt and Conway-Jones, 1958):

$$T = 2.8\frac{(1-\varphi)}{\varphi}\frac{\sigma}{d}$$

(Adhikari 2001)

where, σ is liquid surface tension ($N.m-1$), φ is porosity (dimensionless) of the agglomerate and d is the particle diameter (m).

Funicular and capillary states: When the void space between powder particles is completely filled by water extending to the edge of the pore and forming a concave surface, a negative capillary pressure is exerted in the entire liquid space boosting the tensile strength of the wet agglomerate, which is referred to as a capillary state (Papadakis and Bahu, 1992). The tensile strength of capillary water is given by:

$$T = 8.0\frac{(1-\varphi)}{\varphi}\frac{\sigma}{d}$$

(Adhikari 2001)

The funicular state is the transition between pendular and capillary state. In this state gas still occupies a small fraction in the water continuum.

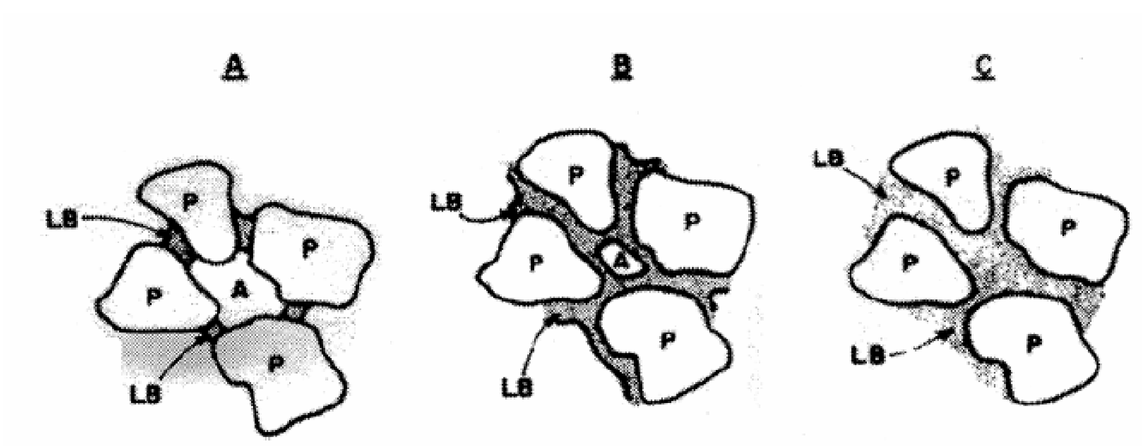


Figure 3 20 : Schematic diagram of liquid bridges. a) Pendular state, b) Funicular state, and c) Capillary state. P, particle; LB, liquid bridge; A, air (Peleg,1977)

Immobile liquid bridges involve a viscous binder between the particles which creates a strong binding force far stronger than the mobile liquid bridges. It can be observed in spray-drying of milk powders. The thermoplastic materials such as sugars at or above glass transition form immobile liquid bridges which lead to unwanted lumping. This type of liquid bridge retains a capacity to transform itself into solid bridges in subsequent drying. This mechanism of bridge formation is driven by surface energy (Downton et al., 1982).

3.5.7 Solid Bridges:

Rumpf (1962) observed that agglomerates can be formed through solid bridges by diffusion of molecules from one particle to another. Pietsch (1997) observed diffusion at the point of contact at elevated temperatures. When amorphous materials crystallize the solid dissolved in the liquid crystallizes and forms solid bridges at the points of contact. Rapid crystallization invariably leads to small crystals with strong bonds (Adhikari 2001).

Mechanical interlocking: Pietsch (1997) observed fibrous, bulky, and flaky particles can interlock or fold about each other resulting in “form-closed” bonds. It happens at elevated temperatures that causes reduced viscosity and molecules at the interface begin to flow into each other. When the temperature decreases interlocking forms between the particles (Griffith, 1991).

Primary chemical bonds: If the contact distance between two surfaces is smaller than 4×10^{-9} m, then chemical bonding (covalent, ionic, metallic, and hydrogen) can take place because of the primary valence forces. According to Allen (1993) adhesive force originated from chemical bonding is the strongest among all the bonding.

3.5.8 Surface Energy and Wetting:

When a liquid drop is placed on the surface of a solid, the shape of the droplet is determined by balance from the three forces of solid, liquid and vapor.

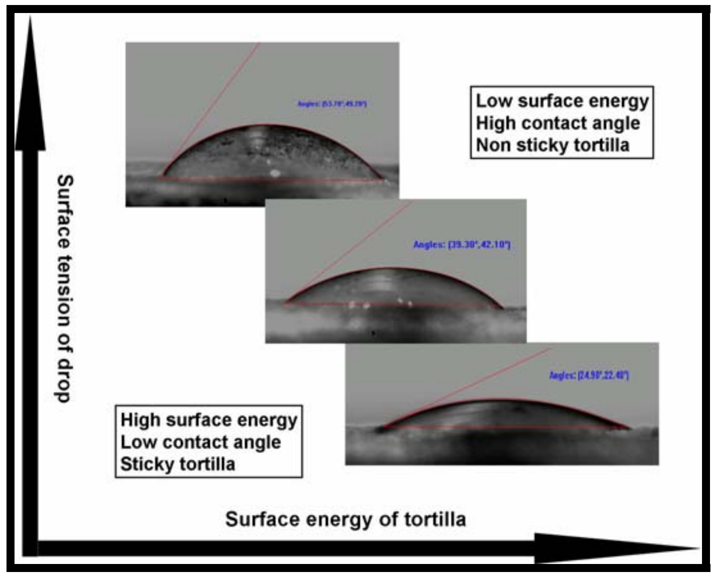


Figure 3 21 : Relationship between contact angle and surface energy of tortilla

The line tangent drawn at the curve of the droplet to the point it intersects the solid surface forms the contact angle. A droplet with high surface tension resting on a low energy solid forms a spherical shape or high contact angle. When the solid surface energy exceeds the liquid surface tension, the droplet forms a flatter, lower profile shape or low contact angle. The correlation of contact angle data with surface tension provides fundamental information for critical surface analysis. Contact angle data reflect the thermodynamics of a liquid/solid interaction and it is useful for characterization of the wetting behavior of a particular liquid/solid pair. Michalski et al (1997) observed that good wettability means the food and the adherent have a strong mutual affinity and are likely to adhere well. This mechanism is based on Young's force equation (Fowkes, 1964) and Dupre's energy equation (Michalski et al., 1997).

3.5.8.1 Surface Energy Calculation:

The contact angle is a measure of the balance between cohesion within the liquid and adhesion at the liquid/solid interface. When the surface forces are larger than the cohesion forces holding the drop together then the liquid spreads and when the converse is true then the drop stays together and a finite contact angle emerges. When a liquid drop is in contact with an ideally smooth, undeformable, homogeneous solid , it exhibits an equilibrium contact angle that can be expressed by Young's equation (Woodward 2000).

$$\gamma_{SV} - \gamma_{SL} = \gamma_{LV} \cos\theta$$	(1)

Where, γ_{LV} is the surface tension of the solid in equilibrium with its own vapor, γ_{SL} the interfacial tension between liquid and solid, γ_{SV} surface tension of the solid in equilibrium with the saturated liquid vapor, and θ the contact angle.

Contact angle is the work related to the competition between cohesive forces and adhesive forces and results in the work of adhesion (work to separate two surfaces):

$$W_a = \gamma_{LV}(1 + \cos\theta)$$	(2)

The work of adhesion can be determined, using a well defined liquid and measuring the contact angle on a solid surface. There are several methods to calculate the surface energy. Harmonic mean method (eqn. 3) is one of the most commonly used methods, where the surface energy is divided into separate components, dispersive (nonpolar) and polar (eqn 4). The method requires probe liquids whose component surface energies are well characterized.

$$(1+\cos\theta_L)\gamma_L = 4\left(\frac{\gamma_L^d \gamma_s^d}{\gamma_L^d + \gamma_s^d} + \frac{\gamma_L^p \gamma_s^p}{\gamma_L^p + \gamma_s^p}\right)$$	(3)
$$\gamma^{total} = \gamma^d + \gamma^p$$	(4)

Where γ_L^d and γ_L^p are the known dispersive and polar surface tension components of test liquid, and θ_L is the contact angle. γ_s^d and γ_s^p are the dispersive and polar components of surface energy of the solid. Since the equation involves two unknowns (γ_s^d and γ_s^p), at least two known surface tension testing liquids are required to calculate the surface energy of the solid using the harmonic mean method (Wu 1982, Shimizu 2000).

3.5.8.2 Example of Testing Liquids and Surface Energy Calculation using Wu's Harmonic Mean Method:

Formamide and DMSO are widely used to determine surface energy of paint and coatings.

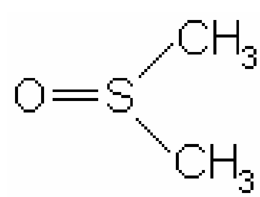


Figure 3 22 : Chemical structure of DMSO

DMSO is a colorless liquid and an important polar aprotic solvent. It dissolves both polar and nonpolar compounds and is miscible in a wide range of organic solvents as well as water. Surface tension of DMSO is 44 dyne/cm (dispersive and polar components 36 and 8 dyne/cm respectively).

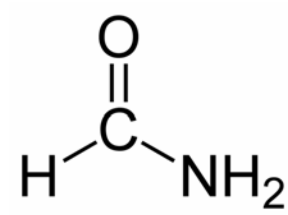


Figure 3 23 : Chemical structure of formamide

Formamide (methanamide) is an amide derived from formic acid. It is a clear liquid which is miscible with water. It dissolves many ionic compounds that are insoluble in water, so it is also used as a solvent. Surface tension of formamide is 58.2 dyne/cm (dispersive and polar components 39.5 and 18.7dyne/cm respectively).

3.5.8.3 Sensitivity analysis:

For Testing liquid (1) Formamide and (2) DMSO the harmonic equations are as below.

$$(1+\cos\theta)\,58.2 = 4\left(\frac{39.5\,\gamma_s^{d}}{39.5+\gamma_s^{d}} + \frac{18.7\,\gamma_s^{p}}{18.7+\gamma_s^{p}}\right)$$

$$(1+\cos\theta)\,44.0 = 4\left(\frac{36.0\,\gamma_s^{d}}{36.0+\gamma_s^{d}} + \frac{8.0\,\gamma_s^{p}}{8.0+\gamma_s^{p}}\right)$$

Table 3.5
Sensitivity Analysis of Formamide and DMSO

θ_1	θ_2	Dispersive	Polar	Total Surface energy
0	10	25.5	49.7	75.2
10		25.8	45.8	71.6
20		26.5	36.4	62.9
30		28.0	25.4	53.4
40		30.7	15.3	46.0
50		37.1	6.4	43.5
0	20	23.6	61.1	84.7
10		23.8	56.0	79.8
20		24.5	43.9	68.3
30		25.7	30.3	56.1
40		28.0	18.6	46.6
50		32.7	8.9	41.6
0	30	20.8	89.8	110.6
10		21.0	80.8	101.8
20		21.5	60.9	82.4
30		22.5	40.8	63.3
40		24.2	25.0	49.2
50		27.4	13.2	40.6
60		37.0	3.2	40.2
0	40	17.5	186.2	203.7
10		17.6	157.1	174.7
20		18.0	104.5	122.5
30		18.7	63.4	82.2
40		20.0	37.1	57.1
50		22.1	20.3	42.4
60		26.4	8.9	35.2

Tortilla surface is a low energy surface. Combination of formamide and DMSO gave reliable results as shown in above table.

3.5.8.4 Application of Contact Angle Analyzer:

Contact angle analyzer is used in the Bio-medical field to measure wettability improvement of contact lenses, bio-compatibility of human implants and micro-fluidity studies of bio-chips. It is used in interfacial chemistry to determine surface tension and wettability of detergents and surfactants. In the field of cosmetics it is used to study skin wettability and spreading of lotions. It is used in the painting and printing industry to evaluate ink spreading and adhesion studies. It is also used to understand the lotus effect on surface nano-structure.

3.5.9 Glass transition Mechanism:

Glass transition temperature (Tg) is a fundamental property of an amorphous material. The crystalline portion remains crystalline during the glass transition. At a low temperature the amorphous regions of a polymer are in the glassy state. In this state the molecules are frozen without any segmental motion. In the glassy state, the motion of the red molecule in the schematic diagram at the right would not occur. When the amorphous regions of a polymer are in the glassy state, it generally will be hard, rigid, and brittle.

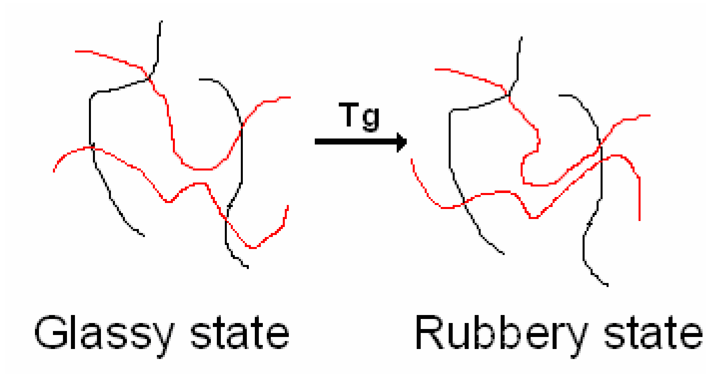


Figure 3 24 : Motion of Semicrystalline amorphous material due to heating[10]

When a glassy material is subjected to moisture and heated it approaches the phase transition (Glass transition). At this temperature portions of the molecules can start to wiggle around as illustrated by the red molecule in the diagram above. The polymer now is in its rubbery state. The viscosity of the tortilla samples decreases dramatically as shown in Figure 3.19 from 10^{8-9} Pa.s to 10^{6-7} Pa.s. The rubbery state lends softness and flexibility to a polymer. This reduced viscosity is unable to support the glassy microstructure giving way to structural collapse causing stickiness (Wallack and King, 1988). Hence a glass transition approach can be successfully used to characterize stickiness in food (Adhikari et al. 2001) Unwanted agglomeration and caking of the food powder can be avoided simply by storing them below their glass transition temperature without moisture absorption from the air (Slade et al. 1993). Phase transitions in foods are not limited to glass transitions.

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[10] http://faculty.uscupstate.edu/llever/Polymer%20Resources/GlassTrans.htm

3.5.10 Stickiness in Tortillas

Stickiness in tortillas has not been well studied and there is very little literature available on the subject. There is a need for fundamental analysis to find out the physical, chemical and physico-chemical causes of stickiness in tortillas. There is no objective method available to measure stickiness in tortillas. Stickiness in tortilla is measured subjectively. In this study we tried to develop an objective test to measure tortilla stickiness.

According to Dr. R. D. Waniska (1999) the primary cause for stickiness in tortilla is moisture migration from the baked tortilla to its smooth surfaces. His qualitative explanation provides a lot of good insight for some of the hypotheses in this thesis. Moisture migration in fact can cause a phase transition and make the surface material rubbery and free flowing resulting in stickiness. He suggested efficient cooling before packaging tortillas which facilitates removal of heat and moisture and/or allows more time for structure of tortillas to stabilize and equilibrate. We also concur with this suggestion since it builds structure and increases the surface viscosity reducing stickiness. He further observed that less severe hot-press conditions (time, temperature, pressure) would yield tortillas with less smooth surfaces, which have less tendency to stick.(Waniska 1999).

Stephen Bright from AB Mauri Ingredients, Inc presented a seminar on "*The Basics & More: Flour Tortilla Trouble Shooting*" at 17th annual Tortilla Industries Association meeting. He discussed the effect of processing conditions and ingredients on tortilla stickiness. According to him over-baking (long time and high temperature) is a major precursor to sticking. It leads to pillowing or puffing which enhances stickiness in tortillas. We concur with this observation as we will discuss later because excessive temperature and moisture causes high gelatinization, some caramelization and the formation of thin sticky blisters that adhere to each other very easily. Other factors which can increases stickiness are moisture migration due to temperature shift and over-packing (excessive weight). He observed that higher levels of L-cysteine and sodium metabisulfite weakens protein by reducing disulfide bonds and making the structure more fluid and crust resilience and increases the occurrence of sticking in tortilla. He suggested maintaining cooler conditions at 40˚F and 80% Relative humidity for tortilla cooling after baking. He also stated that certain enzyme cocktails may contain incorrect enzyme activities breaking down the carbohydrate and protein matrix, liquefying parts of it and causing a surface sticky mass in particular that enhance surface adhesion of the tortillas. On the ingredient front, higher percentages of fat >7 % dilute the gluten-forming protein and thus reduce stickiness. Use of higher MP (melting point) or saturated fats (higher solids at room temperature) might reduce stickiness because liquid oils remain liquid and enhance surface to surface adhesion however solid fats solidify after processing cause surface lubrication and reduce stickiness. The effect of such strategies on the palatability of the product remains unknown.

IV. HYPOTHESIS

Several factors create stickiness in tortillas. We believe that combined effect of moisture content, water activity, glass transition temperature and surface free energy of tortilla results in stickiness.

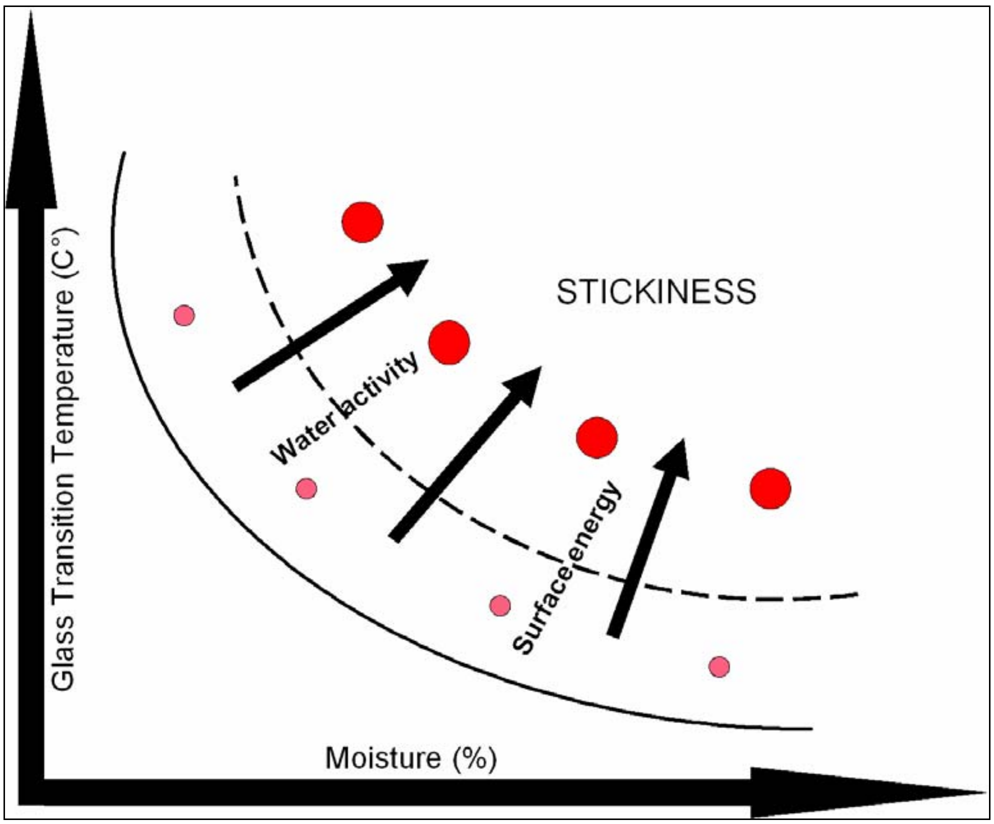

Figure 4 1 : Schematic representation of hypothesis

We expect that any phase change which affects mobility (availability and movement of water) increases surface energy and will have an impact on stickiness. This study hypothesizes that stickiness in tortillas is the result of high surface energy and in particular high polar surface forces which are affected by the presence of moisture and changing phase behavior as a function of relative humidity and temperature at different compositions as shown in figure 4.1.

This study hypothesizes that hydrophilic ingredients such as gums (CMC and XG) and polyols (glycerol and propylene glycol) will compete for water in tortillas and reduce water activity and surface energy. This will result in tortillas with reduced stickiness.

This study also hypothesizes that processing conditions such as tortilla baking temperature, tortilla cooling time, and dough resting or proofing time decides the moisture content, water activity and hence wettability of the tortilla surface. Study of these factors can help create a method to produce tortillas with the least stickiness.

V. MATERIALS AND METHODS

5.1 Basic Recipe for Wheat Flour Tortillas Prepared in Our Lab

Enriched, bleached all-purpose flour with 10% protein was used to prepare hot-pressed wheat flour tortillas.

Figure 5 1 : Nutritional information of Gold Medal wheat flour used in this study

The base tortilla formula is 300 g of bleached all purpose wheat flour (Gold Medal, General Mills Inc.), 4.5 g of salt, 1.8 g of baking powder, 18 g of shortening (all purpose), 0.48 ml of formic acid and 1.8 g of potassium sorbate. Tortillas were made according to the method of Bello et al. (1991) with some modifications. Dry ingredients were mixed for 2 min with a KitchenAid Hobart mixer (figure 5.2) and another 6 min after shortening was added. Shortening was liquidified by microwaving for 45 seconds before adding to the dry mixture. 156 g distilled water (heated to 35°C) was added to the mixture and the mass was mixed for another 5 min. After resting at 35°C for 5 min the dough was divided and rounded using hands into 43 g balls. Dough balls were hot pressed at 350±10°F for 3 to 4 seconds (Villaware model V5955) and baked in an electric pizza oven (Adamatic Hobart) at 350°F or 450° F for 55 seconds as shown in Figure 5.4. The temperature of the oven was measured using a type J-K-T thermocouple (Microprocessor thermometer, Model HH21) to better control processing conditions. Cooked tortillas were cooled to room temperature and stacked in polyethylene (PE) bags for 2 days before they were analyzed.

Using the above basic recipe for tortilla-making we applied a multi-pronged strategy to understand the origins of stickiness in tortillas and devise strategies to reduce it and improve tortilla characteristics.

Strategy 1. Methodology development: An objective method to quantify tortilla stickiness was developed using the TAXT2i texture analyzer. Tortilla samples were manufactured suitable to use contact angle analysis technique for measuring surface energy.

Strategy 2. Origins of tortilla stickiness: Various methods were used to measure properties of tortillas and correlate them to tortilla stickiness**.** Commercial wheat tortillas with a wide range of stickiness scores as indicated by sensory panels were selected and equilibrated to different water activity (Aw) levels (0.12-0.97). Moisture sorption isotherms were developed. Differential scanning calorimetry was used to determine the amount of freezable water present in the tortillas. Mechanical spectroscopy was used to characterize the phase behavior of tortillas. The surface free energy of tortillas was measured using contact angle measurements. Wide-angle x-ray scattering (WAXS) was used to measure relative crystallinity of tortilla samples. X-ray microtomography was used to find tortilla cellularity.

Strategy 3. Utilize ingredients which compete with water: Tortillas were produced with a variety of GRAS ingredients to reduce stickiness and surface energy in tortillas after we demonstrated that surface energy was an excellent marker for stickiness. Specifically, we added ingredients that favorably compete with water. Xanthan gum and carboxymethylcellulose (CMC) were used alone and in combination. We also used glycerol and propylene glycol alone and in combination with the gum blend of xanthan gum and CMC. We used gums (CMC and xanthan gum) and polyols (glycerol and propylene glycol) alone and in combination to test their effectiveness in controlling the aw in wheat flour tortillas. These will be added to the basic recipe at levels of 0.5% for gums and 4% for polyols:

a. (CMC + Xanthan Gum) @ 0.5 %

b. (CMC + Xanthan Gum) @ 0.5 % + Glycerol @ 4 %

c. (CMC + Xanthan Gum) @ 0.5 % + Propylene Glycol @ 4 %

Strategy 4: To understand the effect of processing conditions on stickiness, tortillas were prepared using different combinations of dough resting times (10 and 20min), baking temperatures (350 and 450°F), and cooling times after baking (2, 5 and 10min).

Strategy 5: A shelf life study was conducted to study the effect of storage conditions on tortilla texture. Tortillas were made and stored at freezer, refrigeration, and room temperatures.

Figure 5 2 : Blender used to make dough

Figure 5 3 : Control dough made using modified Bello et al 1999 method

Figure 5 4 : Hot-press and baking oven in Dr. Kokini's lab, Food Science Department, Rutgers University, NJ

Strategy 6: Validation tests to confirm that the findings obtained in model laboratory tortillas could be extended to manufacturing plants were carried out at Casa De Oro Foods pilot plant, on March 12, 2007. Eight different trials were carried out to understand the effect of the addition of gums and polyols and processing conditions on tortilla stickiness. Tortilla samples were packaged in polyethylene bags and transported to Rutgers University, NJ to carry out confirmation tests. Tortillas were stored at room temperature for one day and were analyzed for stickiness (by both sensory and instrumental measurements), moisture and freezable water. In addition the cellularity of the tortillas was measured.

5.2 Basic Recipe for Wheat Flour Tortillas Prepared at Casa De Oro Foods, NE

Bleached flour was used to prepare hot-pressed wheat flour tortillas. The base tortilla formula is shown in Tables 1, 2 and 3. Tortillas were made according to the method of (Bello et al. 1991) with some modifications. Dry ingredients were mixed for 2 min with a Hobart mixer and for another 6 min after shortening was added. Tap water (heated to 97°F) was added to the mixture and mixed for 5 min. The temperature of dough after mixing was 87.5°F. The dough was divided and rounded into 43 g balls using a Dutchess dough divider after resting at 91°F for 5 min. Dough balls were proofed (Dough resting) for either 10 or 20 min with 75% RH. Dough balls were hot pressed and baked using Lawrence Equipment. Four dough balls were pressed at a time using a hot press. Upper and lower presses were adjusted to a temperature of 350°F. A pressure of 950 psi was used to form dough discs. Dough discs were then passed through a gas-fired oven. The oven had three belts, but only the first two belts were heated. The temperature of upper belt was kept either 350°F or 450°F. Lower belt was maintained at 380±15°F. Baking time was kept at 30 sec instead of 55 sec used in our laboratory. This was done because when the baking time was kept at 55 sec at 450 F the tortilla surface showed intense burning. Cooked tortillas were then allowed to pass through a cooling tower (Lawrence Equipment). The cooling tower had seventeen belts and was set to give 5 min room temperature cooling. The temperature of room was 25- 27°C. The humidity of room was 27%. Tortillas were packed in polythene bags and evaluated for stickiness using sensory and instrumental analysis.

The experiment design for the tortillas made is as follows:

1. Control (450_20_5) with 450°F baking temp, 20 min dough resting, 5 min cooling
2. Control (350_10_5)
3. Control (350_20_5)
4. Control (450_20_10)
5. Tortilla added with glycerol (450_20_5)
6. Tortilla added with glycerol with 10 min cooling (450_20_10)
7. Tortilla added with gums and glycerol (450_20_5)
8. Tortilla added with gums and propylene glycol (450_20_5)

Table 5.1
Formula for Control Tortillas

RUTGER'S CONTROL — CONFIDENTIAL — DATE: 3/5/2007

INGREDIENT	Formula %	Bakers %	Lbs.	Grams	#1	#2
Flour	62.166	100.000	4.26	1933.296		
Water	32.326	52.000	2.21	1005.314		
Shortening, NovaLipid	3.730	6.000	0.26	115.998		
Glycerine	0.000	0.000	0.00	0.000		
Xanthan Gum	0.000	0.000	0.00	0.000		
CMC 15	0.000	0.000	0.00	0.000		
Salt	0.932	1.500	0.06	28.999		
Baking Powder	0.373	0.600	0.03	11.600		
Potassium Sorbate	0.373	0.600	0.03	11.600		
Fumaric Acid	0.099	0.160	0.01	3.093		
Propylene Glycol	0.000	0.000	0.00	0.000		
TOTAL	100.000	160.860	6.85	3109.90		

CONFIDENTIAL

6": 43g d-ball 1548g / 36ct

Figure 5 5 : Blender (3 speed control with timer, Hobart) at Casa De Oro Foods

Figure 5 6 : Dough divider (Dutchess)

Figure 5 7 : Tortilla press, oven and cooling chamber (Lawrence Equipment, OD0195-02)

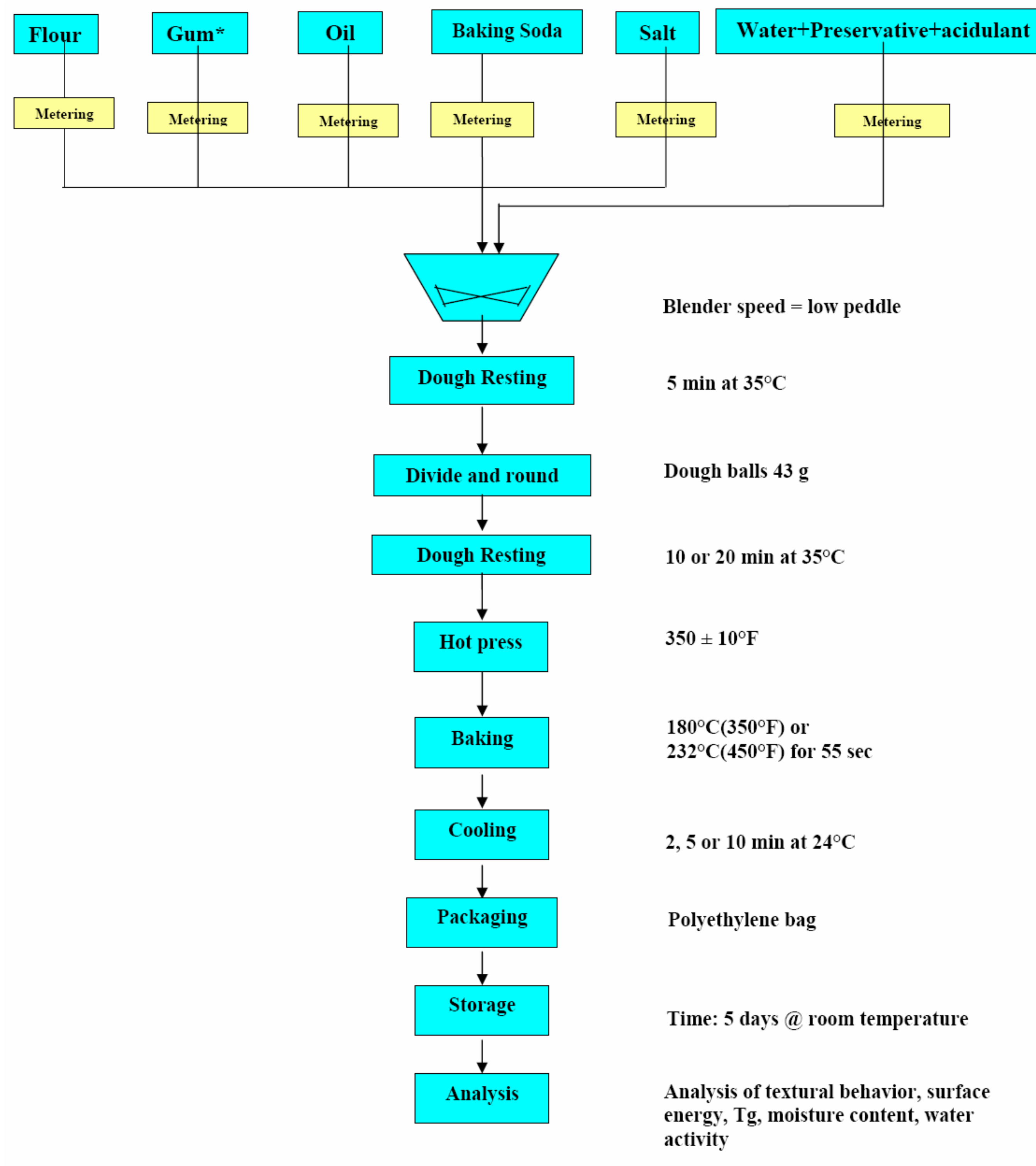

Figure 5 8 : Process flow diagram for tortilla making[11]

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[11] Modified Bello et al 1991method

5.3 Sample Preparation at Food Science Department, Rutgers University,NJ.

i. Identification of the factors which affect stickiness

Six commercial tortillas were selected from a local super market (Stop & Shop). Tortillas were coded as T1 to T6 depending on increasing order of perceived sensory stickiness. The stack of tortillas was removed from the product package two by two in order to keep adjacent tortillas in contact with each other as they were in the package originally. Tortillas were cut into the shape of the rectangular probe used for stickiness measurement with the TAXT2 and left in desiccators for equilibration at water activity levels of 0.75, 0.84, 0.92, 0.94 and 0.97. Tortilla slices were handled carefully in order to prevent detachment of tortilla layers from each other during cutting. The samples to be used for surface energy measurements were also equilibrated in the desiccators. In order to eliminate the variation in contact angle measurements due to surface roughness, tortilla samples were compressed at 200 kg pressure for 80 seconds using the texture analyzer. Samples were then kept in the desiccators for additional 3 days and surface energy measurements were performed. To measure glass transition temperature flour tortillas with higher stickiness and lower instrumental stickiness were selected. Tortillas were cut into rectangular shape (45mm x 12mm) and kept in desiccators until they equilibrated to water activity levels of 0.13, 0.33, 0.46, 0.84, and 0.92.

5.4 Sensory Stickiness

Sensory evaluation of commercial tortilla samples were done by 8 semi-trained panelists using the method of magnitude estimation or ratio scaling (Stevens 1975). This method consists of assigning relative scores to relative magnitudes of sensations, so that the ratio of scores equals the ratio of perception of a particular sensation. (Kokini 1977). Panelists' ages ranged from 22 to 55. Panelists were asked to choose one of the tortillas as a reference, peel it from the stack, and assign a number characteristic as their assessment of stickiness. For example if they assign 10 to the reference tortilla and the second sample feels three times as sticky, they should assign 30 to the second sample. The magnitude estimates were averaged and normalized following the procedure described by (Kokini 1977). First, a logarithmic transformation is performed:

$$W_{ij} = \log S_{ij}$$

Where $\mathbf{S_{ij}}$ is the *i*th individual score of panelist *I*, for the *j*th tortilla sample. Scores were then averaged to correct for variations in individual scales.

$$\mathbf{X_{ij}} = \mathbf{W_{ij}} - \sum_{\mathbf{j=1}}^{\mathbf{J}} \mathbf{W_{ij}} / \mathbf{J}$$

Finally, to correct for variations in panelist judgment (variation among subjects is not of interest here):

$$\mathbf{Y_j} = \sum_{\mathbf{i=1}}^{\mathbf{I}} \mathbf{X_{ij}} / \mathbf{I}$$

Details of the statistical procedure are described by (Stevens 1975) and (Kokini 1977).

5.5 Dough Stickiness Measurement using Chen-Hoseney Dough Stickiness Cell

A Chen-Hoseney (1995) dough stickiness cell was used for quantification of dough stickiness. In this method, the internal screw was rotated to move the piston and increase the sample chamber to its maximum capacity. A small quantity of prepared dough was placed into the chamber and the excess dough was removed with a spatula so that it was flush with the top of the chamber. The extruder lid was screwed on. The internal screw was rotated to extrude a small amount of dough through the holes and the first extrusion from the lid surface was removed using a spatula. The screw was once again rotated to extrude a 1mm high dough sample. The prepared dough surface was allowed to rest for 30 seconds to release the stress produced by extrusion. After this time the cover was removed and the cell was placed directly under the 25 mm cylinder probe attached to the load cell. The adhesive test was then performed. After that the dough was removed from the lid surface and extruded again to repeat the test, using the procedure below:

TA-XT2i Settings:

Option: Adhesive Test
Pre-Test Speed: 0.5 mm/s
Test Speed: 0.5 mm/s
Post-Test Speed: 10.0 mm/s
Distance: 4mm
Force: 250g
Time: 0.1s
Trigger Type: Auto - 5g
Data Acquisition Rate: 500pps

Accessories: 25mm Perspex cylinder probe (P/25P) using 5kg load cell

SMS/Chen-Hoseney Dough Stickiness Cell (A/DSC)

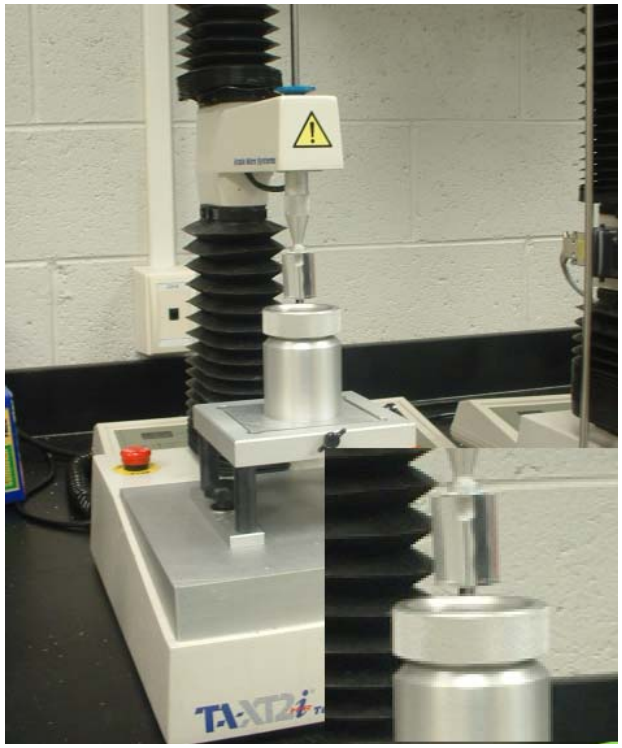

Figure 5 9 : Chen-Hoseney Dough Stickiness Cell

5.6 Textural Measurements

5.6.1 Instrumental Stickiness Measurement:

A TA.XT2i texture analyzer with the pasta stickiness rig attachment (Figure 5.10) was used to measure the instrumental stickiness of tortilla samples. The rectangular probe (Area=1860 mm^2) was attached to the 5 kg load cell and lowered into the retaining plate of the heavy-duty platform. Double-sided tape (Scotch) was applied to both the heavy-duty platform and rectangular probe to keep the tortilla layers stationary. Two tortillas were removed from the package and cut into 1860 mm^2 rectangular shape without separating them and positioned onto the platform directly under the probe. Care was taken to keep tortilla dimensions constant. The probe was set to apply a compression force of 2000 g to the sample for 20 seconds to achieve a good probe-sample contact before withdrawing at maximum speed (10 mm/sec).

Double-sided adhesive tape applied on the platform affixed the lower tortilla while the upper tortilla became attached to the probe during 20 sec compression time. When the probe was withdrawn the upper tortilla separated from the lower tortilla and the force required to separate two tortillas was registered by the load cell in g. as shown in the figure 5.11.

TA-XT2i Settings:

Mode: Measure Force in compression
Option: Return To Start
Pre-Test Speed: 1.5 mm/sec
Test Speed: 1.0 mm/s
Compression force: 2000g/20sec
Post-Test Speed: 10.0 mm/s
Trigger Type: Button
Data Acquisition Rate 400pps

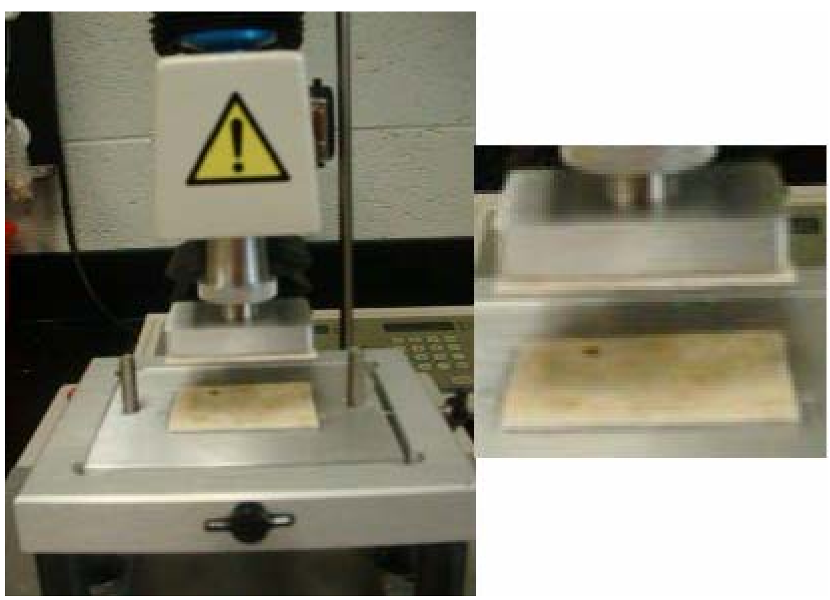

Figure 5 10 : TA.XT2i texture analyzer with pasta stickiness rig

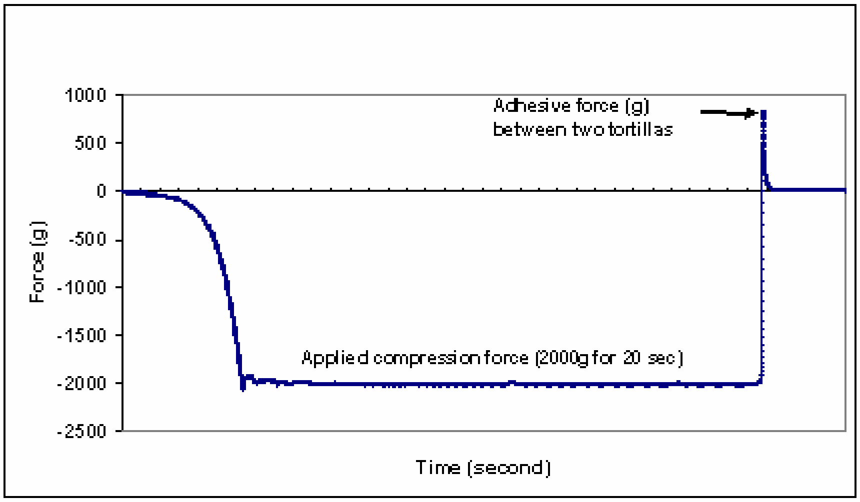

Figure 5 11 : Typical Texture Expert Exceed plot for the stickiness test

Figure 5.11 illustrates the resulting force-time curve. The instrumental stickiness was measured as the maximum peak force to separate the probe from the sample's surface (the higher the force value, the stickier is the sample)

5.6.2 Resistance to Extension Force and Extensibility Measurements

TAXT2i texture analyzer was used to measure tensile force, extensibility and gradient (ratio of force to distance) of tortillas. The upper tensile grip was attached to the load cell carrier and the lower tensile grip was secured to the base of the machine. Tensile grips were calibrated to start from a set distance apart for each test (20mm). Tortilla strips (45 mm × 25 mm) were tightened to tensile grips and tensile strength tests were performed. The TA-XT2i test setting was kept as shown below.

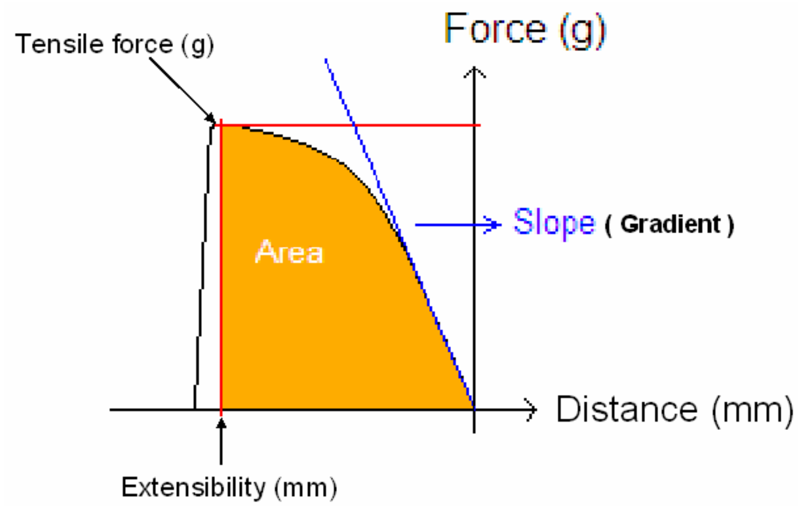

Figure 5 12 : Typical Texture Expert Exceed plot for Tensile Test

Earlier tortillas were prepared using the modified method of Bello et al (1991) and samples were packaged and stored at three different temperatures: room temperature (22 ± 1°C); freezer temperature (-19 ± 1°C); and refrigeration temperature (3 ± 1°C). A Fisher scientific vacuum oven model-228A was used to maintain room temperature. For refrigeration and freezer temperatures a Kenmore refrigerator was used. Tortillas stored at lower temperatures were thawed at room temperature (22 ± 1°C) for 5 hrs before testing. Temperatures were measured using a FISHERbrand 5213 P thermometer. A minimum of ten tests were performed and results were reported as averages of these tests.

TA-XT2i Settings:

Mode: Measure Force in Tension
Option: Return To Start
Pre-Test Speed: 3 mm/sec
Test Speed: 1.0 mm/s
Post-Test Speed: 10.0 mm/s
Trigger Type: Button
Data Acquisition Rate 400pps

Figure 5 13 : TA-96 Tensile Testing

5.7 Water Activity Measurements:

Water activity of tortilla samples were measured at 24°C using an Aqualab water activity meter. The tortilla was divided into small pieces to increase the sample surface area immediately before water activity measurement. Results were reported as the average of three measurements. Tortilla samples were cut into 35 × 10 mm strips and kept in a 100 ml glass beaker. These beakers were then kept in desiccators over saturated salt solutions until samples were equilibrated. The salt solutions used in this study were LiCl, $MgCl_2$, KCO_3, $NANO_2$, NaCl, KCl, $BaCl_2$, KNO_3 and K_2SO_4 with water activities of 0.11, 0.33, 0.43, 0.64, 0.75, 0.84, 0.9, 0.93 and 0.97 respectively. Toluene was kept in desiccators with water activities of 0.64 or more to suppress mold growth.

5.8 Determination of Amount of Freezable Water Present in Tortillas using Differential Scanning Calorimetry (DSC)

The amount of freezable water present in tortillas was analyzed using a TA 4000 Thermal Analysis System with a DSC 30-S Cell/TC11 TA Processor (Mettler Instrument Inc., Hightstown, NJ) and DSC823^e, METTLER TOLEDO STAR^e System. In this test an empty aluminum crucible was used as a reference and calibration of the instrument was performed using indium as a standard. The samples were scanned at a temperature range of -50˚C to 50˚C and a thermogram was obtained. The heating rate was 10˚C/min. Presence of freezable water in the tortilla sample was measured. This is a measure of free water, which is hypothesized to cause stickiness. The amount of freezable water and the instrumental stickiness of tortilla were then correlated.

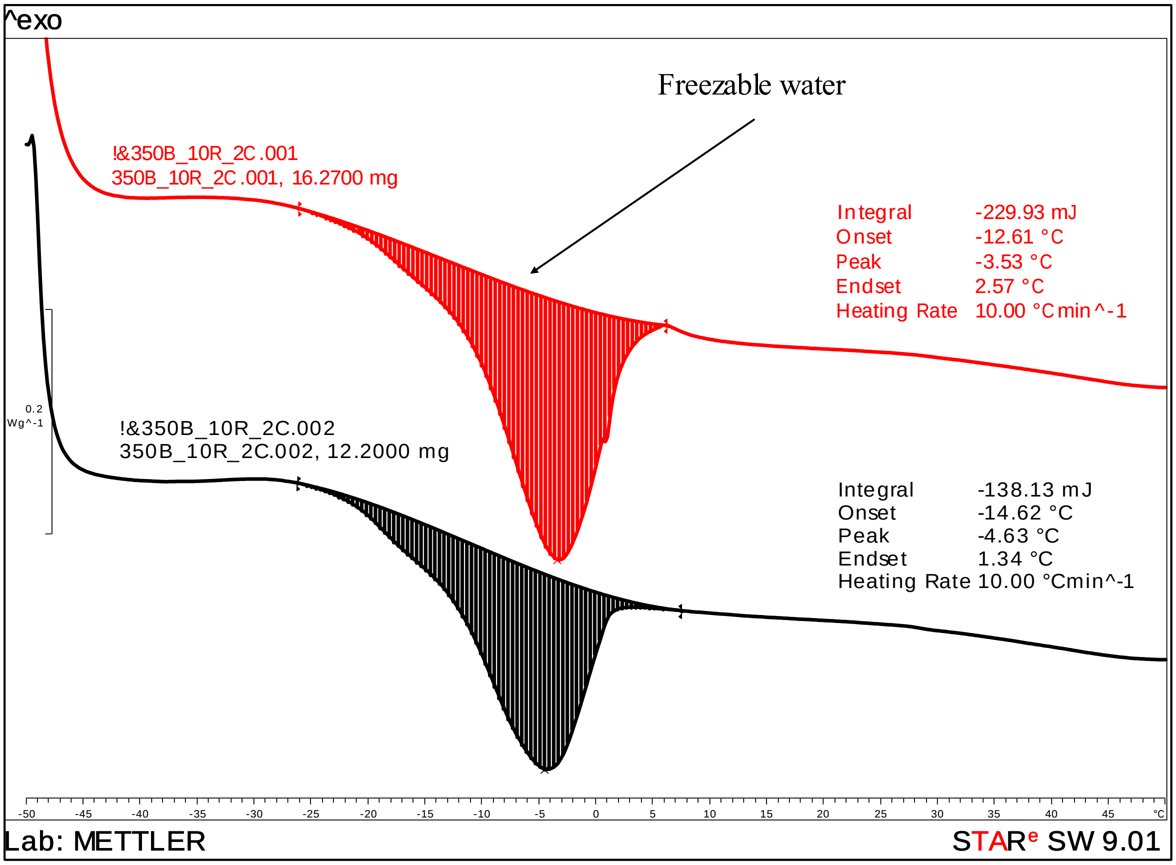

Figure 5 14 : Thermo gram of tortillas baked at 350°F with 10 min dough resting and 2 min cooling time using DSC823 e

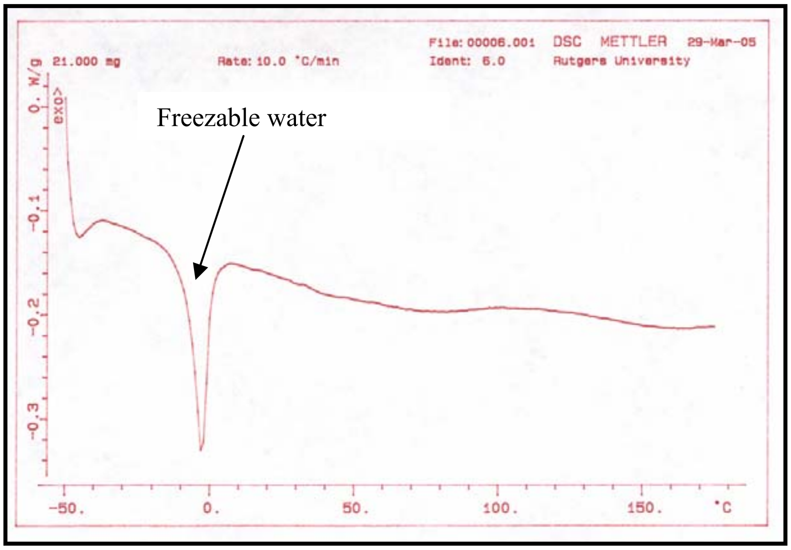

Figure 5 15 : Typical DCS thermo gram of tortilla made using modified Bello et al method

5.9 Glass Transition Temperature (Tg) measurements

Glass transition temperatures of flour tortillas equilibrated to different water activity levels were determined using mechanical spectroscopy. Both Advanced Rheometrics Expansion System (ARES) and a Rheometrics Solid Analyzer (RSA II) were used. ARES was used for flexible tortilla samples having high water activity (Aw≥0.84). In case of hard and brittle samples which is typical for lower water activity levels RSA II was observed to give more reliable results. Torsional rectangular geometry was used to clamp samples in the rheometer. Initially strain sweep tests were done to determine the linear viscoelastic region at particular water activity levels. A strain of 0.05% was applied for tortilla samples of 0.13, 0.33, 0.43 and 0.64 water activity. The strain level for high water activity samples (Aw= 0.84 and 0.92) was 0.1%. A higher strain level was selected to attain the linear viscoelastic region because tortilla samples were resilient at higher water activities. Tortilla samples were covered with grease and heated in an insulated chamber to avoid moisture loss during heat. The temperature ramp test was conducted at a temperature range of -50 to 150°C, heating rate of 5°C/min, and frequency of 1 Hz to find the glass transition temperature.

A temperature range of -50 to 50°C was selected for tortillas at higher water activity and expected to have lower Tg values. In cases of low Aw tortillas a temperature range of 30 to 150°C was used. The higher temperature range was selected because tortilla samples at low water activity were stiff. Plasticization effects of water on tortilla samples were lower and a higher Tg value was expected. Also water in tortilla samples was in the bound water region and water loss during the experiment was minimal.

Test Set Up:

```
Save Data As = 350 20 10.003
Operator = ADMINISTRATOR
AutoSave on; Automatic Data Save at End of Test = On
Geometry Type = Torsion Rectangular (Tors Rect)
      Length = 29.53 [mm]
      Width = 11.45 [mm]
      Thickness = 1.71 [rum]
Read Test Fixture Gap = Off
Tool Serial Num = 0000
Tool Inertia = 0.0 [g·cm2]
Change Gap to Match Tool Thermal Expansion = Off
Tool Thermal Expansion Coefficient = 0.0 [•m/˚C]
Fluid Density = 1.0 [g/cm3]
Test 'type = Dynamic 'temperature Ramp (D'tempRamp)
Frequency = 6.2832 [rad/s]
Initial Temp. = -50.0 [˚C]
Final Temp. = 50.0,0.0,0.0,0.0,0.0,0.0,0.0,0.0 [˚C]
Ramp Rate = 5.0,0.0,0.0,0.0,0.0,0.0,0.0,0.0 [˚C/min.]
Computed Ramp Time = 20:00, 0, 0, 0, 0, 0, 0, 0 [h:m:s]
Soak Time After Ramp = 60, 0, 0, 0, 0, 0, 0, 0 [s or h:m:s]
Time Per Measure = 5, 0, 0, 0, 0, 0, 0, 0 [s or h:m:s]
Strain = 0.1, 0.0, 0.0, 0.0, 0.0, 0.0, 0.0, 0.0 [%]
Computed Test Duration = 21:00 [h:m:s]
ZoneTime = 1260, 0, 0, 0, 0, 0, 0, 0 [S or h:m:s]
Options = Steady PreShear
```

```
Steady PreShear = Off
PreShear Mode = preshear Off
Delay Before Test = Off
Automatically start test when on Temperature = Off
AutoTension Adjustment = On
Mode = Apply Constant Static Force
AutoTension Direction = Tension
Initia1 Static Force = 0.0 [N]
AutoTension Sensitivity = 0.098066 [N]
When Sample Modulus <= 1.00e+06 [Pa]
AutoTension Limits = Default
Max Autotension Displacement = 3.0 [mm]
Max Autotension Rate = 0.01 [mm/s]
AutoStrain = Off
Strain Amplitude Control = Default Behavior
Limit Minimum Dynamic Force Used = No
Minimum ~pplied Dynamic Force = 1.0 [gmf]
Measurement Options = Default Delay Settings
      Cycles = 0.5 []
      Time = 3 [ s or h:m:s]
Correlation: One Cycle Correlate = Off
ElectroRheology Mode = Off
Turn OFF Motor = No
Turn Hold ON = Yes
Turn OFF Temp Controller = No
Set End of Test Temp = Yes
Set End of Test Temp to: = 25.0 [˚C]
Oven Air/N2 Switch = No Change
DieLectric Testing = Off
Steady Stress on Dynamic = 0.0 [Pa]
Analog Data Collection = Off
```

In mechanical spectroscopic techniques, the experimental Tg is determined from the change in storage moduli as a function of temperature either as the inflection point in storage modulus (G′ or E′) as a function of temperature or as the maximum in the loss modulus (G″ or E″). The tan δ peak (tan δ=G″/G′ or E″/E′) is also used to identify the Tg (Kokini *et al*., 1995; Cocero and Kokini, 1991). This study considered the temperature at the midpoint of the drop in storage modulus as the glass transition temperature (Tg). Results were reported as the average of two replicate measurements.

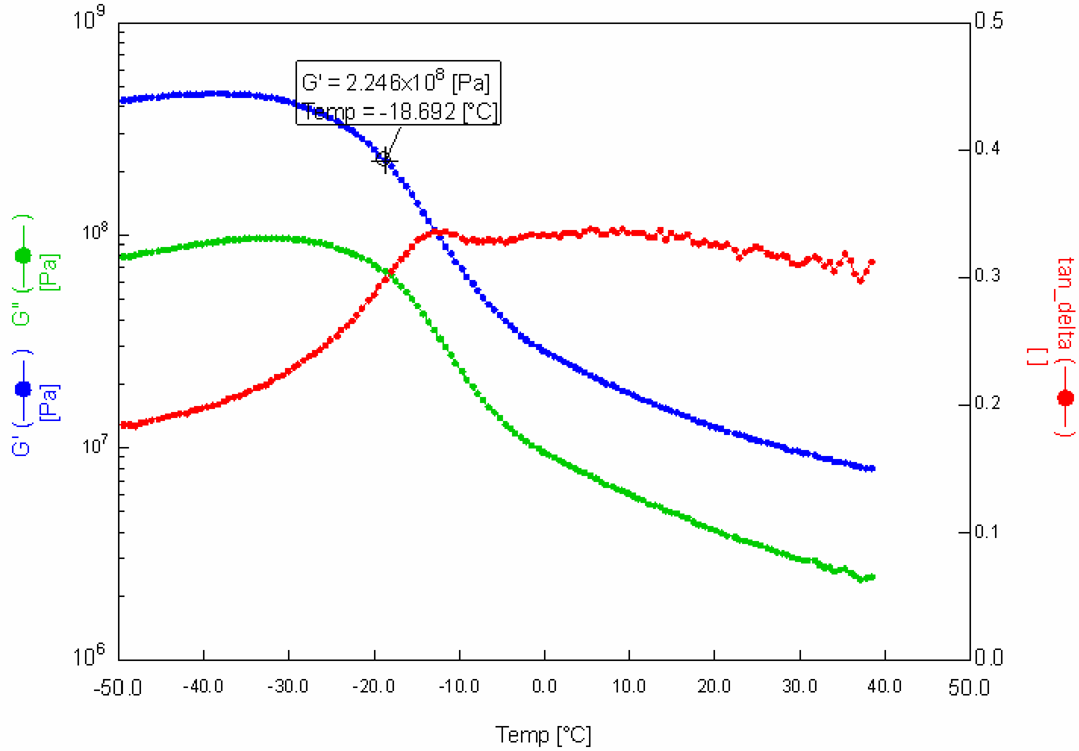

Figure 5 16 : Temperature sweep of a sticky tortilla at 0.92 water activity measured as the drop in the storage modulus in small amplitude measurements

5.10 Contact Angle Measurements

Dynamic Contact Angle and Surface Tension (pendant drop) System (VCAoptima) was used to determine the surface free energy of equilibrated commercial tortilla samples. Formamide and dimethyle sulfoxide (DMSO) were used as test liquids in order to make the calculations needed for the harmonic mean method (Table 5.2).

A drop of liquid was placed on the surface of the tortilla sample and the contact angle was determined as the line tangent drawn at the curve of the droplet to the point it intersects the sample surface after 5 seconds.

The surface free energy of tortilla samples was calculated using the harmonic mean method (from Eqn 3) below. The polar and dispersive component of the surface free energy was measured by the SE 2500 software available with the VCAoptima contact angle analysis instrument.

5.11 Table 5.2

Properties of the Test Liquids Used

Types of Liquids	Surface Tension	Dispersive	Polar
Formamide	58.2	39.5	18.7
DMSO	44.0	36.0	8.0

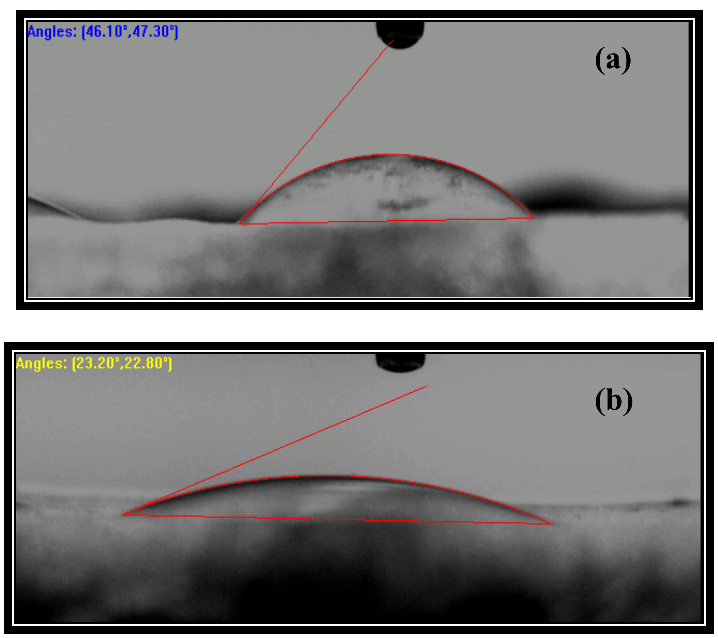

Figure 5 17 : Contact angles measured for MSN tortilla using formamide (a) at Aw=0.75 and (b) at Aw=0.97 as shown in the earlier report

5.11 Moisture Content Measurement

Moisture content of tortillas was measured using a hot air oven. In this technique the samples were dried in an oven (Thermolyne Oven, series 9000) at 103°C for 18 to 20 hours to a constant weight. The weight of samples before and after drying was carefully reported and the difference reported as percentage moisture content. The moisture content of tortillas, the amount of freezable water, and the stickiness of the tortillas were correlated.

5.12 X-ray Diffraction Analysis of Tortillas at Chemistry Department, Rutgers University:

The wide-angle x-ray scattering (WAXS) patterns of tortilla samples were obtained using a Bruker HiStar area detector and a rotating-anode x-ray generator equipped with a 0.5 mm collimator and a graphite monochromator (Cu Kα; λ = 1.5418 Å) operating at 40 kV and 75 mA. Samples were prepared at room temperature by placing them in 1mm glass capillaries in a constant water activity container, and then sealing them with molten beeswax (to minimize moisture loss during measurements) just prior to x-ray data collection at room temperature (20°C). The sample to detector distance was 9 cm. The capillary was placed in the sample holder (goniometer head) such that χ~45 deg. The collected images were 512x512 pixels. Data for a LaB6 powder standard were taken to confirm the true detector distance and beam center at $2\theta = 0$. X-ray data in each image were collected by rotating the sample 4 degrees in ω (only) from the initial $\omega=-2°$ and rotating completely in ϕ (spindle axis). The detector angle, 2θ, was fixed at 25°. Scan times were 600 sec to achieve about 48,000,000 total counts from the detector.

The intensities in the spatial diffraction patterns (Figure 5.18) obtained were integrated over a range of 90° to obtain intensity vs. 2θ plots using WinPlot graphical interface system software. The intensity versus 2θ plots ware analyzed for crystalline to amorphous ratio by the method of Cheetham and Tao (1998) as shown in Figure 5.19.

The intensity versus 2θ plot for each sample was fed into the SigmaScan Pro software for image analyses. A smooth curve which connected the peak baselines was plotted on the diffractograms. The area above the peak base line corresponded to the crystalline portion and the lower area between the smooth curve and a linear baseline which connected the initial and final 2θ values was taken as the amorphous region. The ratio of the areas corresponding to the crystalline and amorphous region was calculated as a measure of relative crystallinity.

$$\text{Relative Crystallinity (\%)} = \frac{\sum \text{Area under peaks}}{\text{Area of amorphous region}}$$

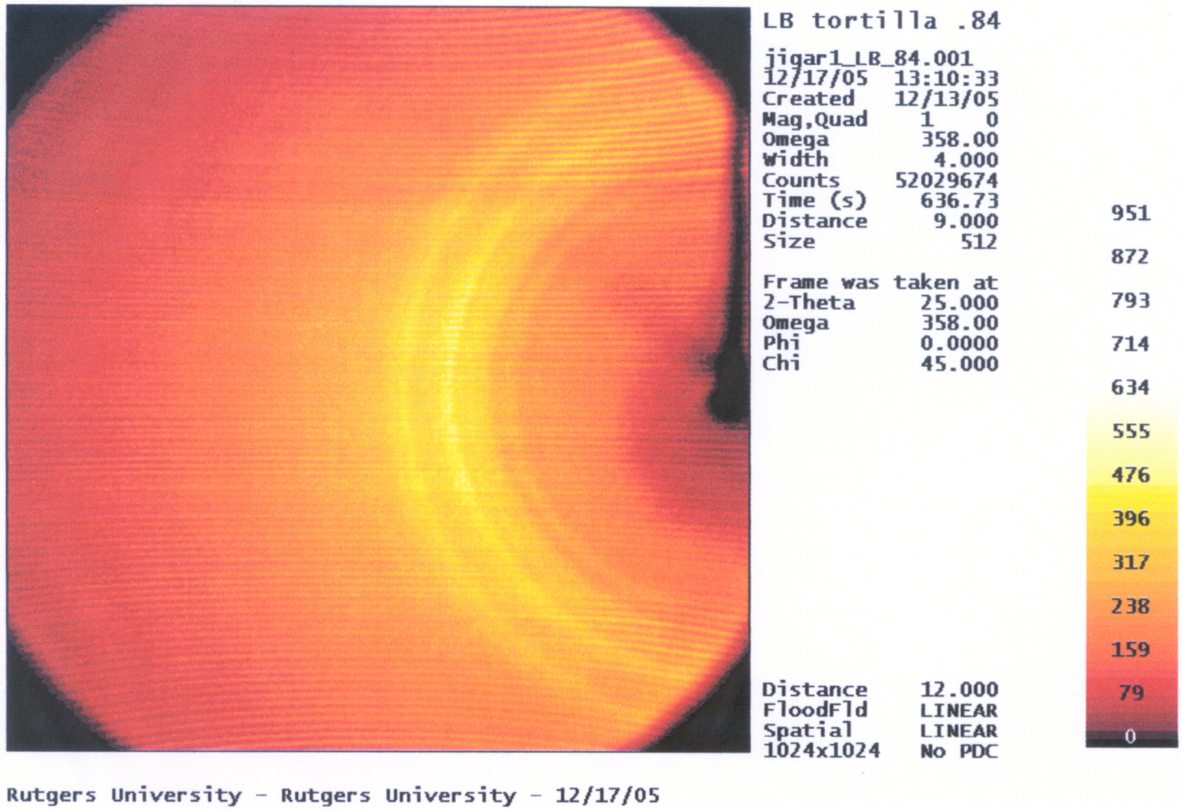

Figure 5 18 : Detector image of a non-sticky tortilla at 0.84 Aw using WAXS

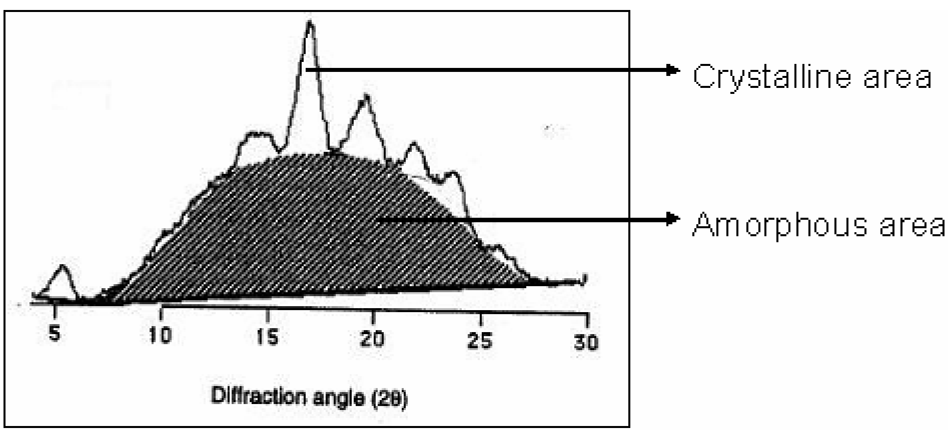

Figure 5 19 : Calculation of crystalline to amorphous ratio in maize (Cheetham and Tao, 1998)

5.13 X-ray Microtomography at Micro Photonics Inc. Allentown, PA:

High resolution X-ray microtomography, an emerging technique proven to provide non-destructive 3-D investigation of cellular materials was used to analyze cellularity of tortilla samples. We used a high-resolution (<5 μm) microtomograph (Skyscan 1072, Aartselaar, Belgium) for structural characterization of all tortilla samples.

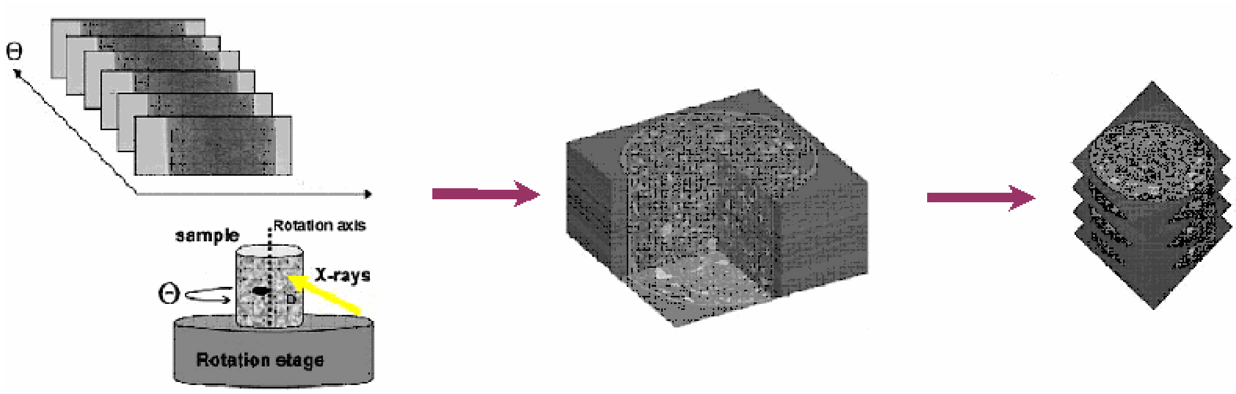

Figure 5 20 : Typical X-ray microtomography set up

Similar to medical CT scanners, the microtomograph used in this study consists of a monochromatic X-ray source, a rotatable stage and a panoramic detector. The sample is placed in a sample holder, which is rotated around the axis perpendicular to the beam direction. Components in the sample with different density (i.e. various solid materials or fluids) absorb X-radiation at different degrees. Images are recorded in the range 0-180° and angular projections are used to generate 2-D cross-sectional images. Software is used to reconstruct a 3-D object from the multiple 2-D images. Once a 3-D structure is constructed it can then be virtually sliced in any direction at any thickness. A set of 2-D images (i.e. slices) for the entire tortilla sample was obtained and analyzed to quantify the cell size distribution and average cell wall thickness using image analysis software (SigmaScan Pro 4.0).

The area and numbers of cells could be calculated using the software. The uniformity of pore size distribution was evaluated by using a polydispersity index, which is the ratio of weighted average cell area to number average cell area.

$$\mathrm{PDI} = \frac{A_W}{A_N} = \frac{\sum A_i^2 N_i / \sum A_i N_i}{\sum A_i N_i / \sum N_i}$$

where *Ai* is cell area and *Ni* is number of cells having *Ai*. PDI ranges between 1.0 and infinity, values closer to 1.0 signify a more uniform cell size distribution.

Average cell wall thickness was calculated using the formula below:

$$\text{Ave cell wall thickness (mm)} = \frac{A_{sample} - \sum_i^n A_i}{\sum_i^n P_i}$$

where the area of the solid portion of the sample (cell walls) was calculated as the difference between the cross-sectional area of the sample and total cell area, and *Pi* is the measured perimeter of the air cells. Then the following cellular characteristics were derived: cell density (number of cells per cm^2), average pore area (mm^2) and pore shape, and ratio of the average cell wall thickness to average pore radius, *t*/*R*.

$$\text{Ave cell area (mm}^2\text{)} = \frac{\sum_i^n A_i}{n}$$

$$\text{Cell density (\# cell/cm}^2\text{)} = \frac{n}{A_{sample}}$$

5.14 Image Analyses

Cross-sectional images of tortilla samples were analyzed using image analysis techniques proposed by (Smolarz 1989), (Barrett and Ross 1990) using SigmaScan Pro 4.0. The analysis procedure consists of a number of steps: The grabbed image was subjected to a process called "histogram stretch" to increase the contrast. Then "intensity threshold" was applied to identify the pores to be analyzed. Thresholding is simply detecting only those areas whose color value or monochrome value falls within a specified range. The image was then inverted and black pixels were converted to one of the four colors: red, green, yellow or blue, because the analyzer only measures objects with these colors.

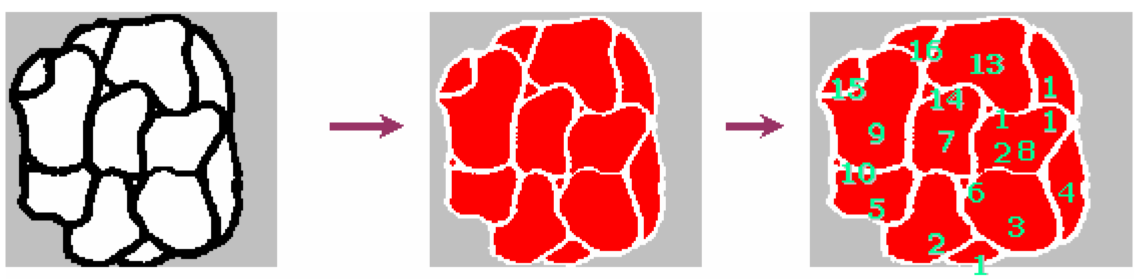

Figure 5 21: Image analysis using SigmaScan Pro software of a pet food sample done in Dr. Kokini's Lab, Food Science Department, Rutgers University, NJ

Upon analysis, the number of cells in each section and the area and perimeter, shape factor and diameter of each cell were measured. Average cell wall thickness was calculated using the formula below:

$$t_{wall} = \frac{A_{solid}}{\sum_i P_i}$$

Where, *Asolid* is the solid portion of the sample cross-sectional area, calculated as the difference between the cross-sectional area of the sample and total cell area, and *Pi* is the measured perimeter of the air cells. Then the following cellular characteristics were derived: cell density (number of cells per cm2), average pore area (mm2) and pore shape, and ratio of the average cell wall thickness to average pore radius, *t*/*R*.

5.15 Statistical Analysis of Data

Data were subjected to correlation, average, and standard deviation analysis on Microsoft Excel (Microsoft office XP, 2003). Average, standard deviation, correlation coefficient, and R^2 were measured and reported.

Correlation coefficient (CORREL): $\rho_{X\,Y}$ between two random variables X and Y with expected values μ_X and μ_Y and standard deviations σ_X and σ_Y is defined as:

$$\rho_{X,Y} = \frac{\mathrm{cov}(X,Y)}{\sigma_X \sigma_Y} = \frac{E((X-\mu_X)(Y-\mu_Y))}{\sigma_X \sigma_Y},$$

Standard Deviation (STDEV): is a measure of the spread of values. It is usually denoted with the letter σ and defined as the square root of the variance.

$$\sqrt{\frac{n\sum x^2 - \left(\sum x\right)^2}{n(n-1)}}$$

R^2 : The r-squared value was measured using following formula.

$$R^2 = 1 - \frac{SSE}{SST}$$

where

$$SSE = \sum \left(Y_i - \hat{Y}_i \right)^2$$

and

$$SST = \left(\sum Y_i^2 \right) - \frac{\left(\sum Y_i \right)^2}{n}$$

VI. RESULTS AND DISCUSSION

6.1 Causes of Tortilla Stickiness:

6.1.1 Correlation between Sensory and Instrumental Stickiness

A good positive correlation (R^2 = 0.72) between sensory and instrumental stickiness was found (Figure 6.1). A linear equation was used to calculate the trend line, which was y = 0.0013x-0.5003. It was observed that the tortillas which showed stickiness values away from the trend line values were T2 (Instrumental stickiness of 90.21 g) and T3 (instrumental stickiness of 362.05 g). Tortillas with low to moderate stickiness were found difficult to identify using sensory analysis. More panelist training before sensory stickiness evaluation might have resulted in an improved R^2 value.

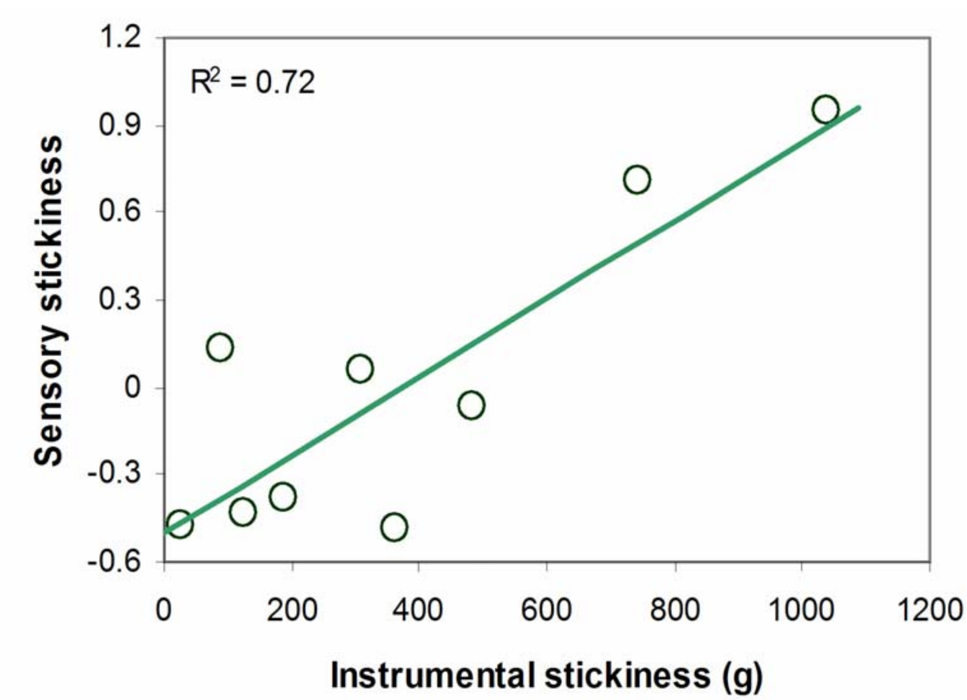

Figure 6 1 : Correlation between sensory and instrumental stickiness

6.1.2 Effect of Water Activity on Tortilla Stickiness:

Experimental results have shown that instrumental stickiness increases significantly with an increase in water activity. Below a water activity level of 0.84 tortilla samples did not show measurable stickiness (Figure 6.2). Tortillas at water activities lower than 0.84 had Tg values higher than room temperature (23˚C) which can be seen in the phase diagram later in this section. According to Slade (1993) if a product is stored below its Tg value it will maintain its glassy state. This might be a reason for reduced or no stickiness in tortilla samples.

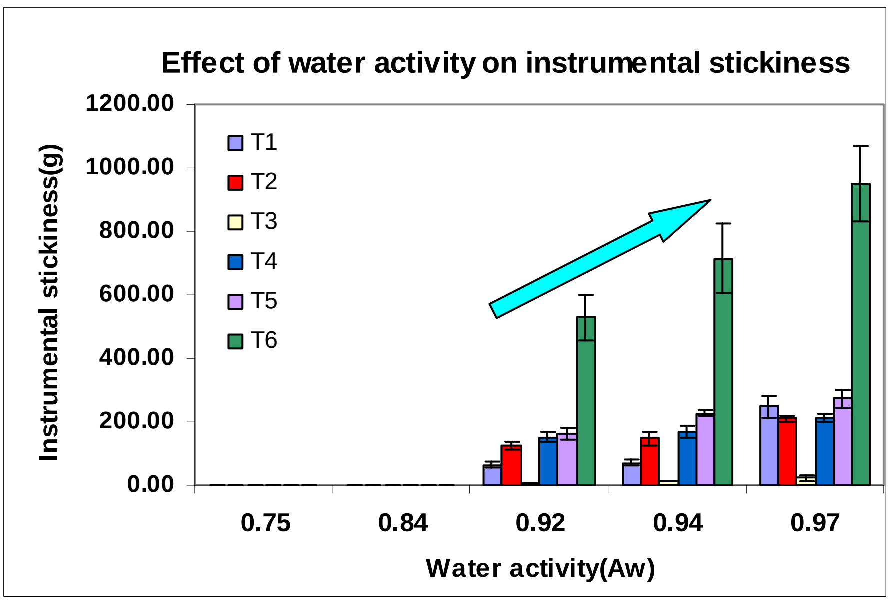

Figure 6 2 : Effect of water activity on instrumental stickiness

6.1.3 Moisture Sorption Isotherm of Tortillas

Figure 6.3 shows moisture sorption isotherms for sticky, moderately sticky, and non-sticky tortillas. From the isotherms it can be seen that up to Aw=0.84 tortilla samples have shown a comparable increase in their moisture contents with increasing Aw. After this point the sticky tortilla (T6) has higher moisture content and therefore free water available at any given water activity level as compared to the non-sticky tortilla indicating higher hygroscopic behavior of the product which is due to the compositional differences. This high moisture content in sticky tortillas at high Aw levels is believed to increase the molecular mobility and ultimately the surface adhesion forces, which are responsible for the stickiness in tortillas. The moderately sticky tortillas (T4 and T5) follow the same pattern as T6 but to a lower extent. Whereas non-sticky tortillas T1, T2, and T3 showed lower moisture content values (30.81, 31.16, and 31.84 % respectively) at 0.94 water activity level.

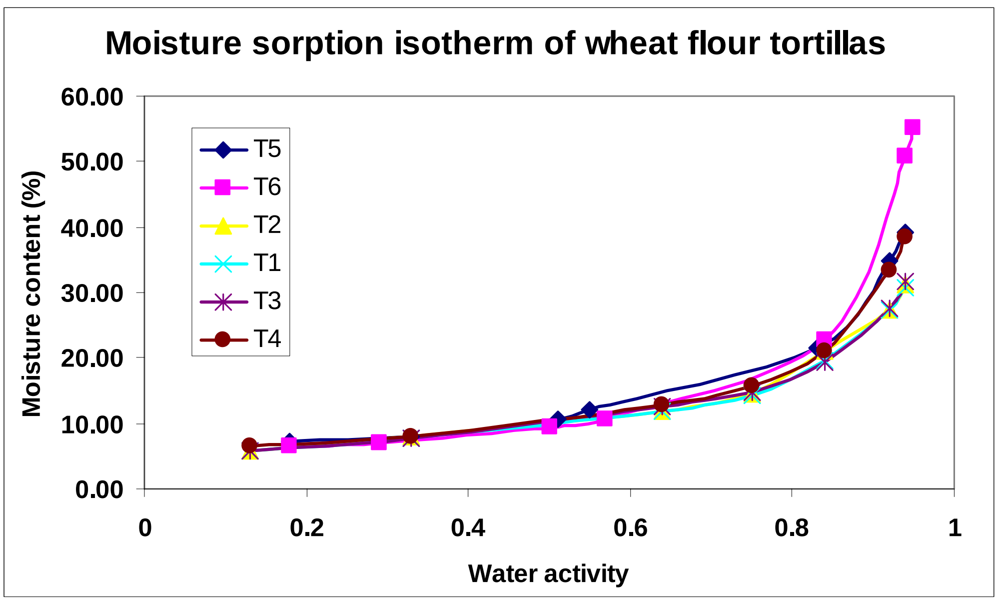

Figure 6 3 : Moisture sorption isotherms of sticky and non-sticky flour tortillas

6.1.4 Effect of Tg on Tortilla Stickiness (Phase/state diagram)

The change in glass transition temperatures of tortilla samples as a function of water activity obtained using combined data from ARES and RSA II is given in Figure 6.4.

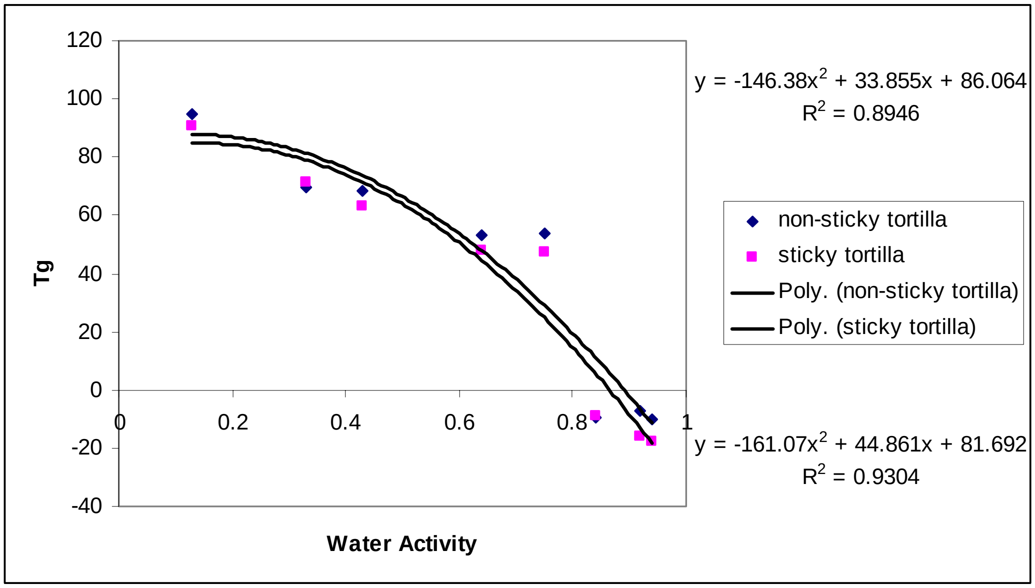

Figure 6 4 : State diagram of sticky and non-sticky flour tortillas

The glass transition temperature (Tg) both for sticky and non-sticky tortillas dropped significantly with an increase in water activity due to the well-known plasticization effect of water on biopolymers. The Tgs ranged from -20˚ to 100˚C in the range of water activities from 0.13 to 0.94. The Tg dropped sharply at water activity levels above 0.8. Sticky and non-sticky tortillas showed different phase behavior as shown in the above Figure 6.4. Although trend lines were very close to each other, the non-sticky tortillas had a slightly higher glass transition temperature compared to sticky tortillas. Tortillas were sticky at water activities above 0.84. So the phase behavior (Tg) of tortilla above 0.84 water activity is important. At 0.94 water activity sticky tortillas showed a Tg value of -16.07˚C as compared to -7.4˚C for non-sticky tortillas. That is a difference of 8.62˚ C. Slade et al., 1989 attributed, the rate of decrease of *Tg* to water plasticization as ~10˚C per 0.01g of water/g of material. Suppose tortillas have an average weight of 40g. According to Slade et al sticky tortillas should have 0.35 g more moisture compared to non-sticky tortillas. This free moisture might contribute to higher mobility which ultimately leads to association, cross linking, bonding at tortilla surfaces, and hence higher stickiness. The moisture difference (g) in sticky and non-sticky tortillas was 9.77 g at 0.94 water activity as shown in the moisture sorption isotherm (moisture content = 55.25% for T6 and 30.81% for T1 tortilla). This is many times the 0.34 g as calculated by Slade et al. method. Tortillas are a complex food with many ingredients and that is why they showed a very large range for Tg. That made it difficult to find a clear Tg. There is a need to develop a method that can show a clear Tg value.

6.1.5 Crystallinity of Sticky and Non-sticky Tortillas

Crystallinity of tortilla samples with different stickiness were characterized as a function of water activity levels. Samples from both the surface and the crumb of flour tortillas were analyzed. Figure 6.5 shows intensity vs. 2Θ plots obtained for the sticky and non-sticky tortilla samples through intensity integration of the region $8 < 2\theta < 48°$ and $-134 < \chi < -44°$. Overall, the patterns show a broad background with breadth from amorphous content approximately the same for all water activities. The presence of crystallinity is confirmed by the presence of peaks in the diffraction patterns. For the strongest identifiable peak (near $2\theta = 20°$), the degree of crystallinity increased slightly with increase in Aw level for non-sticky samples. The sticky tortillas have a broad amorphous region as compared to non-sticky tortillas.

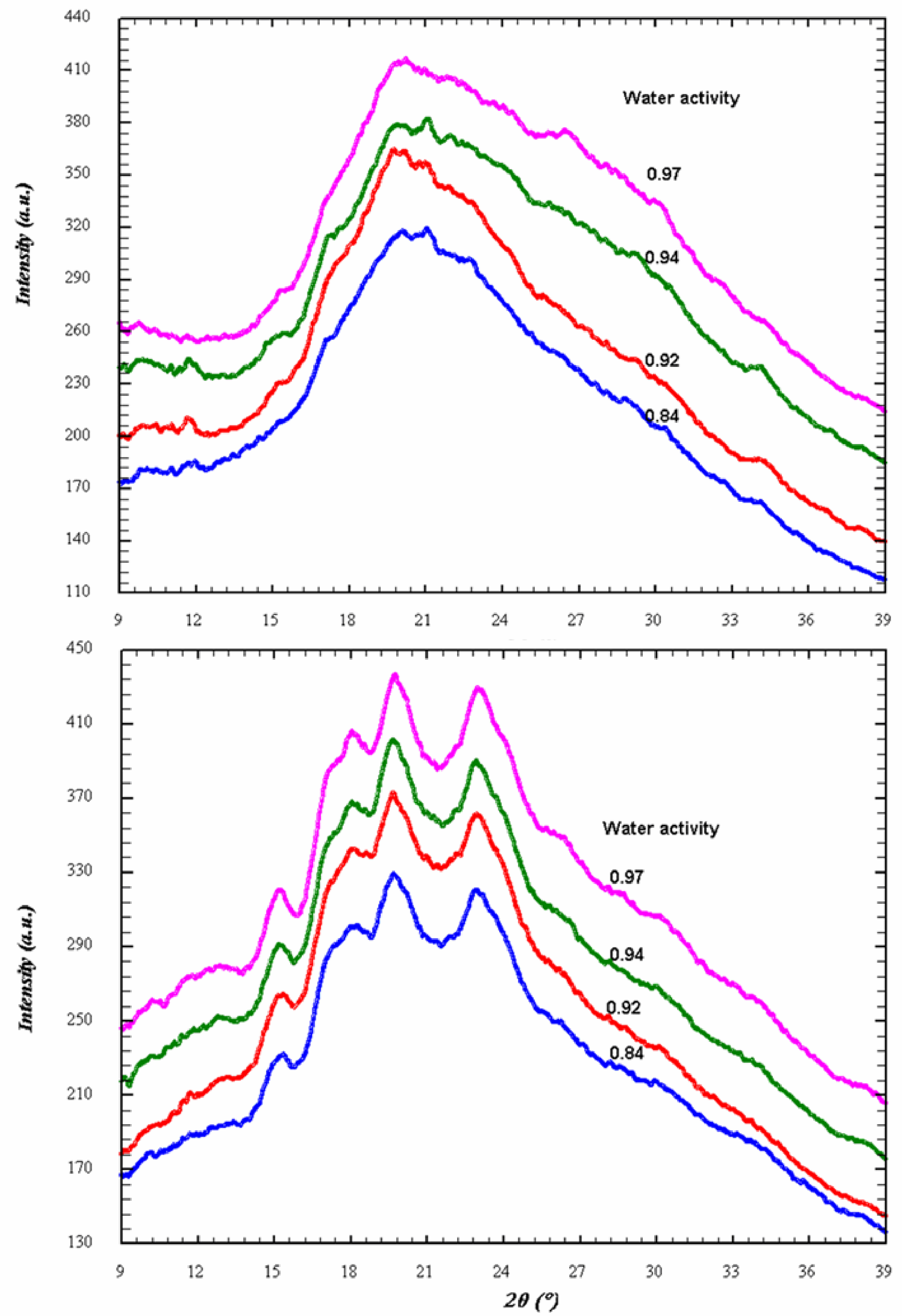

Figure 6 5 : Crystallinity of (a) sticky and (b) non-sticky tortilla samples

Comparison of the diffraction patterns for the samples with those of typical cereal starches (Zobel 1988) confirmed that the crystallinity in the samples predominantly originated from starch. Native cereal starch shows A type crystallinity with the principal peaks appearing at 15°, 17° and 23.4° of 2Θ (Becker 2001). The peaks around 20° of 2Θ indicates the presence of V_h type crystallization patterns due to amylose-lipid complexes. Re-crystallization of the amylopectin during cooling and storage may also possibly account for the presence of A-type crystallinity in the samples.

As shown in Figure 6.5 the sticky tortilla has lower degree of crystallinity and a larger amorphous region. The characteristic peaks seen in the non-sticky sample at 2θ ≈21° and 23° were not observed for sticky samples. The relative crystallinity calculated for tortilla surface and crumb for non-sticky, moderately sticky, and sticky tortillas for four different water activity levels are given in the Table 6.1. Slightly higher crystallinity was observed in the core than on the surface. This result was unexpected. The core of tortillas had higher moisture than the surface because tortillas dry out more quickly on the surface. In the case of T1 the tortillas' surface and core moisture were 30.06 and 28.59% respectively. A possible reason that could explain this phenomenon is that higher core moisture might enhance molecular activity of amylose and amylopectin molecules to rearrange and crystallize. Retrogradation in tortilla core crumb could be happening upon cooling as moisture is being lost. The starch molecules rearrange themselves and the moisture earlier trapped between them oozes out to the surface. This also promotes crystallinity (Schiraldi 1996). Both non-sticky and moderately sticky tortillas show higher relative crystallinity than sticky tortillas. Sticky tortilla T6 surface at 0.94 water activity had 0.7 % relative crystallinity. The non sticky tortilla T1 and moderately sticky tortilla T4 showed 4.13 and 5.24 % relative crystallinity respectively. Less crystallinity and thus higher amorphous regions in sticky tortillas may be partly responsible for stickiness. The amorphous regions tend to agglomerate at temperatures higher than Tg and can cause stickiness. The crystallinity results for the tortilla surface are more important here because stickiness appears on the surface of tortilla.

Table 6.1
Relative (%) Crystallinity of Tortilla Surface and Core

Water Activity	**Non-Sticky Tortilla**		**Moderately Sticky Tortilla**		**Sticky Tortilla**	
	Surface	**Core**	**Surface**	**Core**	**Surface**	**Core**
0.84	3.42	4.67	3.53	4.71	1.39	1.30
0.92	3.17	4.74	5.45	6.28	1.39	1.30
0.94	4.13	5.71	5.24	5.62	0.70	1.65
0.97	5.30	5.54	6.00	6.21	1.18	1.64

The reason for higher crystallinity on a moderately sticky tortilla surface compared to a non-sticky tortilla surface could be the solid bridges theory as explained by Pietsch, 1997. He observed that when amorphous materials crystallize the solid dissolved in the liquid crystallizes and forms solid bridges at the points of contact.

6.1.6 Effect of Surface Energy on Tortilla Stickiness

Reduced surface energy of tortillas corresponded with a higher contact angle which is a manifestation of high surface tension of the liquid selected. When the surface energy of tortillas is higher, to overcome the surface tension of liquid the droplet spreads resulting in a reduced contact angle. Contact angle measurements and corresponding surface energy calculations have also shown a strong dependence on water activity (Figures 6.6, 6.731, 6.8). The polar component of surface energy corresponding to hydrophilic surface energy increased with an increase in Aw levels while the dispersive component of surface energy corresponding to hydrophobic surface energy decreased with an increase in Aw levels (Figures 6.6 and 6.7). An increase in the polar component indicated an increase in surface hydrophilicity. The increase in the polar component with an increase in Aw was much more significant than the drop in the dispersive component in terms of affecting the overall change in surface energy. Thus, the overall change in the surface energy ($\gamma^{total}=\gamma^{dispersive}+\gamma^{polar}$) was observed to increase with increasing Aw (Figure 6.8).

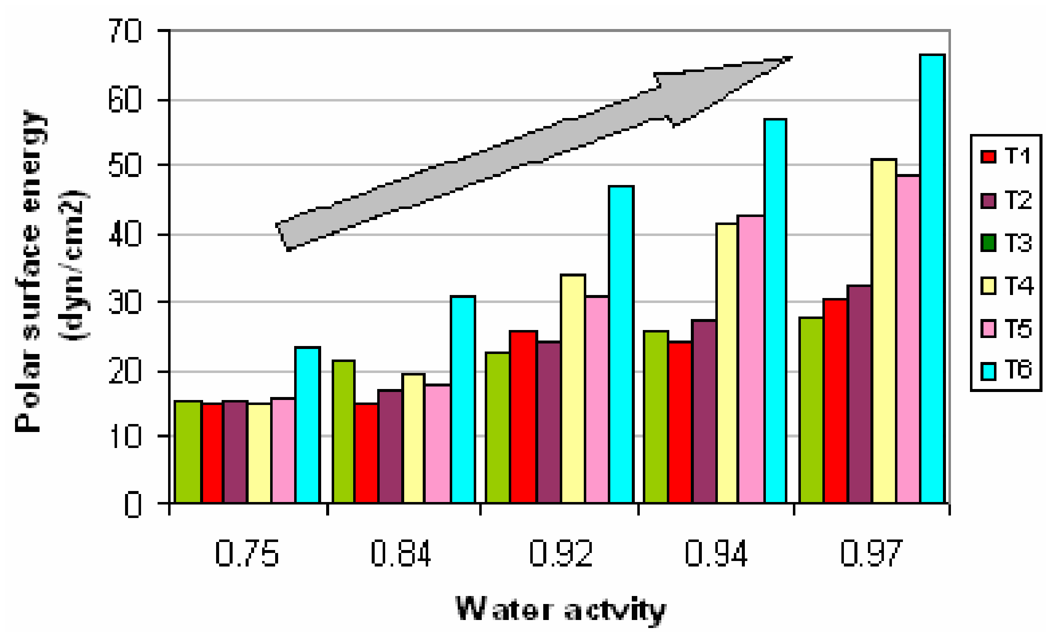

Figure 6 6 : Effect of water activity on polar component of surface energy

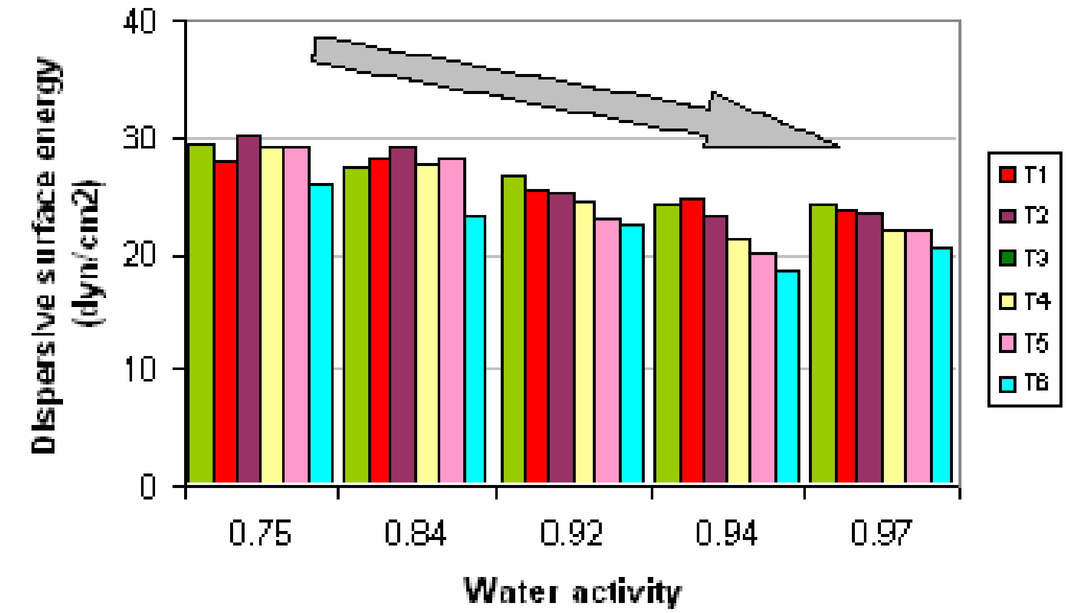

Figure 6 7 : Effect of water activity on dispersive component of surface energy

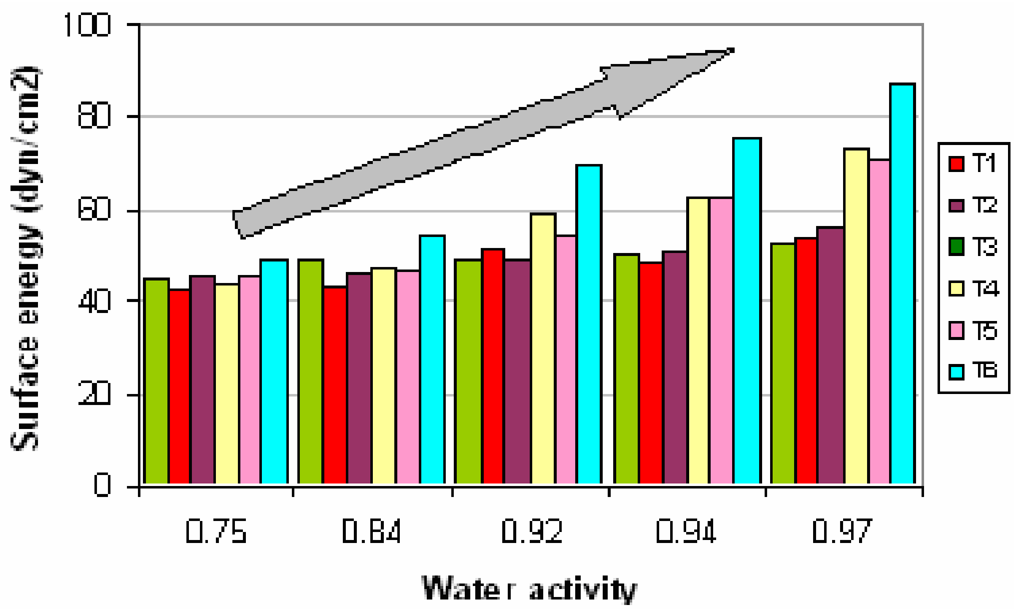

Figure 6 8 : Effect of water activity on total surface energy

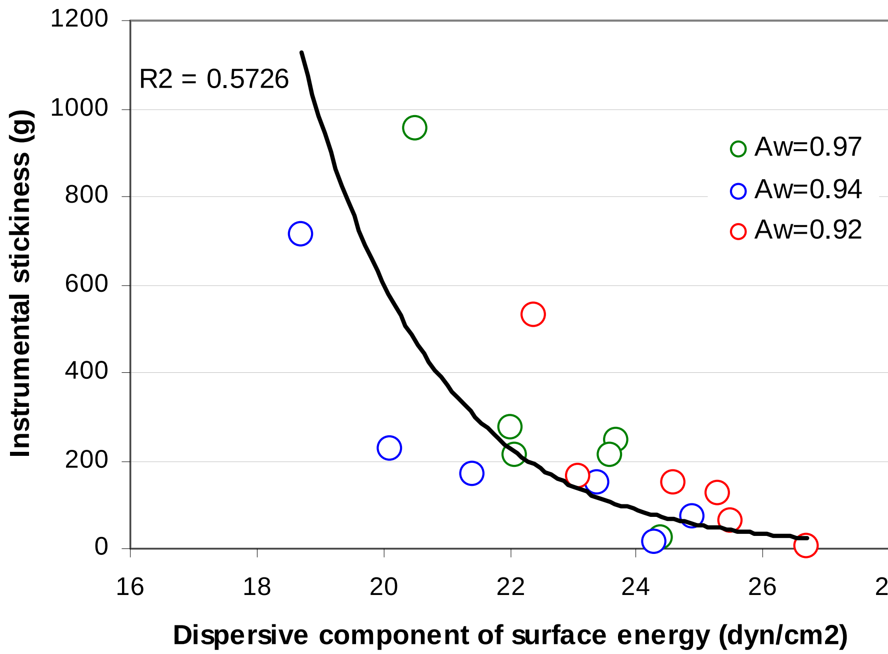

Figure 6 9 : Variation in the instrumental stickiness with the dispersive surface energy

The dispersive component of surface energy was not as sensitive to type and composition of tortilla products as the polar component of the surface energy. The differences between the dispersive surface energy values calculated for six tortilla samples did not show much difference, although there was a clear inverse relationship with Aw in dispersive surface energy.

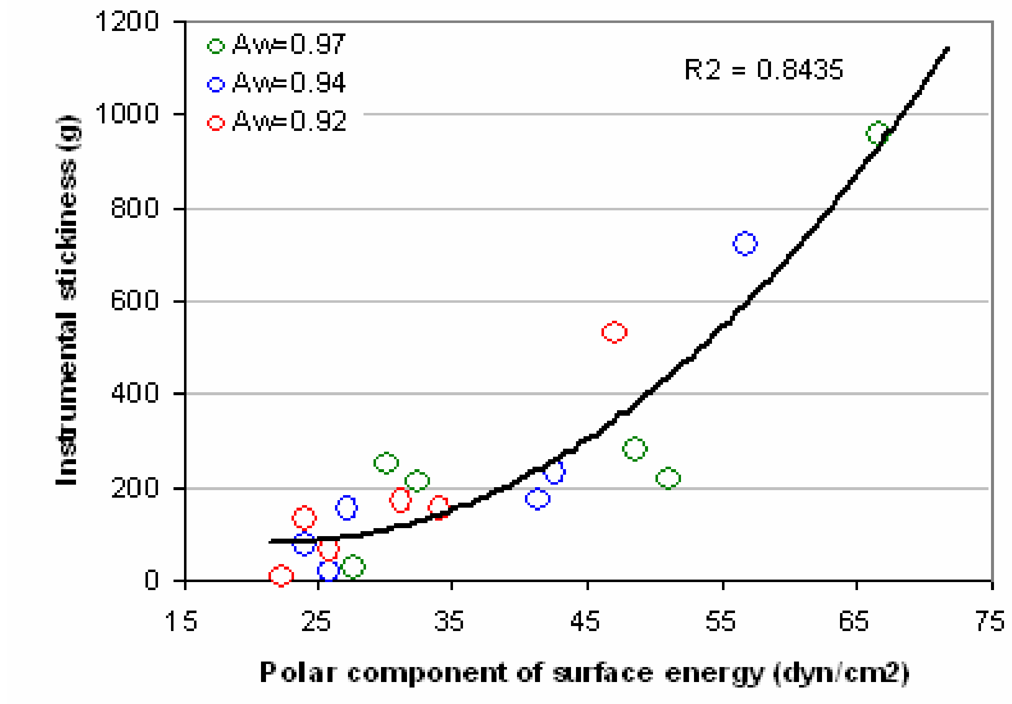

Figure 6 10 : Variation in the instrumental stickiness with polar surface energy

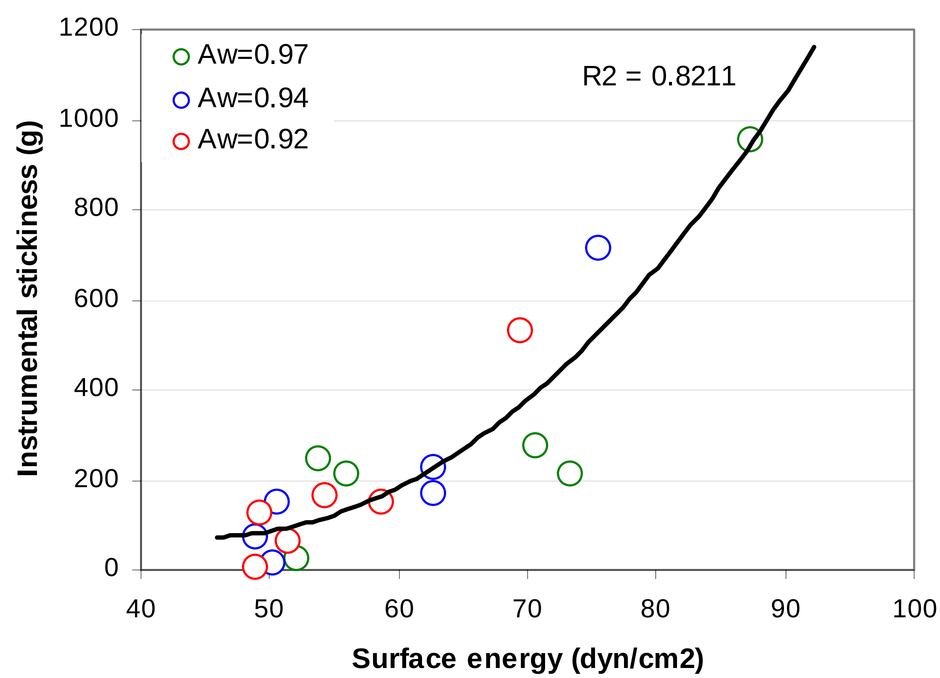

Figure 6 11 : Variation in the instrumental stickiness with the surface energy

Instrumental stickiness was found to correlate well with surface energy and its components and particularly with the polar component of surface energy (Figures 6.10, 6.11). Correlation coefficients (R^2) between instrumental stickiness and the dispersive surface energy, the polar surface energy, and the total surface energy were calculated to be -0.74, 0.88 and 0.87, respectively. The strong positive correlation between the instrumental stickiness and the polar component of the surface energy indicated that stickiness observed in tortilla products is a moisture-driven property which causes an increase in the hydrophillicity of surfaces and creates attractive forces between the two tortilla surfaces. There seems to be a threshold surface energy above which the tortilla stickiness is observed.

6.2. Effect of Addition of Gums and Polyols on Dough Moisture Content and Stickiness

We studied the average moisture content and its relationship to stickiness of the raw uncooked dough as a diagnostic characteristic of final tortilla stickiness. The machinability (ease in handling the dough) of tortilla dough changes with moisture content. As shown in Table 6.2 the moisture content of the dough decreases approximately ten percent with the addition of gums and glycerol.

Table 6.2
Effect of Addition of Gums and Polyols on Dough Moisture Content and Stickiness

Type Dough	Dough moisture %	Dough stickiness (g)
Control	38.44 ± 0.02	56.39 ±16.24
Gum	38.12 ±0.14	77.61 ±26.91
Gum and glycerol	36.9 ±0.14	67.33 ±12.07
Glycerol	37.34 ±0.05	67.33 ±17.45
Propylene glycol	37.47 ±0.92	75.44 ±11.42
Gum and propylene glycol	38.01 ±0.06	64.43 ±14.98

Dough moisture content in the control was 38.44 %. Addition of glycerol (4% by weight of flour) reduced the moisture content of the dough to 37.34 %. The lowest moisture content, 36.9%, was observed when gums and glycerol were added in combination. Dough stickiness was measured using a Chen-Hoseney dough stickiness cell. As shown in the above table the stickiness of dough increases with the addition of gums and polyols. The dough stickiness increased from 56.39 g in the control to 75.44 g in the case of dough with propylene glycol. However, these increases in the stickiness did not affect the machinability of dough. Instead, the doughs with gums and polyols, alone and in the combination, were found to be more pliable. Tortillas made using these ingredients were rounder. The changes observed in dough stickiness were not large but within the range of standard deviation.

6.3 Effect of Ingredient Strategy on Tortilla Stickiness

6.3.1 Effect of Addition of Gums and Polyols on Tortilla Water Activity:

As shown in Figure 6.12 water activities of tortillas decreased when polyols were added to the formula. The water activity decreased from 0.94 to ~ 0.91 when tortillas were made using polyols alone and in combination with gums.

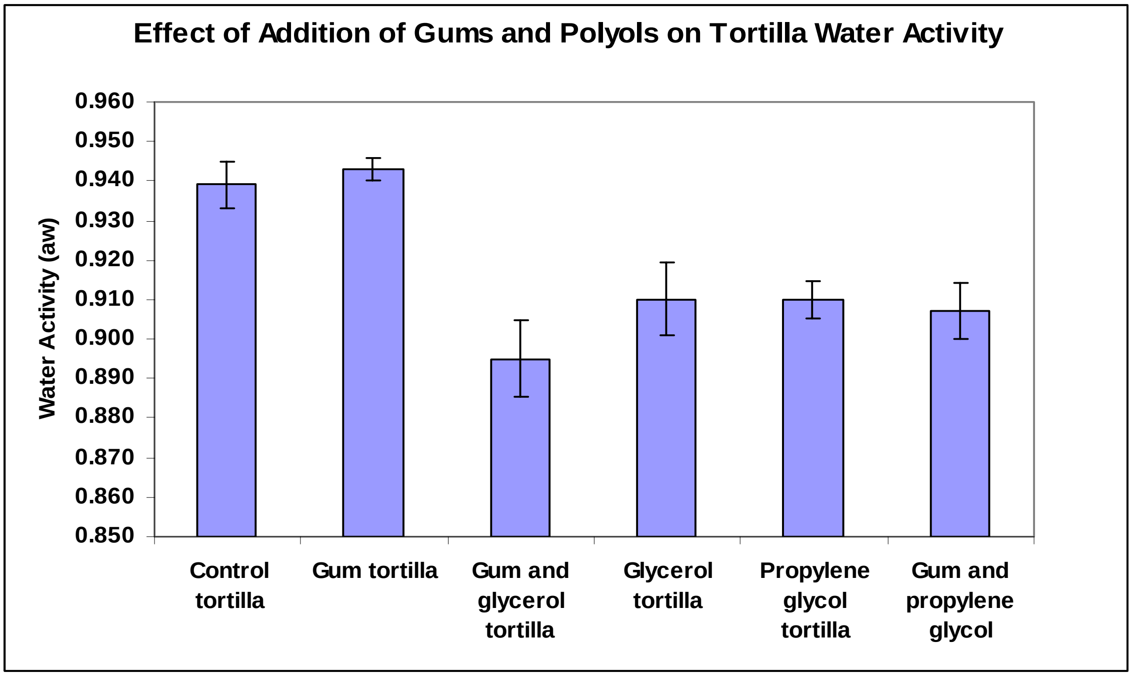

Figure 6 12 : Effect of addition of gums and polyols on tortilla water activity

6.3.2 Effect of Addition of Gums and Polyols on Tortilla Moisture Content

As shown in Figure 6.13 a decrease in moisture content was observed when tortillas were made using gums and glycerol. The lowest moisture percentage was 24.57% in the case of tortillas made with propylene glycol in combination with XG and CMC gums.

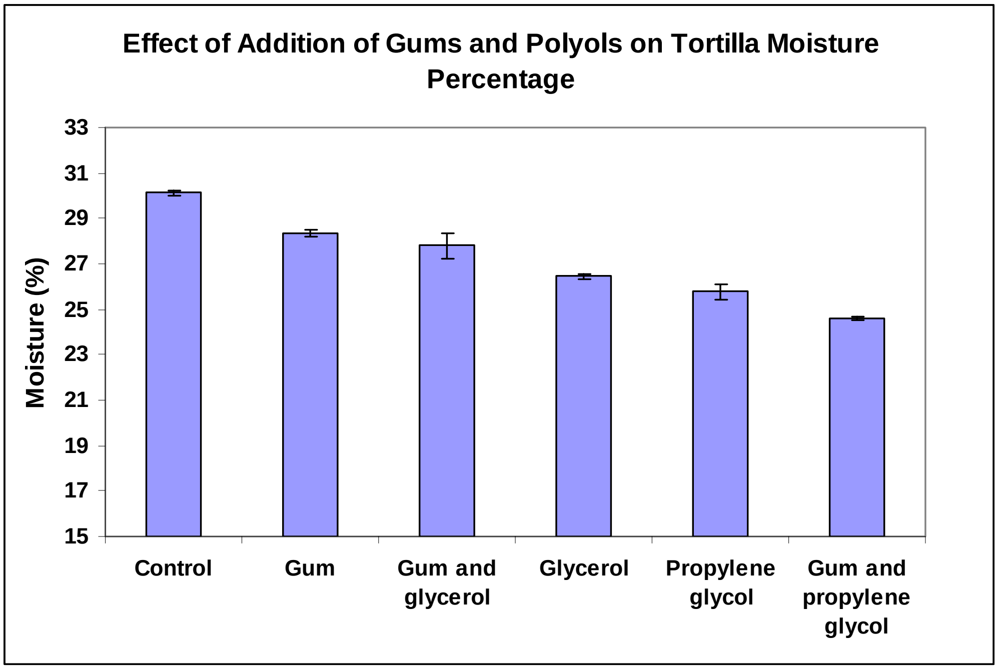

Figure 6 13 : Effect of addition of gums and polyols on tortilla moisture content

Tortilla moisture percentage decreased from 30.13 % (control tortilla) to 28.35% in tortillas added with XG and CMC gums.

6.3.3 Effect of Addition of Gums and Polyols on Freezable Water Present in Tortillas

The thermal behavior of tortillas was monitored by DSC. Figure 6 14 shows the thermogram of tortillas. The freezable water region in the control tortilla was the highest. Addition of gums slightly reduced the freezable water present in tortillas. Striking differences were observed when tortillas were made with polyols. Polyols reduced the available freezable water in tortillas. This is because of the free –OH groups in polyols which bound water molecules and thus reduced freezable water present in tortillas. Reduction in freezable water (FW) induced by glycerol was previously reported for other bakery products (i.e. bread (Baik and Chinachoti, 2001)). Baik stated that this reduction in FW may be related to an increase in viscosity of the hydrophilic phase of the product resulting in a decrease in motion of the water molecules that could form ice crystals detectable by DSC.

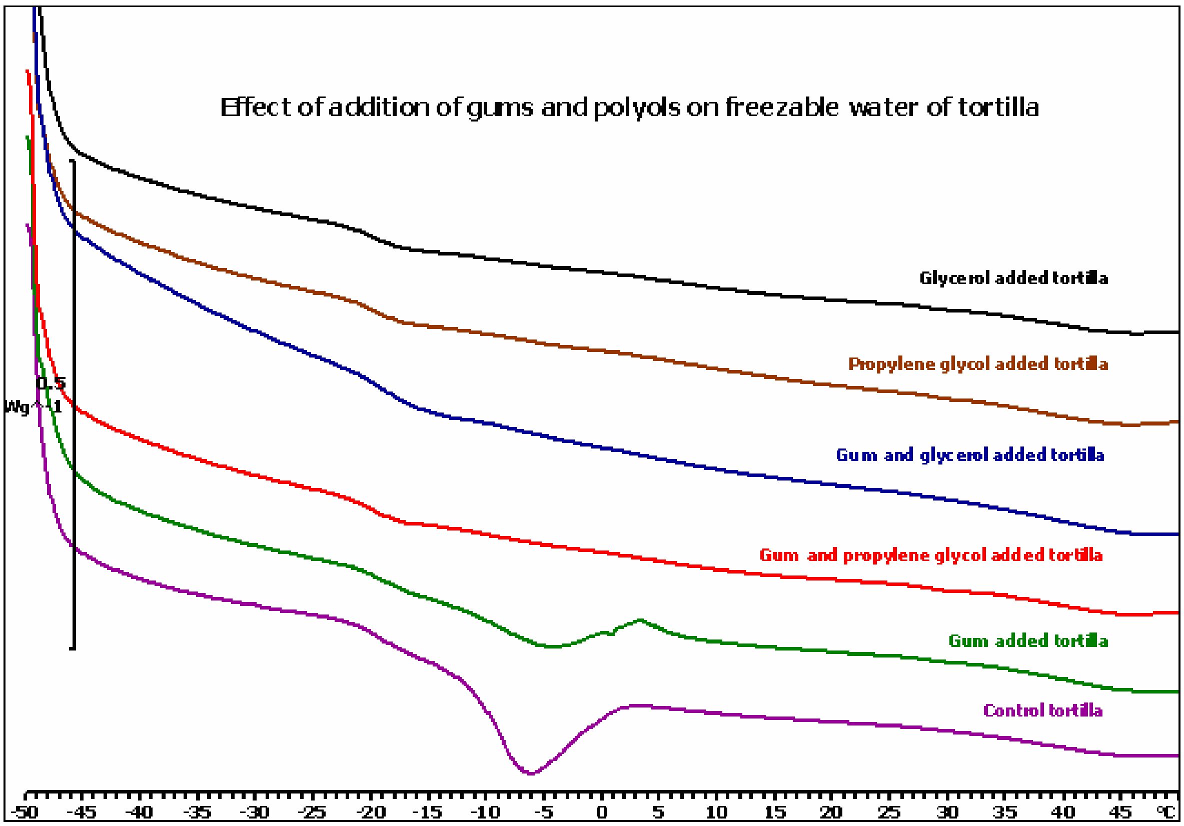

Figure 6 14 : Thermo gram showing amount of freezable water in tortillas using DSC823 e

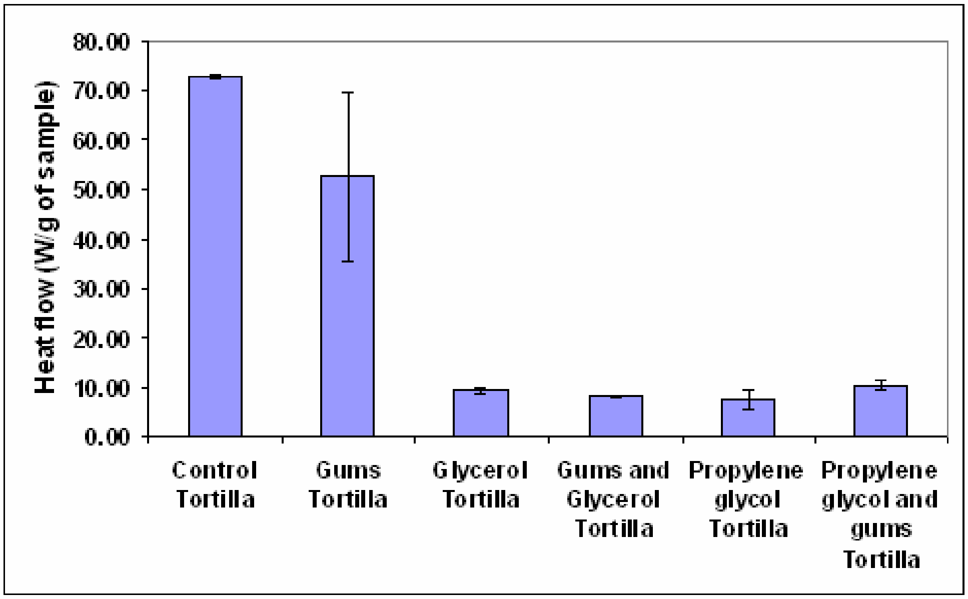

Figure 6 15 : Effect of addition of gums and polyols on freezable water

Polyols tend to stabilize water–starch systems by becoming incorporated in the structure of the water that surrounds the starch chain (Miura et al., 1992). This interaction of glycerol with water makes the water 'unfreezable', resulting in a reduction of FW in the system (Miura et al., 1992).

6.3.4 Effect of Addition of Gums and Polyols on Tortilla Cellularity

As shown in the Figure 6 16 tortillas prepared with different processing conditions and ingredients shows different cellularity. Tortillas were arranged in trial 1 to 8 in the following figure.

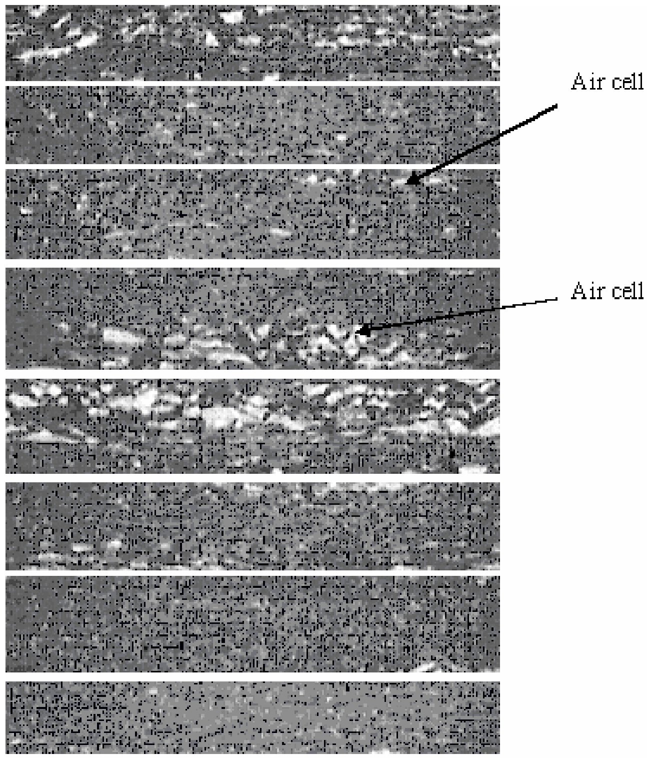

Figure 6 16 : X-ray microtomography images of tortilla trials

Table 6.3
Effect of Addition of Glycerol on Tortilla Cellularity

Type of tortilla	Ave cell area, A (mm2)	Ave cell radius, R (mm)	Cell density (cell/cm2)	Ave cell thickness, t (mm)	t/R	PDI
Tortilla 1 = Control (450_20_10)	0.024±0	0.087±0	696.40 ± 74.38	0.12± 0.03	1.4 ± 0.31	4.8 ± 0.34
Tortilla 2 = PG+Gums(450_20_5)	0.014±0.01	0.066±0.02	621.6±114.08	0.159±0.04	2.61±1.26	5.27 ± 4.39
Tortilla 3 = Control (350_10_5)	0.018±0.01	0.075±0.01	484.88±82.55	0.201±0.02	2.71±0.32	5.2 ± 0.68
Tortilla 4 = Glycerol	0.02±0.02	0.081±0.01	441.9±37.27	0.172±0.04	2.16±0.6	4.17 ± 1.41
Tortilla 5 = Control (450_20_5)	0.023±0.01	0.084±0.02	506.64±132	0.165±0.05	2.13±1.08	5.2 ± 2.05
Tortilla 6 = Gum+glycerol (450_20_5)450-380	0.014±0	0.066±0.01	514.26±94.15	0.191±0.03	2.94±0.86	5.05 ± 3.29
Tortilla 7 = Control(350_20_5)	0.009±0	0.052±0.01	377.14±75.45	0.268±0.05	5.523±2.06	3.77 ± 1.11
Tortilla 8 = Gum+glycerol (450_20_5) 450_420	0.012±0.01	0.059±0.02	457.07±89.81	0.217±0.05	3.98±1.98	5.36 ± 3.29

To understand the effect of glycerol addition in tortillas compare T4 (tortilla added with 4% glycerol) to T5 (control tortilla). As shown in Table 6.3 addition of glycerol does not have an effect on air cell size (0.08 mm radius in both cases). The average cell area reduced to 0.021 from 0.23 mm^2 when glycerol was added. The cell density was reduced to 441.90 from 506.64 $cell/cm^2$. But the average cell thickness was found unchanged (0.17mm).

To understand the effect of addition of 4% propylene glycol and 0.5% XG and CMC on tortilla cellularity compare T5 (control tortilla) to T2 (tortilla added with gums and PG). It was noticed that addition of both gums and PG reduced the air cell size (0.08 to 0.07 mm radius) but cell thickness almost remained the same (~0.17). Addition of PG and gums led to increased air cell density of 621.60 from 506.64 $cell/cm^2$.

To understand the effect of addition of 4% glycerol in combination with 0.5% CMC and XG in tortilla compare T5 (Control) to T6. Average cell area reduced to 0.014 from 0.023mm^2 and average cell radius reduced to 0.066 from 0.084 mm when tortillas were incorporated with glycerol and gums. As expected the cell wall increased to 0.191 mm from 0.165 mm.

6.3.5 Thermal Behavior of Tortillas Containing Added Gums and Glycerol

The average glass transition temperature of control tortillas at 0.94 water activity was -4.32°C. The Tg value decreased to -3.48 °C and -9.27 °C for tortillas containing gums (XG and CMC) and both gums (XG and CMC) and glycerol respectively as shown in Figures 6.17 and 6.18. It was noticed that even though addition of polyols reduced tortilla water activity from 0.94 to 0.9 it did not increase the Tg value. Instead addition of propylene glycol reduced the Tg value of tortillas to -10 °C. That means if tortillas with added polyols were equilibrated at 0.94 water activity and analyzed for Tg, the Tg value would have been even lower. Tortilla Tg is lower than 0˚C irrespective of the addition of ingredients. That means all tortillas are in the rubbery state and prone to stickiness.

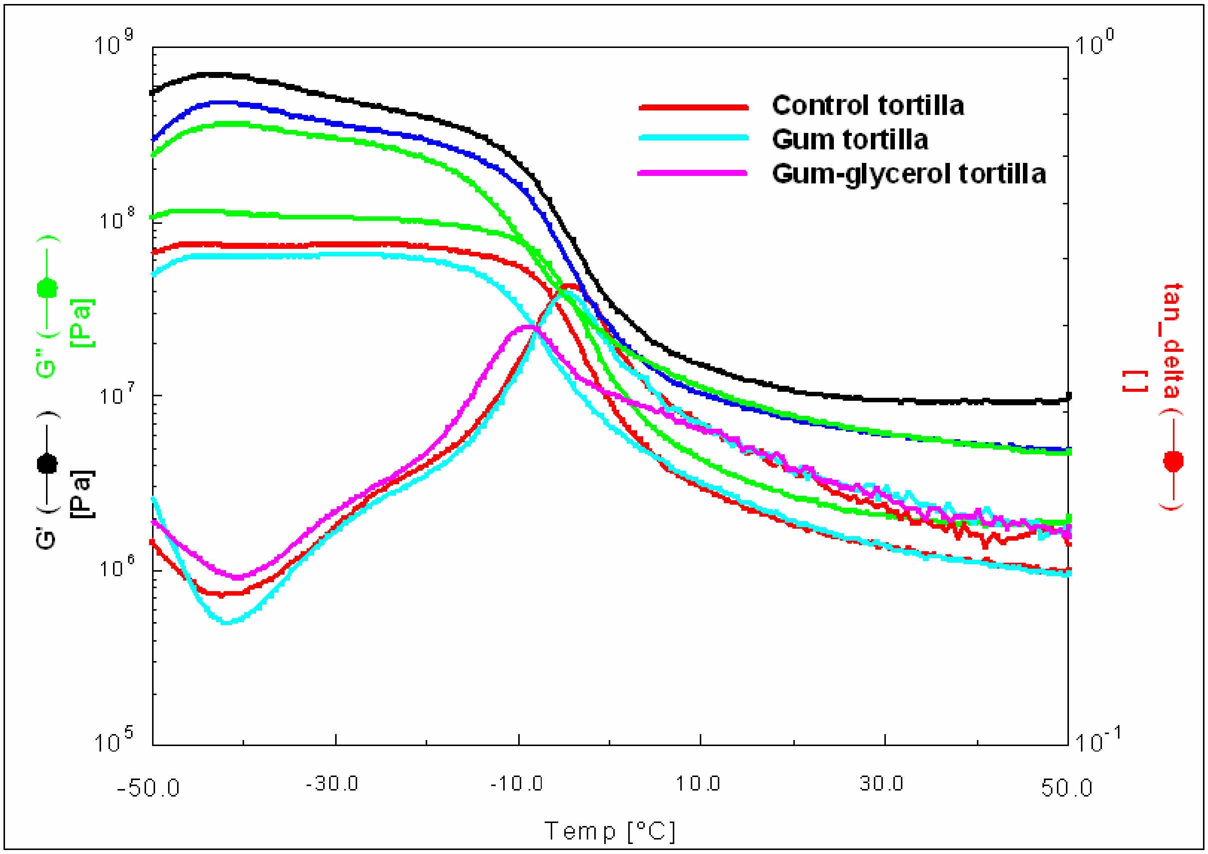

Figure 6 17 : Superimposition of temperature sweep of control, gum and gum-glycerol tortillas

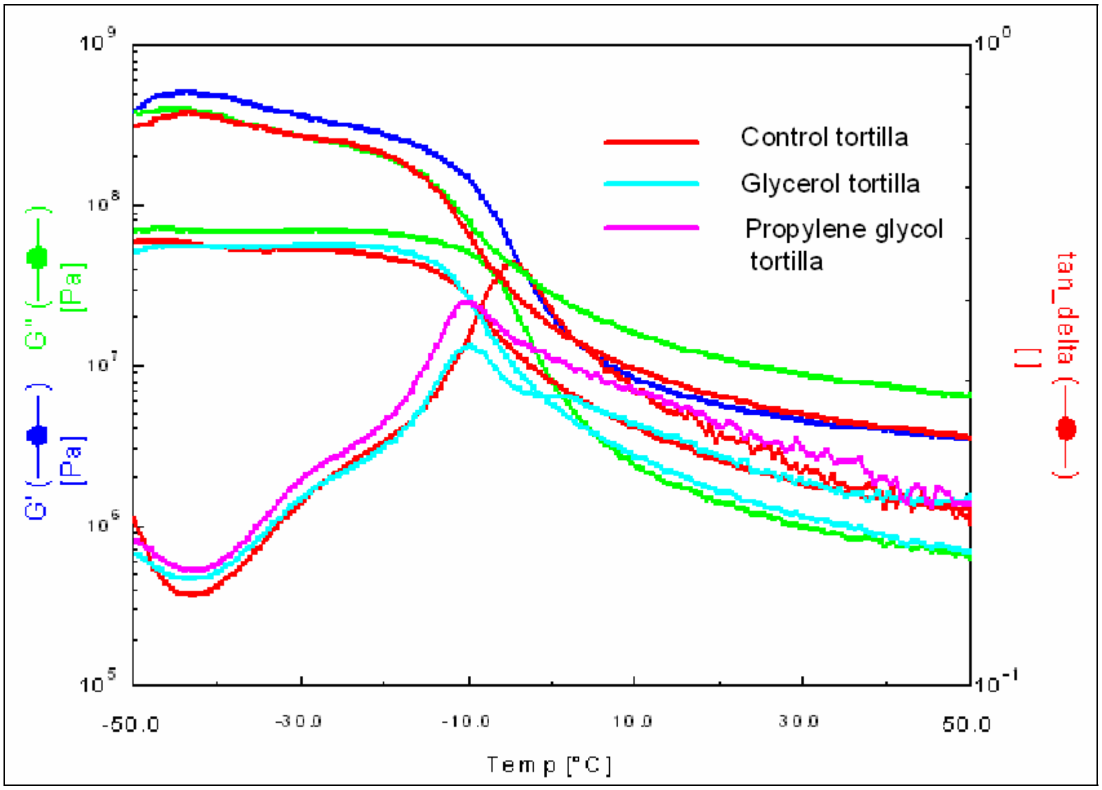

Figure 6 18 : Superimposition of temperature sweep of control, glycerol and propylene glycol tortillas

The reduction in Tg value in the case of tortillas containing polyols was due to the plasticizing effect. This allowed the tortillas to remain relatively soft while binding significant amounts of water.

6.3.6 Effect of the Addition of CMC and XG Gums and Polyols on Relative Crystallinity of Tortillas

As shown in Table 6.4 the relative crystallinity of control tortilla is the highest (8.99%) followed by CMC and XG gum incorporated tortilla and gum (CMC+XG)-glycerol tortilla with 8.76% and 8.19% respectively. Addition of polyols reduced the crystallinity of tortillas. The lowest crystallinity was observed in the case of tortillas made with the addition of propylene glycol.

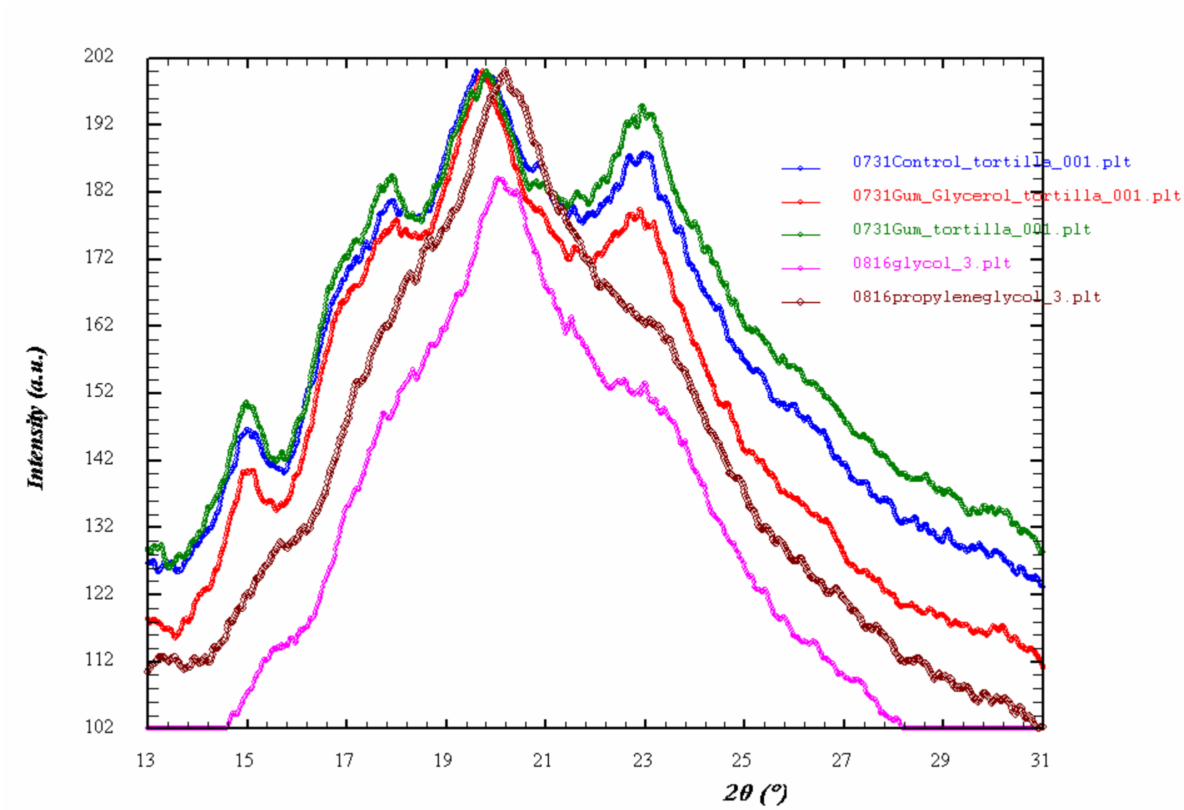

Figure 6 19 : Intensity integration of tortilla samples with gums and polyols

Table 6.4

Relative Crystallinity of Control, Gum Tortilla and Gum-Glycerol Tortilla

Type of tortilla	Relative crystallinity (%)
Control	8.99 ± 0.66
Gum Tortilla	8.76
Gum and Glycerol tortilla	8.19 ± 0.66
Glycerol Tortilla	7.01
Propylene glycol tortilla	4.06

Comparison of the diffraction patterns for the samples with those of typical cereal starches (Zobel, 1988) confirmed that the crystallinity in the samples predominantly originated from starch. Native cereal starch shows type A crystallinity with the principal peaks appearing at 15°, 17°, and 23.4° of 2Θ. The peak around 2Θ = 20° indicates the presence of V_h type crystallization patterns due to amylose-lipid complexes. Re-crystallization of the amylopectin during cooling and storage may account for the presence of A-type crystallinity in the samples. No distinguishing new peak was observed in tortillas made with added ingredients as compared to the control tortilla. The only feature that was affected was the magnitude of the peak, showing that the extent of crystallinity was affected. Sticky tortillas showed reduced crystallinity as shown in the earlier discussions which is not the case here. That means crystallinity is not only reason for the stickiness in tortillas. Stickiness is a complex phenomenon governed by more than one factor such as water activity, moisture content, freezable water present, glass transition temperature, stacking, surface energy, and crystallinity. These factors are correlated to each other such as water content affects water activity or freezable water present in the product. It is clear that not a single factor by itself but the balance of these factors decides stickiness of tortillas.

6.3.7 Effect of Addition of Gums and Polyols on the Surface Energy of Tortillas

As shown in Figure 6.20 total surface energy of the control tortilla was observed to be 63.7 dyne/cm. Addition of CMC and XG gums reduced total surface energy of the tortilla to 54.9 dyne/cm. Addition of glycerol alone and in combination with CMC and XG gums reduces the total surface energy of the tortilla to 56.9 and 50.9 dyne/cm respectively.

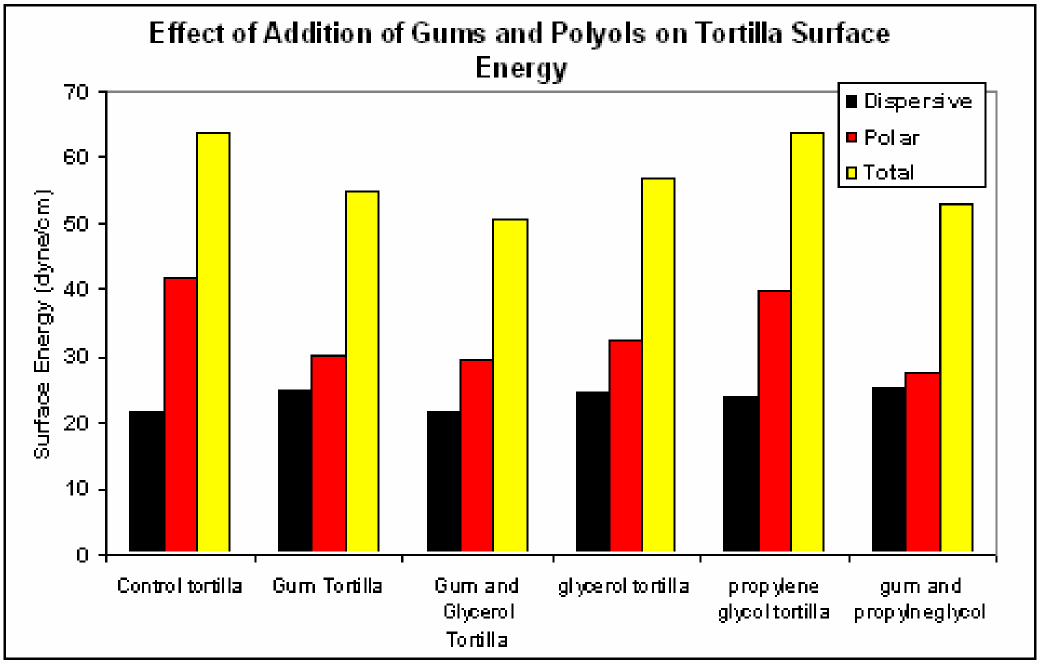

Figure 6 20 : Effect of addition of CMC and XG gums and polyols on tortilla surface energy

When propylene glycol was added, total surface energy was found to remain the same. However the polar component of surface energy which is mainly responsible for stickiness in tortillas was observed to be lower in tortillas containing propylene glycol as compared to the control. A combination of gums and glycerol showed lower total surface energy. The change in polar surface energy is very important. Polar surface energy was shown to be mainly responsible for the stickiness as we have seen in the earlier part of this thesis. The polar component of surface energy was highest (41.9 dyne/cm) in the case of the control tortilla. Lowest polar surface energy was observed in the case of tortillas prepared using gums and glycerol. Addition of gums and polyols in tortillas showed lower polar surface energy as compared to the control. The dispersive component of surface energy which represents hydrophobic energy remained more or less same.

6.3.8 Effect of Addition of Gums and Polyols on Tortilla Stickiness

As shown in Figure 6.21 addition of 0.5% CMC and Xanthan gum and 4% polyols (glycerol and propylene glycol) effectively reduce the stickiness of tortillas. The control tortilla showed a stickiness value of 211.05 g. This substantially reduced to 65.4 g when tortillas were made using 4% glycerol. Tortillas made with 0.5% CMC and XG gums, 4% propylene glycol, and a combination of CMC and XG gums and 4% glycerol also showed reduction in stickiness.

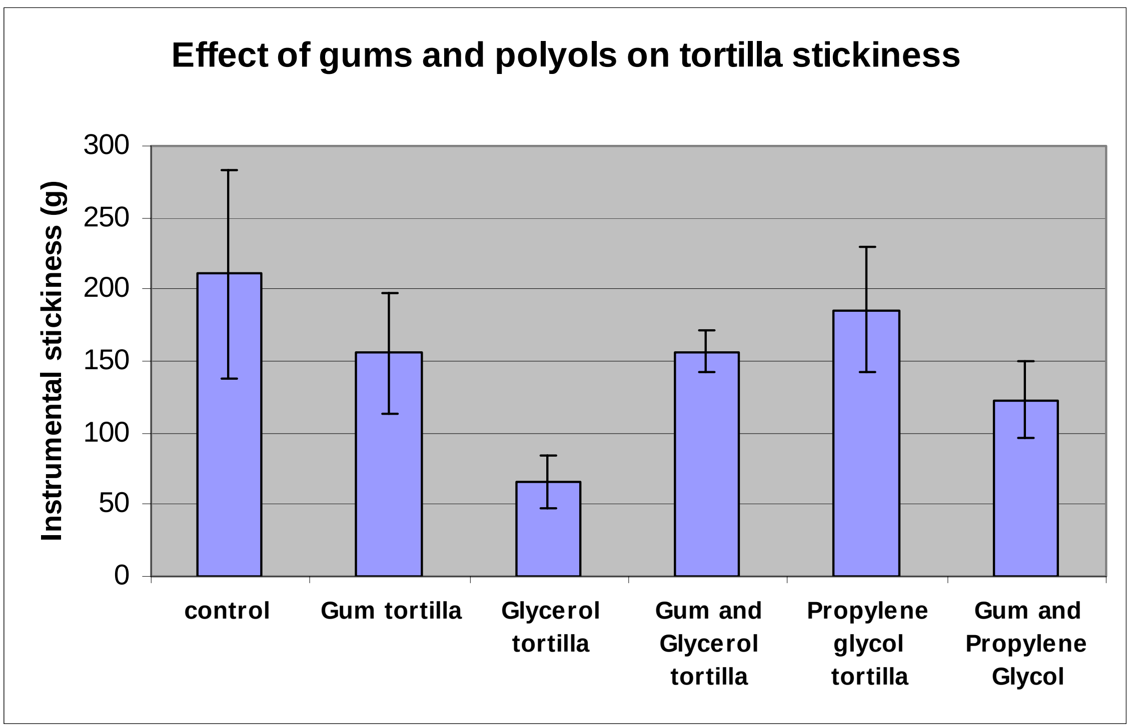

Figure 6 21 : Effect of addition of gums and polyols on tortilla stickiness

Addition of 4% Glycerol was found to be more useful in controlling stickiness as compared to 4% propylene glycol. This may be because of the chemical structure of these polyols. Glycerol has three free –OH groups to bind water whereas propylene glycol has only two. However, a combination of CMC and XG at 0.5% and 4% propylene glycol produced reduced stickiness (122.93 g) compared to the combination of 4% glycerol and 0.5% CMC and XG (156.67 g). A positive soft correlation (0.40) was found between water activity and stickiness of tortillas**.** A positive correlation (0.63) was observed between polar surface energy and instrumental stickiness of tortillas. Moisture percentage was also positively correlated with stickiness (0.54)

A positive correlation of 0.56 was observed between stickiness in tortillas and the amount of freezable water present. Once again these soft to moderate correlations suggests that stickiness is a complex phenomenon and the balance of tortilla properties such as surface energy, water activity, etc. decides stickiness.

6.4 Effect of Processing Parameters on Tortilla Properties

To understand the effect of processing conditions on the stickiness of tortillas we made tortillas using two levels of processing temperatures (350°F and 450°F). The time of tortilla cooling after baking was also studied at three levels to understand its effect on stickiness. These cooling times are: 2 min, 5 min, and 10 min cooling after baking. Dough resting times of 10 min and 20 min were selected. The main hypothesis in this particular case was that dough resting time and baking temperature have a major impact on the distribution of water and therefore will affect the balance between free and bound water.

6.4.1 Effect of Processing Conditions on Tortilla Moisture Content

As shown in Table 6.5 baking temperatures have a mixed effect on tortilla moisture content. Tortillas baked at 350°F with 10 min dough resting and 2 min cooling time showed a lower moisture content compared to tortillas baked at 450°F but when dough resting time was 20 min tortillas baked at 350°F showed a higher moisture content.

Table 6.5
Effect of Processing Condition Parameters on Moisture Content and Water Activity

Tortilla Number	Resting time(min)	Baking Temp(ºF)	Cooling Time(min)	Moisture content (%)	Water activity
1	10	350	2	27.45± 0.27	0.930 ±0.01
2	10	350	5	26.87± 0.39	0.926 ±0.00
3	10	350	10	28.2 ± 0.00	0.926 ±0.00
4	20	350	2	29.02 ± 0.82	0.926 ±0.01
5	20	350	5	27.79 ± 0.74	0.935 ±0.00
6	20	350	10	26.93 ± 0.46	0.930 ±0.00
7	10	450	2	29.10 ± 0.62	0.931 ±0.00
8	10	450	5	27.19 ± 0.16	0.923 ±0.01
9	10	450	10	27.70 ± 0.16	0.916 ±0.01
10	20	450	2	27.50 ± 0.82	0.929 ± 0.00
11	20	450	5	25.92± 0.93	0.925 ±0.00
12	20	450	10	27.16 ±1.53	0.925 ±0.00

A tortilla cooling time of 5 min was found to be most effective in controlling tortilla moisture content as compared to 2 and 10 min. The lowest moisture content of 25.92% was observed for tortillas made with a baking temperature of 450°F, dough resting time of 20 min, and cooling time of 5 min. At the high resting time moisture loss is more pronounced than at low resting times as would be expected from moisture transfer in tortillas from the surface. A cooling time of 2 min showed a higher moisture percentage in all cases because moisture loss at this cooling time is less as expected.

As shown in Table 6.5 dough resting time of 20 min showed a higher moisture content for tortillas baked at 350°F. However for tortillas baked at 450°F it showed a lower moisture percentage. Again as explained above this is due to moisture loss at higher temperature and high resting time where moisture flash off and movement out of the tortilla is highest. The highest moisture percentage was found for tortillas baked at 450°F, with a dough resting time of 10 min, and cooling time of 2 min.

6.4.2 Effect of Processing Conditions on Tortilla Water Activity

The water activity of baked and cooled tortillas was found to be the most important parameter in controlling tortilla stickiness as we have seen in the earlier part of this thesis. Tortillas made using a baking temperature of 350°F were found to have a higher water activity because they also have the highest moisture content. The highest water activity was observed for tortillas baked at 350°F with a dough resting time of 20 min and cooling temperature of 5 min.

As shown in Table 6.5 an increase in the tortilla cooling time from 2 min to 10 min reduced the water activity of tortillas because of increased moisture loss during cooling. The lowest water activity was found in tortillas baked at 450°F with a dough resting time of 10 min and cooling time of 10 min because during manufacture this tortilla is subjected to the highest temperature and then is given the longest time to rest and lose moisture. This is also the tortilla with the lowest moisture content.

As shown in Table 6.5 the general trend suggests that a higher dough resting time prior to processing and baking allows dough to retain more moisture in the gluten matrix and subsequently increases the water activity of tortillas.

6.4.3 Effect of Processing Conditions on the Amount of Freezable Water and Crystallinity in Tortillas

As shown in Table 6.6 a baking temperature of 450°F reduced the amount of freezable water present in tortillas. These are tortillas that have been baked and subjected to different cooling times. The moisture content of the tortillas baked at 450°F is one of the lowest (as reported in earlier section) which also reflected itself in their water activity.

Table 6.6:
Effect of Processing Conditions on the Amount of Freezable Water and Crystallinity in Tortillas

Tortilla Number	Resting time(min)	Baking Temp(°F)	Cooling Time(min)	Freezable water (mJ)	Relative crystallinity(%)
1	10	350	2	184.03 ± 4.91	7.39
2	10	350	5	142.40 ± 113.28	6.20
3	10	350	10	155.60 ± 35.38	6.48
4	20	350	2	348.54 ± 0.99	7.53
5	20	350	5	193.78 ± 75.33	6.67
6	20	350	10	171.87 ± 5.16	6.57
7	10	450	2	271.36 ± 47.69	5.54
8	10	450	5	130.73 ± 35.79	3.86
9	10	450	10	69.65 ± 36.92	4.86
10	20	450	2	92.76 ± 1.20	6.10
11	20	450	5	91.04 ± 53.90	6.45
12	20	450	10	79.73 ± 43.66	5.23

This results in less free water in the tortillas which in turn reduces the freezable water. Tortillas baked for 450°F with dough resting and tortilla cooling times of 10 min showed the lowest freezable water (69.65 mJ). On the other hand, tortillas which were baked at 350˚F which were shown to have the highest moisture content and higher water activity have more free water which is the source of the freezable water and results in high freezable water.

It can be seen from Table 6.6 that a lower amount of freezable water was found in tortillas cooled for 10 min compared with tortillas at 2 and 5 minutes. As discussed earlier this is due to the fact that moisture loss is highest at 10 min cooling time where the tortilla starts from its baking temperature and gradually approaches room temperature. In this process moisture is lost progressively as the tortilla is cooled longer because the tortilla is hot and moisture continues to evaporate from this hot tortilla. This results in less moisture in the tortilla and also less free water resulting in turn in less freezable water. More freezable water was found in tortillas rested for a lower time (10 min). A higher resting time allowed the dough to bind more water in the gluten matrix.

6.4.4 Effect of Tortilla Processing Conditions on Relative Crystallinity of Tortilla Surface

As shown in Table 6.6 a higher baking temperature (450°F) reduced the relative crystallinity of tortillas. The lowest relative crystallinity of 3.86 % was observed for tortillas baked at 450°F with a dough resting time of 10 min and cooling time of 5 min. The highest crystallinity (7.53 %) was observed for tortillas baked at 350°F with 20 min dough resting time and 2 min cooling time. This result was unexpected. Tortillas baked at 450°F were expected to show higher crystallinity because at higher temperatures the surface dries out faster and enhances retrogradation. The possible explanation is that at baking temperature of 450°F starch and protein molecules denaturized and insolubility increased. Lack of moisture and high heating temperature might have led to reduced gelatinization of starch. Reduced moisture led to reduced mobility of amylose and amylopectin. These phenomena might have resulted in lower crystallinity. Moreover the XRD tests were performed three days after tortilla manufacture. Longer storage of samples might have showed different results.

A cooling time of 5 min gave the lowest relative crystallinity of 3.88%. A cooling time of 2 min gave the highest relative crystallinity of 7.53%. When tortillas were cooled for only 2 min and packaged in the PE bags, they retained higher moisture as discussed earlier. This happened because tortillas were hot after 2 min cooling and upon packaging in PE bags moisture condensed which ultimately went back into the tortillas. Higher moisture on the surface might have resulted into higher mobility for amylose and amylopectin molecules to rearrange and crystallize. Ten min of cooling time resulted in more than required cooling. Tortillas dried out faster which was reflected in lower moisture content. This might have led to higher crystallinity.

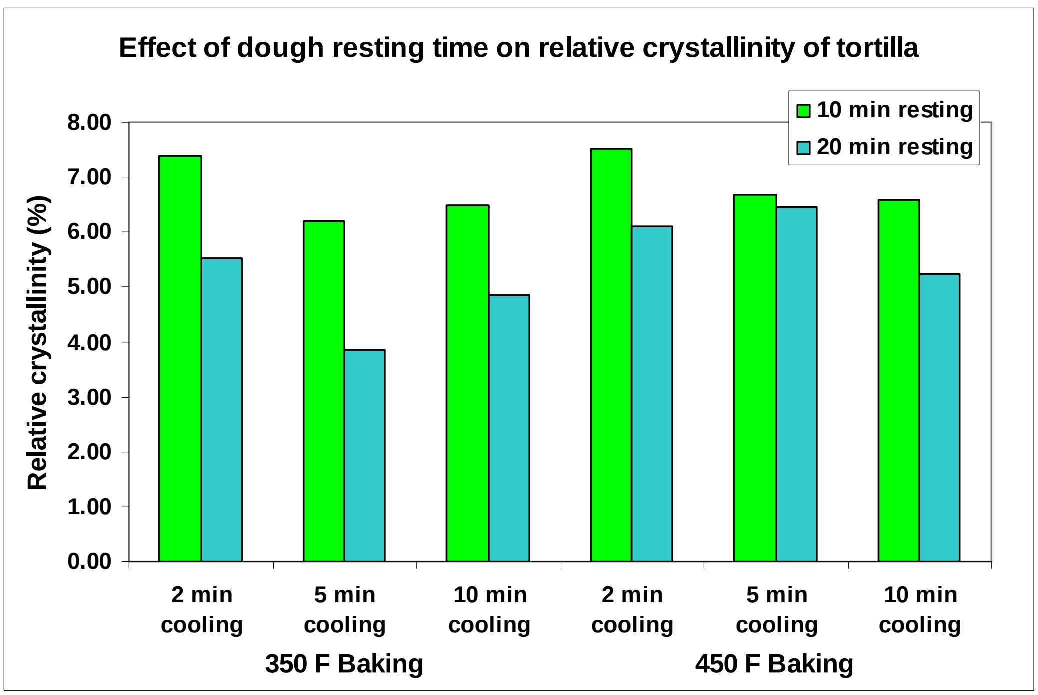

Figure 6 22 : Effect of dough resting time on relative crystallinity of tortillas (see table 6.6)

The lower dough resting time of 10 min gave higher relative crystallinity as compared to 20 min. Dough resting time of 20 min allows starch molecules time to imbibe moisture and it gets evenly distributed in the gluten starch network. Higher dough resting time allows enough time for the gluten network to reduce the stress of mixing and contract to form islands which are slightly connected to each other (Food Chemistry, 3rd edition by Hans-Dieter Belitz, Peter Schieberle, Werner Grosch). This new network reduces loss of moisture from gluten during retrogradation compared to the dough with 10 min resting time resulting in lower crystallinity in dough with 20 min resting time.

6.4.5 Effect of Processing Condition on Tortilla Cellularity

As shown in Table 6.3, to understand the effect of baking temperature on tortilla cellularity T5 with 450˚F baking temperature was compared to T7 which was baked at 350˚F. The baking temperature of 450˚F increased average cell area to 0.023 from 0.009 mm^2. Average cell radius was found higher (0.08 mm) in tortillas baked at 450˚F. Higher baking temperature of 450˚F made tortillas evaporate moisture at a faster rate compared to the lower baking temperature of 350˚F. This resulted in the formation of larger and higher number of air cells (506.64 $cell/cm^2$). Tortilla baking temperature of 450˚F also increased the PDI value. Lower PDI value reflects uniform distribution of air cells. It was noticed that higher baking temperature resulted in increased PDI value. From the above data it can be predicted that a baking temperature of 450˚F might also increased puffing or blistering on the tortilla surface. These puffed spots on the tortilla surface are sticky due to high gelatinization and because they are thinner they can stick to each other very easily.

6.4.6 Effect of Processing Conditions on Surface Energy of Tortillas

As shown in Table 6.7 a baking temperature of 450°F with cooling time and dough resting time of 10 min resulted in the highest total surface energy of 66.7 dyne/cm. When tortillas were baked at 350°F with cooling time of 5 min and dough resting time of 20 min it resulted in lower total surface energy of 54.9 dyne/cm. It was observed that baking temperatures of 350°F and 450°F had mixed effects on surface energy. No strong correlation was observed between surface energy and baking temperature. Baking temperatures of 350°F and 450°F with 10 min of dough resting time and 2 min of tortilla cooling time showed almost the same surface energy (61.4 and 61.1 dyne/cm). But the variation in surface energy was evident at 10 min of tortilla cooling time (60.1 dyne/cm for 350 F and 66.7 dyne/cm for 450 F).

Table 6.7
Effect of Baking Temperature on Total Surface Energy of Tortillas

Tortilla Number	Resting time(min)	Baking Temp(ºF)	Cooling Time(min)	Dispersive (dyn/cm)	Polar (dyn/cm)	Total Surface energy(dyn/cm)
1	10	350	2	25.6	35.8	61.4
2	10	350	5	26.3	31.3	57.6
3	10	350	10	22.2	37.9	60.1
4	20	350	2	25.2	34.2	59.4
5	20	350	5	25.8	29.1	54.9
6	20	350	10	25.2	33.7	58.9
7	10	450	2	25.3	35.8	61.1
8	10	450	5	26.1	32.5	58.6
9	10	450	10	24.4	42.3	66.7
10	20	450	2	26.3	29.0	55.3
11	20	450	5	25.9	33.1	59
12	20	450	10	25.7	31.9	57.6

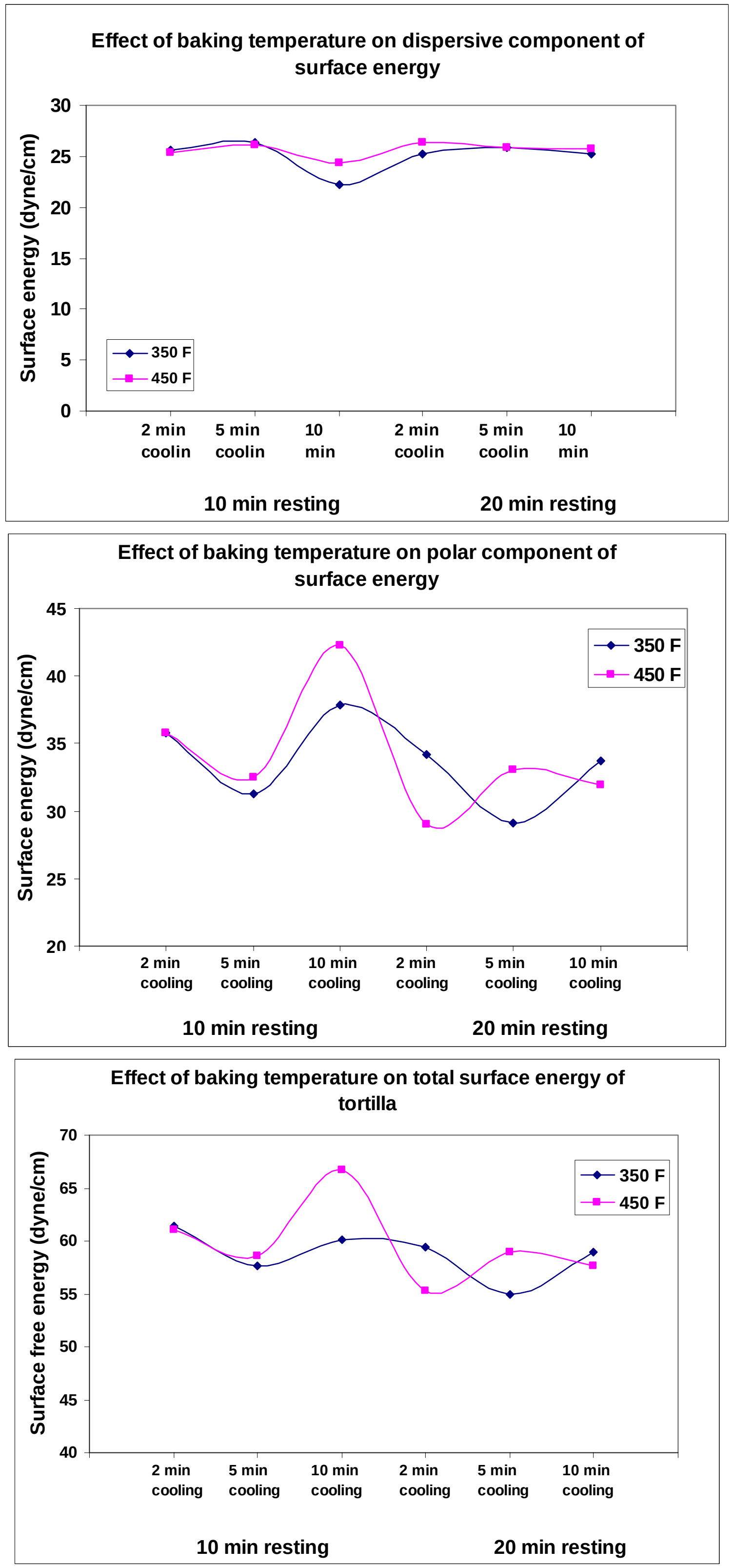

Figure 6 23 : Effect of baking temperature on (a) Dispersive (2) Polar and (c) Total surface energies of tortillas (see table 6.7)

The dispersive (hydrophobic) component of surface energy did not show much difference. The highest polar surface energy was observed at 42.3 dyne/cm for tortillas baked at 450°F with 10 min dough resting and 10 min tortilla cooling. Tortillas baked at 350°F and cooled for 5 min (with 20 min dough rest time) showed the lowest total surface energy of 29 dyne/cm.

Tortilla cooling time of 5 min resulted in lower surface energies as shown in Figures 6.24 and 6.25. Tortillas cooled for 5 min after baking did not show any initial signs of condensation in PE bags. They also did not show dried out surfaces which were found in 10 min cooled tortillas. Tortillas cooled for 5 min with a baking temperature of 350°F and 20 min dough resting time resulted in 29.1 dyne/cm polar surface energy.

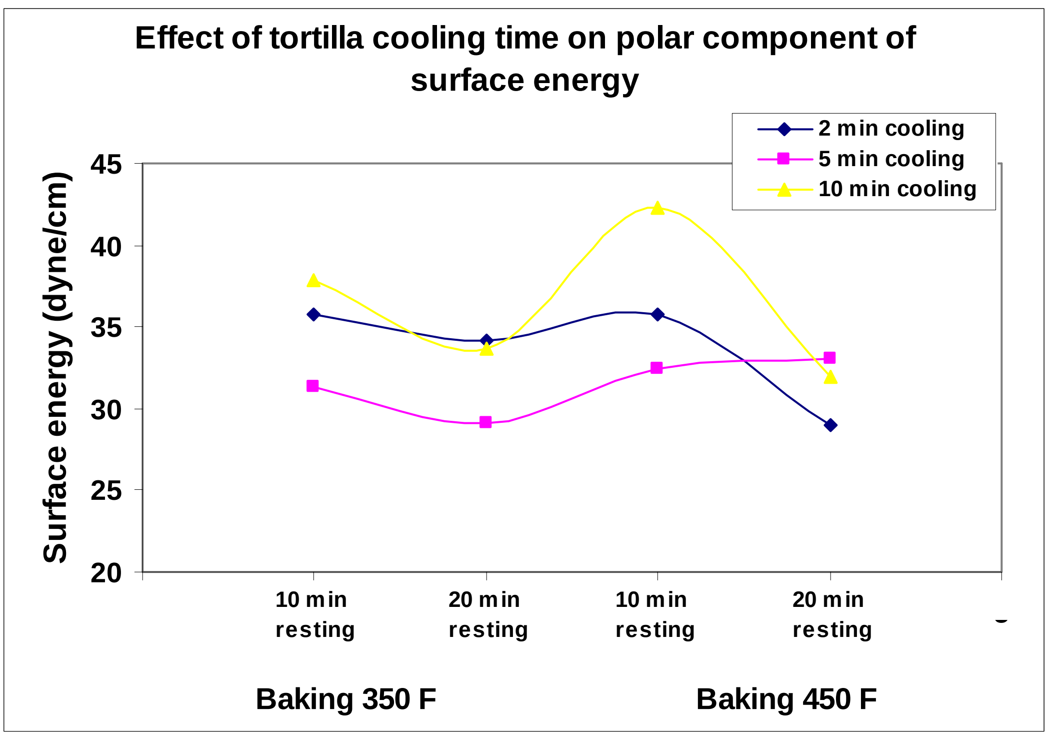

Figure 6 24 : Effect of cooling time on polar component of surface energy (table 6.7)

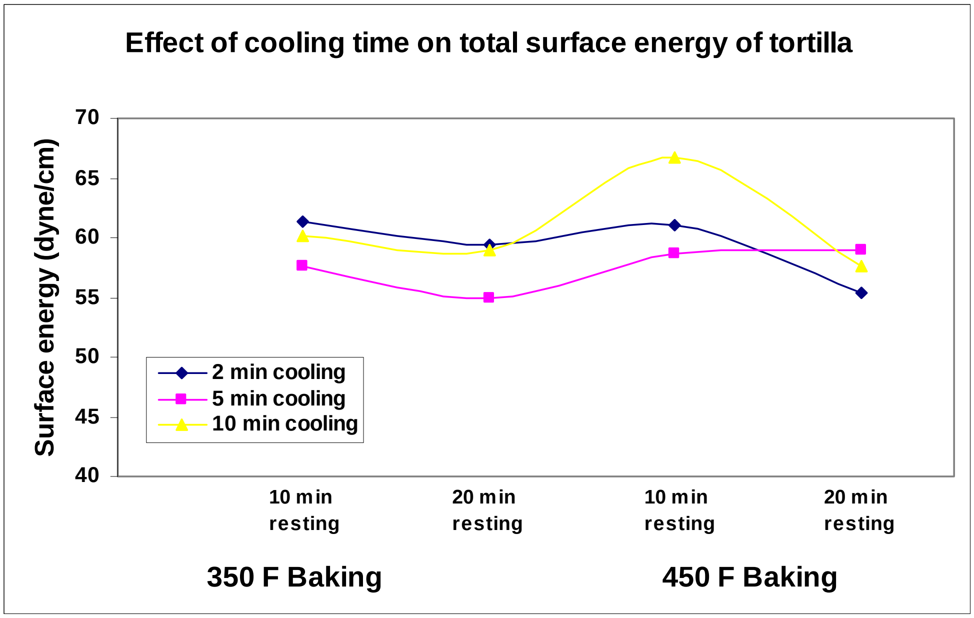

Figure 6 25 : Effect of cooling time on total surface energy of tortillas (table 6.7)

A dough resting time of 20 min resulted in reduced surface energies as shown in Figures 6.26 and 6.27. Tortillas baked at 450°F with tortilla cooling and dough resting times of 10 min resulted in the highest polar surface energy of 42.3dyne/cm. The lower dough resting time of 10 min is not enough for dough to relieve the stress of dough mixing. Higher dough resting time might let the gluten matrix relax, form a stronger network, and bind moisture strongly before baking. This might have increased surface energies of tortillas with 10 min dough resting time.

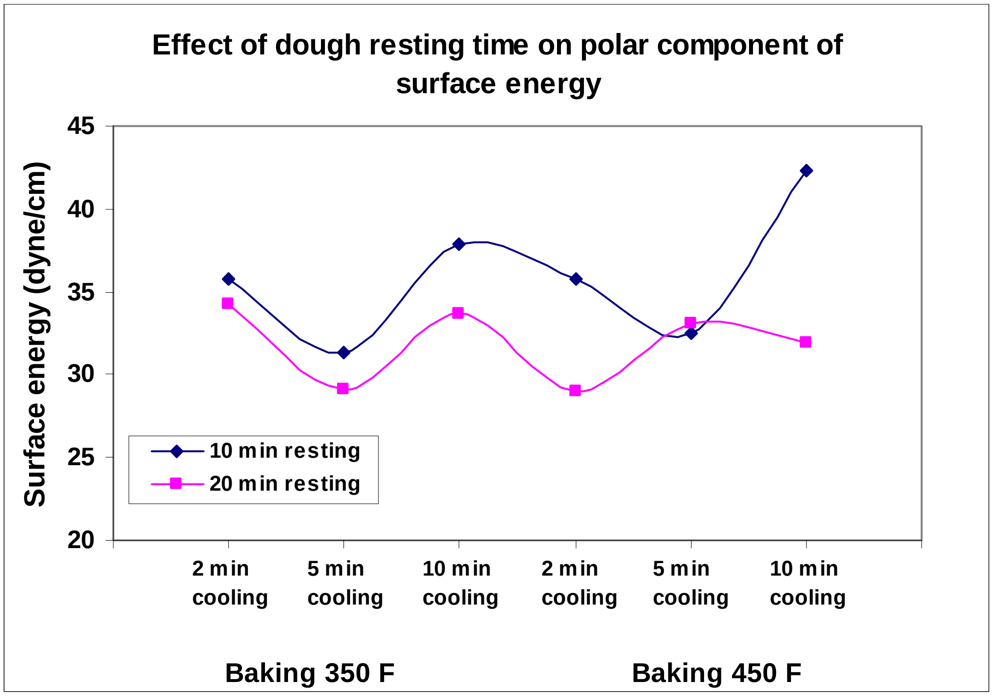

Figure 6 26 : Effect of dough resting time on polar component of surface energy (Table 6.7)

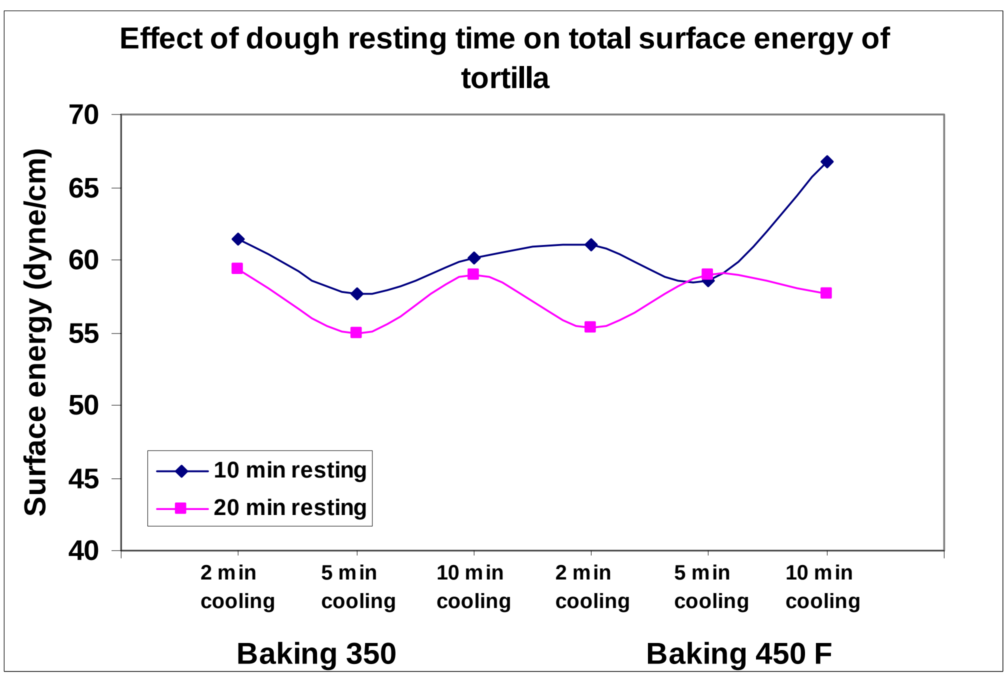

Figure 6 27 : Effect of tortilla dough resting time on total surface energy

6.4.7 Effect of Processing Conditions on Tortilla Stickiness

6.4.7.1 Effect of Baking Temperature on Tortilla Stickiness

As shown in Figure 6.28, tortilla stickiness increased with higher baking temperature (450°F). The lowest tortilla stickiness (135.35 g) was observed when tortillas were baked at 350°F (with 10 min dough resting, 5 min cooling). The highest tortilla stickiness was 731.62 g when tortillas were baked at 450°F with 20 min dough resting and 10 min tortilla cooling time. A baking time of 450°F compared with 350°F might have reduced micro-roughness of the tortilla surface. This might have allowed tortillas to come closer to each other resulting in formation of liquid, solid bridges as explained earlier in this thesis. If the distance between two tortilla surfaces is smaller than 4×10^{-9} m (Adhikari 2001), then chemical bonding (covalent, ionic, metallic, and hydrogen) can take place because of the primary valence forces. These forces are the strongest among of the bondings (Allen, 1993). Higher baking temperature of 450°F might have denatured starch and protein. This might have changed the chemistry of the tortilla surface. This might have led to increased molecular adhesion forces between tortillas. Higher baking temperature might have enhanced the formation of immobile liquid bridges or solid bridges between two tortilla surfaces resulting in higher stickiness. As we discussed earlier, a baking temperature of 450°F (as compared to 350°F) resulted in reduced crystallinity on the tortilla surface. Lower relative crystallinity (see Figure 6.5) led to higher stickiness as discussed earlier.

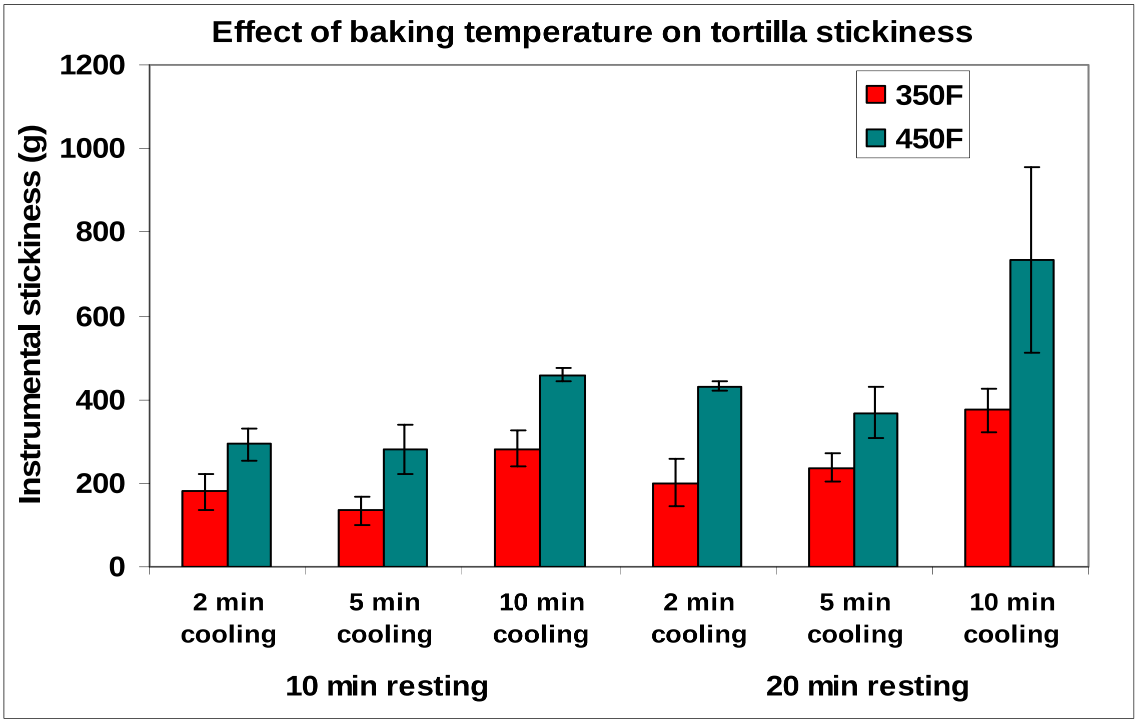

Figure 6 28 : Effect of baking temperature on tortilla stickiness

6.4.7.2 Effect of Tortilla Cooling Time on Tortilla Stickiness

A tortilla cooling time of 5 min was most suitable with the lowest stickiness values. Tortillas baked at 350°F with 10 min dough resting time and 5 min tortilla cooling time had the lowest stickiness of 135.35 g. Tortillas baked at 450°F with dough resting time of 20 min and tortilla cooling time of 2 min resulted in higher (432.38 g) stickiness. Tortilla cooling time of 2 min is not enough. Tortillas were hot (~60 ˚C) when packaged in PE bags. This led to moisture condensation in the bags, which was eventually absorbed by tortillas. This might have resulted in mechanical interlocking as described by Pietsch, 1997. He observed that fibrous, bulky, and flaky particles can interlock or fold about each other resulting in "form-closed" bonds. This interlocking happens at elevated temperatures that cause reduced viscosity and molecules at the interface begin to flow into each other. When the temperature decreases it forms interlocking between the particles (Griffith, 1991). The steam inside hot tortillas might have become trapped between two tortilla layers, condensing upon cooling. This free liquid between tortillas resulted in higher water activity and freezable water as discussed earlier. Tortillas cooled for 2 min had higher crystallinity compared to 10 min cooling (Table 6.6). During crystallization in tortillas the solid dissolved in the liquid might have formed solid bridges at the points of contact upon crystallization. This might have resulted in higher stickiness.

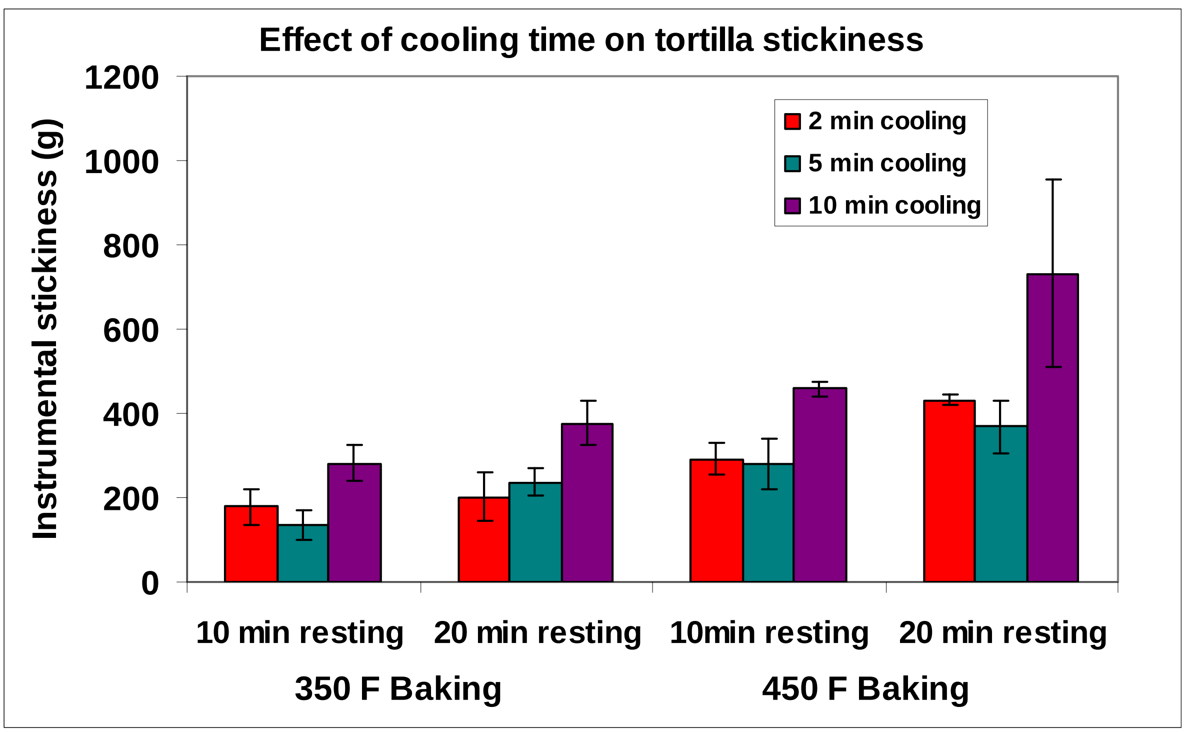

Figure 6 29 : Effect of cooling time on tortilla stickiness

The highest stickiness of 731.62 g was observed when tortillas were baked at 450°F with 20 min dough resting time and 10 min tortilla cooling time. Prolonged cooling of tortillas might have modified tortilla surface topography in a way that promoted stickiness.

6.4.7.3 Effect of Dough Resting on Tortilla Stickiness:

Dough resting time of 20 min resulted in increased tortilla stickiness. The highest stickiness of 731.62 g was observed when dough was rested for 20 min with 450°F baking temperature. Lower crystallinities were observed when dough was rested for 20 min as discussed earlier. Higher dough resting time allows enough time for the gluten network to reduce the stress of mixing and rearrange itself. This new network holds more moisture during retrogradation compared to the dough with 10 min resting time. This might be the reason for lower crystallinity in dough with 20 min resting time. As discussed earlier in this thesis lower crystallinity and therefore higher amorphous material in tortillas results in higher stickiness.

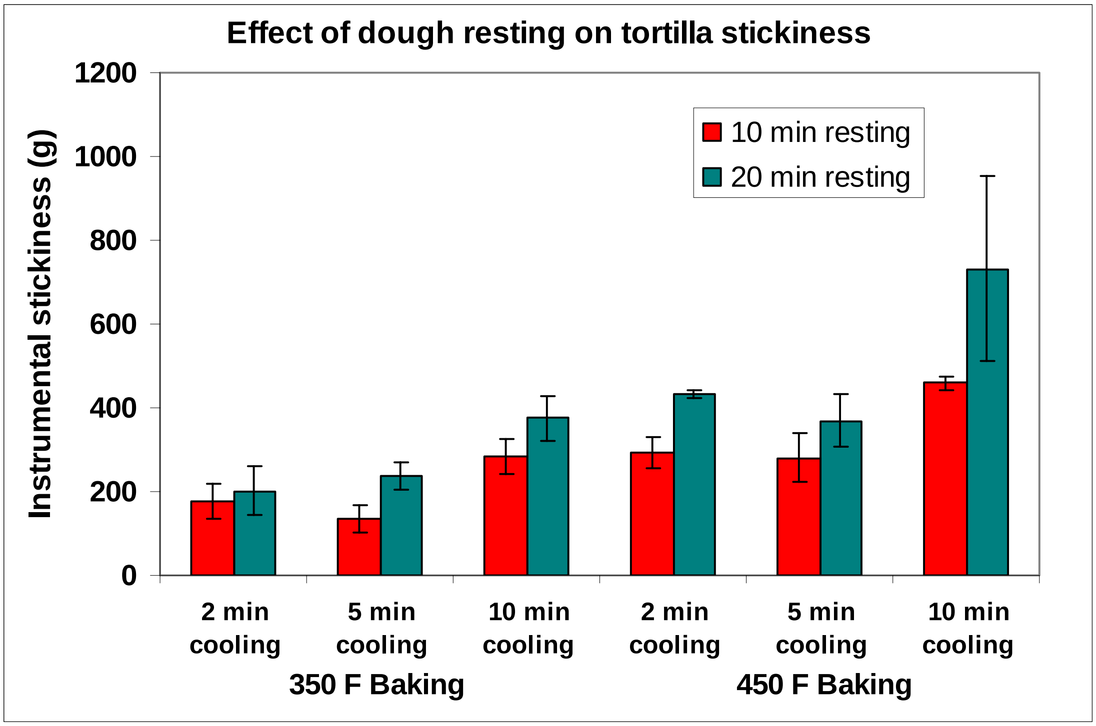

Figure 6 30 : Effect of dough resting on tortilla stickiness

6.5 Effect of Storage Condition on Tortilla Texture

As shown in Figure 6.31 the force required to extend tortillas increased when tortillas were stored at higher temperatures. Tortillas become firmer because of staling or retrogradation of starch. Kelekci et al 2003 observed that changes in protein hydration and association with other components in flour tortillas could be responsible for the increased firming during storage of tortillas. The force required to extend tortillas is lowest at freezer temperature and highest at room temperature. Storage of tortillas at lower temperatures retains freshness as shown by reduced rupture force values. This is because structural polymers are in the glassy state at frozen temperatures and are not mobile. Amylose and amylopectin can not associate or crystallize.

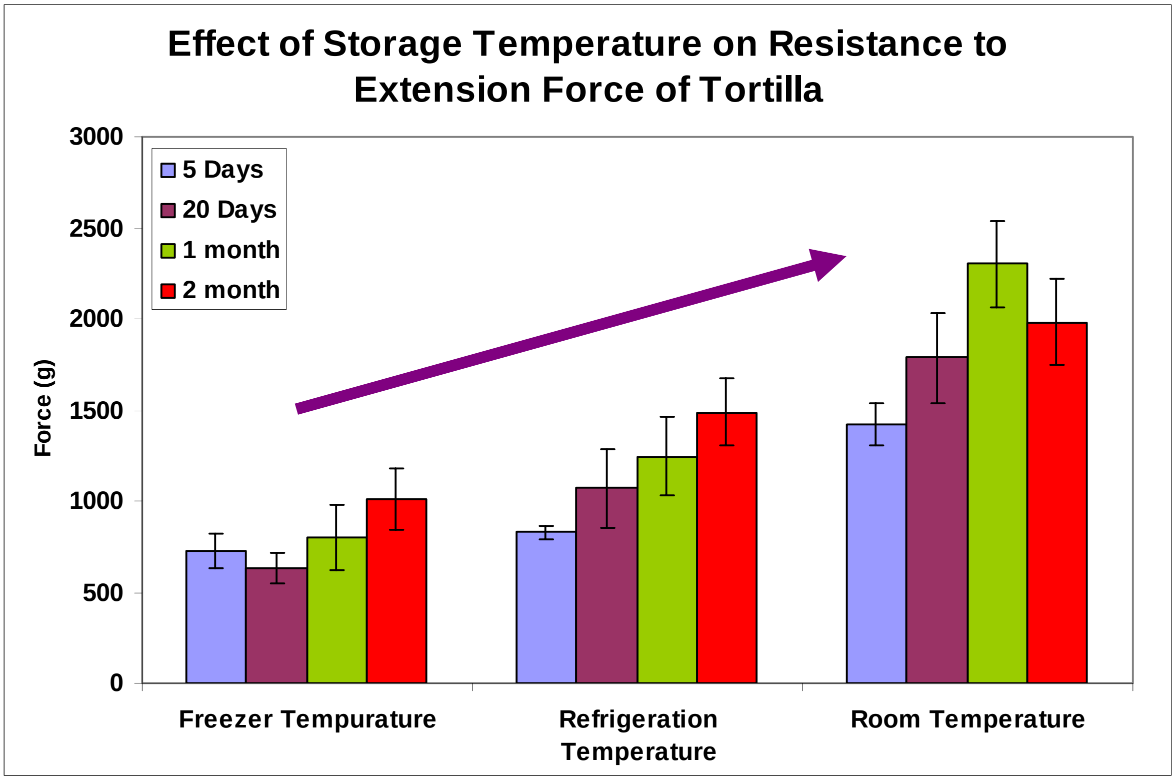

Figure 6 31 : Effect of storage temperature on rupture force of tortilla

As shown in the figure the lowest extension force of 631.56 g was observed for tortillas stored at freezer temperature (-19± 1˚C) for 20 days. The highest tortilla extension force of 2301.22 g was observed when tortillas were stored at room temperature (23 ± 1˚C). This means tortillas retrograded faster at room temperature as compared to freezer and refrigeration temperature (3± 1˚C). As discussed earlier in the thesis the Tg of tortillas is ~-5˚C depending on the formulation and processing conditions. Retrogradation occurs above the Tg. Rate of retrogradation is faster at room temperature. Since the product is stable at freezer temperature and product stability is a function of lack of mobility this can be interpreted to mean that tortillas were in the glassy state at freezer temperature.

Extensibility was observed to be highest when tortillas were stored at freezer temperature. With an increase in storage temperature and time extensibility decreased. This is due to retrogradation as discussed earlier. Tortillas retrograded within 5 days of storage when stored at room temperature. No change in tortilla extensibility was observed after 5 days of storage. The extensibility value showed a sharp decline for tortillas stored at freezer temperature between 1st and 2nd month to 2.77 mm from 7.37 mm. Once again it was seen from the results that at lower temperatures, especially in freezer conditions, tortillas retained freshness. Rubbery tortillas ruptured at long distances.

As shown in Figure 6.33 with an increase in temperature and time of storage the gradient value increases. Higher gradient values suggest lower elasticity of tortillas.

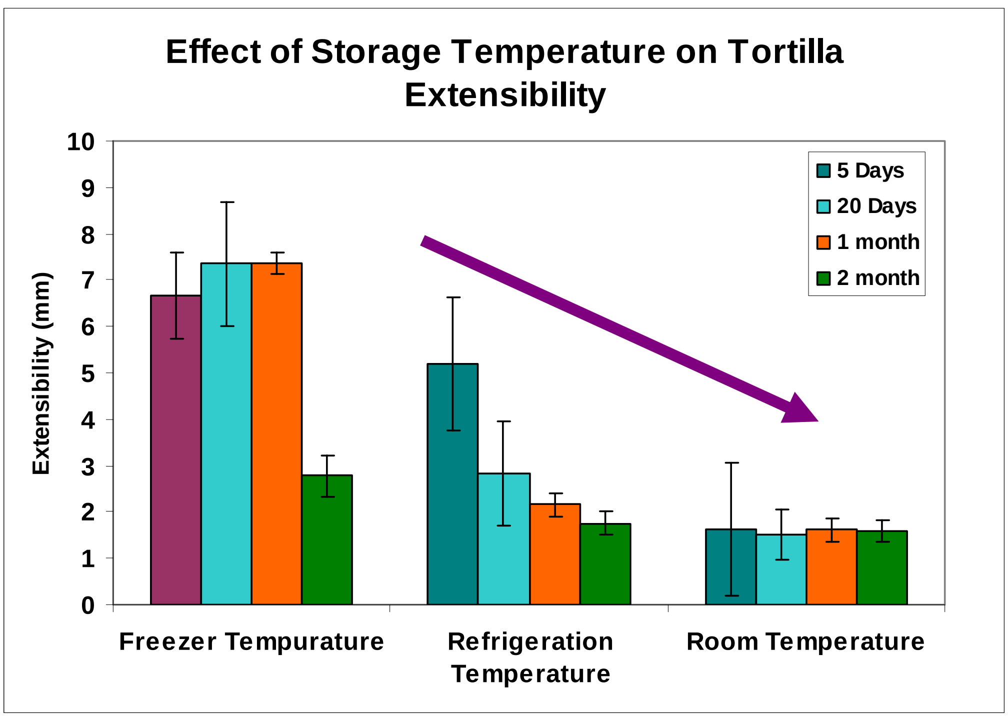

Figure 6 32 : Effect of storage temperature on tortilla extensibility

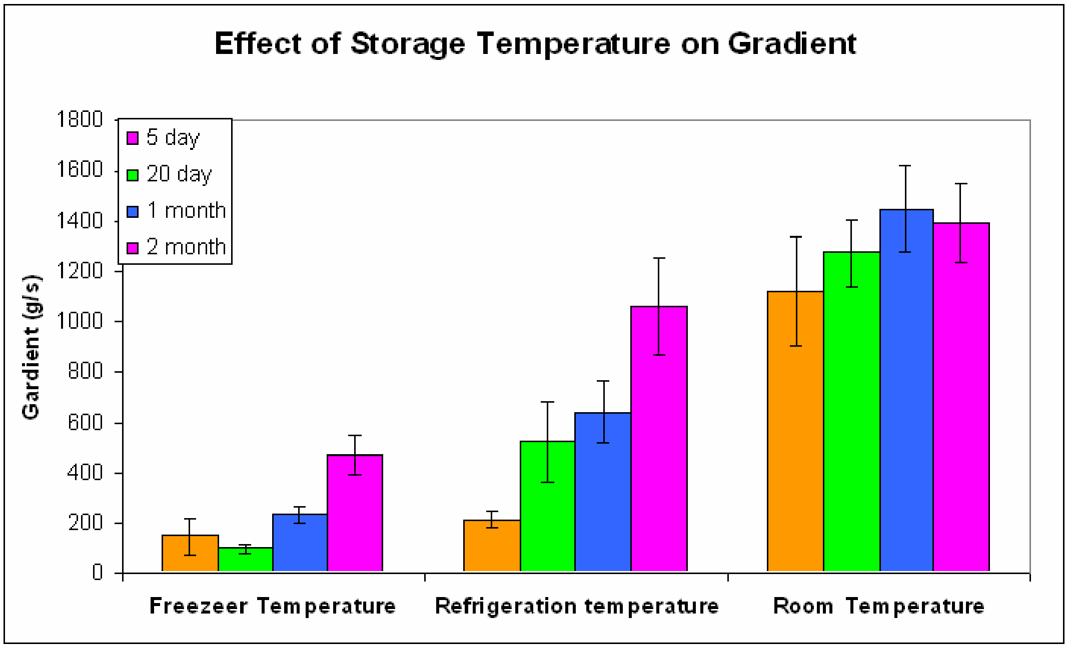

Figure 6 33 : Effect of storage temperature and time on gradient of elasticity

VI. CONCLUSIONS

An objective method to measure tortilla stickiness was successfully developed using a texture analyzer. In this method two sticky tortillas were separated by a rectangular probe and the force (g) required to separate them was measured by a load cell which gave stickiness value. Water activity has a significant impact on tortilla stickiness. The polar component of surface energy, which is an indication of hydrophilic surface energy contributions, was found to correlate positively with increased stickiness. Tortilla samples with high surface energy were shown to stick to each other while those with low polar surface energy did not stick to one another. The state diagram has shown that sticky tortillas showed lower glass transition temperatures compared to non-sticky tortillas. The moisture sorption isotherm indicated availability of high moisture content in sticky tortillas as compared to non-sticky tortillas at a particular water activity level for Aw>0.75. Relative crystallinity of sticky flour tortillas was observed to be lower than that of non-sticky tortillas at the range of water activity levels used. High amorphous regions in sticky tortillas are believed to provide points for stickiness. An increase in product water activity resulted in an increase in surface energy (i.e. high surface hydrophilicity) which in turn caused an increase in instrumental stickiness scores. The dispersive component of surface energy on the other hand, which is an indication of hydrophobic contributions to surface energy, showed a negative correlation with instrumental stickiness. The product composition, processing, and storage history may also have an effect on product stickiness, which needs to be investigated. The results of moisture content, water activity and surface energy measurements of tortillas supported the hypothesis of the project. It was observed that as all fresh tortillas are in the rubbery state at room temperature they are likely to experience stickiness. The sticky tortillas showed slightly lower values of glass transition temperature compared to non-sticky tortillas at given water activities. These results did not support the original hypothesis. Glass transition temperature measurement in a complex food like tortilla is difficult using currently available instruments because they give a range for glass transition temperature not an absolute value.

It can be concluded that stickiness is a complex phenomenon governed by more than one factor such as water activity, moisture content, freezable water present, glass transition temperature, stacking, surface energy, crystallinity, chemical bonding, liquid/solid bridges between tortilla surfaces, intermolecular forces, electrostatic forces, and mechanical interlocking. These factors are correlated to each other such as water content affects water activity or freezable water present in the product which in turn increases surface energy of the product. Some factors are more important than the other. This study found that water activity, polar surface energy and crystallinity are the main factors causing stickiness in tortilla. Still it is clear that not a single factor by itself but the balance of these factors decides stickiness in tortillas.

Addition of hydrophilic ingredients such as CMC, XG, and polyols (glycerol and PG) resulted in increased water retention and decreased free water present in tortillas. Addition of glycerol significantly reduced the water activity of tortillas from 0.94 to 0.91. Addition of gum and glycerol reduced the surface free energy, mainly the polar component of surface free energy. The experimental results have shown a reduction in instrumental stickiness of tortillas when Xanthan gum and CMC were added in tortillas. Low freezable water showing less free water was observed when tortillas were made with gums and glycerol. Lower glass transition temperatures were observed for tortillas made with gums and glycerol.

Higher baking temperature and cooling time reduced the water activity of tortillas. Higher moisture was retained in tortillas when cooled for less time (2 min) after baking. Tortillas cooled for 5 min after baking showed a lower moisture percentage. Tortillas baked at 450°F showed higher stickiness whereas tortillas cooled for 5 min showed reduced stickiness. Highest stickiness was observed when tortillas were cooled for longer time after baking (10 min). Higher baking temperature and cooling time showed a reduced amount of freezable water present in tortillas. Higher baking temperature of tortilla showed increased size and density of air cells. Longer tortilla cooling time resulted in lower cell wall thickness, on the other hand higher dough resting time showed thicker cell walls. Addition of gums and polyols reduced air cell diameter.

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VIII APPENDIX

Fig. 6.1: Correlation between sensory and instrumental stickiness

Tortilla	Instrumental Stickiness	Panelist score
T-1	189.35	-0.376
T-2	362.05	-0.486
T-3	90.21	0.129
T-4	309.44	0.063
T-5	742.94	0.709
T-6	481.4	-0.071
T-7	27.21	-0.479
T-8	123.64	-0.435
T-9	1039.53	0.948

Fig. 6.2: Effect of water activity on instrumental stickiness

Water Activity	T1	T2	T3	T4	T5	T6
0.75	0.00	0.00	0.00	0.00	0.00	0.00
0.84	0.00	0.00	0.00	0.00	0.00	0.00
0.92	62.7±9.38	124.09±13.86	5.66±134	150.55±15.49	164.22±19.24	528.91±72.46
0.94	71.36±7.02	148.15±20.82	12.5±2.88	168.64±18.86	227.07±8.83	715.12±108.94
0.97	247.12±31.58	210.33±8.04	23.71±9.15	212.42±14.96	274.7±27.83	952.5±118.98

Fig. 6.3: Moisture sorption isotherms of sticky and non-sticky flour tortillas

Aw	T1	T2	T3	T4	Aw-T6	T6	Aw-T5	T5
0.13	5.79	5.75	5.83	6.46	**0.18**	6.44	**0.18**	7.36
0.33	7.82	7.90	7.84	7.96	**0.29**	7.00	**0.33**	7.91
0.64	11.79	11.93	12.56	12.87	**0.5**	9.51	**0.51**	10.59
0.75	14.29	14.47	14.67	15.75	**0.57**	10.73	**0.55**	12.18
0.84	19.61	20.97	19.43	21.02	**0.84**	22.71	**0.83**	21.51
0.92	27.37	27.37	27.48	33.38	**0.94**	50.73	**0.92**	34.90
0.94	30.81	31.16	31.74	38.46	**0.95**	55.25	**0.94**	39.25

Fig. 6.4: State diagram of sticky and non-sticky flour tortillas

Water Activity	Non-sticky tortilla	Sticky tortilla
0.13	107.5	100.0
0.33	82.0	81.5
0.43	76.0	70.7
0.64	62.0	59.0
0.84	-2.0	-8.0
0.92	-7.0	-13.0
0.94	-8.0	-14.0

Raw data for Surface energy measurements **Fig.6.7: and Fig. 6.8 .**

Surface Energy Table of tortillas at 0.97 water activity

Type	Liquids	contact angles										AVG	SD	D-SE	P-SE	HSE
T1	F	36.47	31.52	38.67	29.97	44.6	37.13	32.01	38.19	36.91	32.88	35.84	4.34	23.7	30.2	53.9
	DMS	21.39	23.54	20.92	34.39	31.22	28.89	37.37	36.14	27.88	30.02	29.17	5.87			
T2	F	33.95	35.93	32.68	42.49	37.33	40.97	34.37	29.95	35.76	30.28	36.31	4.58	24.4	27.8	52.2
	DMS	20.35	21.44	26.54	30.28	26.24	26.2	34.6	28.72	35.47	23.92	27.38	5.03			
T3	F	31.18	33.99	32.42	32.82	32.68	32.29	37.4	34.76	34.04	38.56	33.89	3.39	23.6	32.4	56
	DMS	30.13	31.48	25.26	29.29	34.08	30.56	28.99	24.6	30.12	24.31	28.44	3.07			
T4	F	29.38	17.38	20.69	30.98	34.99	18.97	20.71	23.01	21	21.96	23.91	5.8	22.1	51.2	73.3
	DMS	26.08	29.61	25.48	30.55	27.09	26.54	32.29	33.04	31.77	34.65	29.27	3.6			
T5	F	22.63	20.47	27.8	31.64	31.38	31.04	28.95	23.37	20.82	19.04	25.71	4.9	22	48.7	70.7
	DMS	27.68	35.21	27.72	28.49	28.4	30.35	33.45	28.13	29.85	29.12	29.84	2.55			
T6	F	25.58	21.3	20.25	18.87	16.45	17.54	26.59	23.75	21.23	19.65	21.01	3.14	20.5	66.8	87.3
	DMS	37.53	33.01	31.22	36.64	31.33	29.07	30.67	28.31	38.04	27.46	33	3.85			

Surface Energy Table of tortillas at 0.94 water activity

Type	liquid	contact angle										AVG	SD	D-SE	P-SE	HSE
T1	F	37.64	31.63	40.04	31.15	46.13	43.99	43.49	32.52	44.81	43.27	39.47	5.84	24.9	24	48.9
	DMS	25.24	33.86	27.49	27.03	24.92	32.2	26.88	26.69	29.01	25.17	27.95	2.88			
T2	F	43.52	42.77	39.48	33.62	40.04	37.8	38.42	35.61	38.36	37.51	38.67	2.72	24.3	25.9	50.2
	DMS	21.31	23.74	21.01	24.25	35.25	37.63	38.47	26.35	32.07		28.9	7			
T3	F	35.84	39.22	33.44	39.85	40.75	40.04	42.24	41.74	43.53	42.26	39.44	4.15	23.4	27.3	50.7
	DMS	34.96	26.17	36.84	28.77	34.8	32.13	25.38	37.89	31.12	28.01	31.6	4.45			
T4	F	35.04	30.2	33.37	38.28	32.87	28.64	30.07	32.39	37.99	36.83	33.03	3.91	21.4	41.4	62.8
	DMS	37.37	29.76	36.94	32.5	29.89	30.27	28.94	38	39.91	36.98	34.06	4.17			
T5	F	47.69	42.38	47.66	41.17	38.34	38.35	47.69	42.38	33.84	41.17	41.73	4.52	18.1	42.6	60.7
	DMS	43.51	41.38	47.55	50.36	44.99	40.29	40.67	44.95	50.86	45.03	45	3.77			
T6	F	37.87	30.19	30.99	32.74	33.1	30.21	34.25	37.89	28.78	32.72	32.87	3.1	18.7	56.9	75.6
	DMS	41.72	39.12	40.16	45.25	42.68	42.69	42.67	38.13	42.61	37.12	40.81	2.74			

Surface Energy Table of tortillas at 0.92 water activity																
Type	liquid	contact angle										AVG	SD	D-SE	P-SE	HSE
T1	F	35.7	36.7	33.52	35.1	36.8	36.49	34.9	36.1	36.8	–	–	35.8	1.11	25.5	25.9	51.4
	DMS	27.55	27.2	24.28	27.3	24.1	23.42	21	19.7	24.2	–	–	24.2	2.62			
T2	F	40.55	40.7	35.77	32.3	35.4	30.86	41.3	39.8	37.8	38	39.5	37.5	3.5	26.7	22.3	49
	DMS	22.63	23.2	14.78	23	21.8	22.8	21	23.2	23.5	23.6	19.3	21.7	2.63			
T3	F	40.08	41.8	47.61	35.4	37.7	33.79	35.9	36.4	36.3	36.7	42.1	38.5	4.01	25.3	24	49.3
	DMS	29.83	32.3	30.72	24.6	27.8	22.85	22.5	27.4	24.5	24	23.5	26.4	3.42			
T4	F	27.99	26.9	25.66	28.6	30.2	29.83	32.2	27	27.3	32.2	35	29.4	2.83	24.6	34.1	58.7
	DMS	25.3	26.7	19.58	22.8	21.3	26.71	27.9	17.8	–	–	–	23.5	3.71			
T5	F	38.63	30.6	37.73	36.3	37.1	36.43	35.6	38.1	36.7	35.9		36.3	2.23	23.1	31.2	54.3
	DMS	29.79	32.6	30.9	41	25.6	27.35	31.7	27.5	–	–	–	30.8	4.76			
T6	F	28.14	28.4	27.78	27.9	21.2	24.22	25.1	26.1	20.6	–	–	25.5	3	22.4	47.1	69.5
	DMS	29.94	27.5	32.76	33.7	32.2	28.38	26.3	27.6	25.4	26.7	25.1	28.7	3.04			

Surface Energy Table of tortillas at 0.84 water activity

Type	Liquid	contact angle										AVG	SD	D-SE	P-SE	HSE
T1	F	43.9	37.5	42.97	40	39.3	47.76	48.9	46	52.6	51.59	45.63	4.93	28.2	14.9	43.1
	DMS	22.6	32.8	32.31	20.8	21.2	23.06	25.7	17.8	24.8		24.56	5.08			
T2	F	31.5	31	37	32.5	46.2	33.3	40.8	37.1	38		36.38	4.95	27.7	21.4	49.1
	DMS	12	20.4	14.43	17.8	15.3	18.23	17.6	17.7	22.5	21.92	17.78	3.29			
T3	F	39.7	40.8	33.22	32.4	36.7	44.32	39.9	47.8	45.8	48.79	40.39	5.73	29	17	46
	DMS	22.3	14.2	16.16	13.7	12.4	16.72	20.5	23.1	14.2		17.2	3.79			
T4	F	36.2	37.8	38.96	38.8	39.5	40.87	41.6	40.6	42.8	35.36	39.2	2.64	27.8	19.4	47.2
	DMS	23.1	14.8	17.8	19.8	23.3	19.53	23.2	18.8	14.3		20	3.71			
T5	F	43.1	43.6	41.28	39.1	35.9	39.85	34.9	44.2	39.5		40.16	3.27	28.3	18.1	46.4
	DMS	26.3	21.2	21.84	15.5	13.7	12.69	21.9	23.9	20.1	20.29	19.29	4.45			
T6	F	37.4	36.3	37.3	35.7	36	33.83	32.4	34.5	37	36.01	35.82	1.56	23.3	31.2	54.5
	DMS	31.3	34.1	34.99	28	29.1	21.36	18.2	26.9	35.9	35.9	30.12	6.09			

Surface Energy Table of tortillas at 0.75 water activity

Type	liquid	contact angle										AVG	SD	D-SE	P-SE	HSE
T1	F	45.3	49	44.73	46.4	44.6	41.81	43.3	47.5	46.3	47.82	45.67	2.18	28	15	43
	DMS	28.3	27.3	20.4	22.8	22.1	21.04	25.9	24.5	27.2	26.92	24.9	2.6			
T2	F	44.5		41.51	37.7	43.2	42.23	42.4	37.9	40.2	51.89	42	4.16	29.5	15.5	45
	DMS	20.4	20.5	11.72	23.1	12.1	19.04	21	20.3	13.7	14.61	17.64	4.16			
T3	F	37.5	40.8	37.03	41.4	45.8	38.28	42.3	46.7	42.8	38.68	41.13	3.2	30.2	15.2	45.4
	DMS	16.3	9.4	14.6	11.1	10.5	21.39	20.2	11.5	12.1	12.26	13.94	4.12			
T4	F	46.3	45.7	42.72	43.8	42.3	46.69	41.9	48.5	41.8	39.11	43.58	2.89	29.3	14.8	44.1
	DMS	20.7	17.7	21.2	20.3	22.8	19.85	20	12	21.5	22.38	19.79	2.95			
T5	F	44.1	43	39.06	45.4	39	43.19	38.4	40.5	44	41.81	41.84	2.66	29.3	15.9	45.2
	DMS	16.9	17.2	19.39	20.3	19.1	18.32	16.1	19	19.5	15.04	18.08	1.7			
T6	F	36.5	39.2	35.87	37.7	36.2	37.19	33.3	32.7	43	47.8	37.4	4.28	26.2	23.2	49.4
	DMS	19.3	22.2	16.59	24	27.2	21.22	20.7	26.9	27.9	26.45	23.2	3.7			

Fig. 6.12: Effect of addition of gums and polyols on tortilla water activity

Tortilla type	Water Activity
Control tortilla	0.939±0.006
Gum tortilla	0.943±0.00
Gum and glycerol tortilla	0.895±0.010
Glycerol tortilla	0.910±0.009
Propylene glycol tortilla	0.910±0.005
Gum and propylene glycol	0.907±0.007

Fig. 6.13: Effect of addition of gums and polyols on tortilla moisture content

Tortilla type	Tortilla moisture %	STDEV
Control tortilla	30.13	0.13
Gum tortilla	28.35	0.16
Gum and glycerol tortilla	27.79	0.54
Glycerol tortilla	26.46	0.1
Propylene glycol tortilla	25.79	0.34
Gum and propylene glycol tortilla	24.57	0.07

Fig. 6.15: Effect of addition of gums and polyols on freezable water

Type of tortilla	FW-1 (mJ)	FW-2 (mJ)	AVG FW(mJ)	STDEV
Control Tortilla	72.8	72.33	72.57	0.33
Gums Tortilla	64.83	40.66	52.75	17.09
Glycerol Tortilla	8.94	9.91	9.43	0.69
Gums and Glycerol Tortilla	8.12	8.4	8.26	0.20
Propylene glycol Tortilla	6.25	8.64	7.45	1.69
Propylene glycol and gums Tortilla	9.53	10.85	10.19	0.93

Fig. 6.20: Effect of addition of CMC and XG gums and polyols on tortilla surface energy

Type of	Dispersive surface energy (dyne/cm)	Polar surface energy (dyne/cm)	Total surface energy (dyne/cm)
Control tortilla	21.8	41.9	63.7
Gum Tortilla	24.7	30.2	54.9
Gum and Glycerol Tortilla	21.5	29.4	50.9
glycerol tortilla	24.5	32.4	56.9
propylene glycol tortilla	23.8	39.9	63.7
gum and propylneglycol	25.3	27.5	52.8

Fig. 6.21: Effect of addition of gums and polyols on tortilla stickiness

Type of Tortilla	Instrumental Stickiness (g)	STDEV
control	211.0513	72.74739
Gum tortilla	155.9067	42.2693
Glycerol tortilla	65.40636	18.39091
Gum and Glycerol tortilla	156.67	14.56497
Propylene glycol tortilla	185.8167	43.61478
Gum and Propylene Glycol	122.93	26.9461

Fig. 6.28, 6.29 and 6.30: Effect of processing condition on tortilla stickiness

Tortilla Number	Resting time(min)	Baking Temp(ºF)	Cooling Time(min)	STICKINESS (g)	STDEV
1	10	350	2	282.3	41.91
2	10	350	5	178.88	42.04
3	10	350	10	135.35	34.17
4	20	350	2	201.47	58.49
5	20	350	5	236.95	33.36
6	20	350	10	375.3	52.32
7	10	450	2	432.38	11.12
8	10	450	5	368.99	62.73
9	10	450	10	731.62	221.73
10	20	450	2	292.18	36.6
11	20	450	5	280.89	59.11
12	20	450	10	459.01	17.04

Fig.6.31: Effect of storage temperature on rupture force of tortilla

Storage time (days)	Force at Freezer Temperature (g)	Force at Refrigeration Temperature (g)	Force at Room Temperature (g)
5	722.95±94.79	827.88±81.69	1421.65±176.98
20	631.576±40.08	1072.47±216.69	1785.56±218.66
1 month	802.73±118.48	1247.13±246.59	2301.22±237.14
2 month	1011.69±169.64	1489.09±186.23	1984.11±234.45

Fig.6.32: Effect of storage temperature on tortilla extensibility

Storage time (days)	Extensibility at Freezer Temperature (mm)	Extensibility at Refrigeration Temperature (mm)	Extensibility at Room Temperature (mm)
5	6.67±0.94	5.19±1.33	1.62±0.23
20	7.36±1.45	2.84±1.12	1.52±0.25
1 month	7.37±1.43	2.17±0.56	1.61±0.24
2 month	2.77±0.44	1.76±0.26	1.59±0.22

Fig.6.33: Effect of storage temperature and time on gradient of elasticity

Temperature of Storage	5 day (g/mm)	20 day(g/mm)	1 month(g/mm)	2 month(g/mm)
Freezeer	148.85±74.92	101.24±15.47	235.41±35.55	472.23±77.45
Refrigeration	216.45±34.29	523.72±160.00	641.989±125.89	1062.58±190.91
Room	1121.76±216.74	1273.58±136.73	1447.98±171.55	1395.68±158.2