Investigation of the impact of different sugar compositions on the moisture absorption of hard candy when exposed to a humid environment

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June 2023

Abstract

Hard candy is hygroscopic, implying that hard candy readily absorbs moisture from the environment, resulting in adverse changes in the texture and appearance of the candy. This thesis investigates the impact of different sugar compositions on the moisture absorption of hard candy when exposed to a humid environment.

Five different hard candy formulations were exposed to an 8-day storage experiment at 75% relative humidity, allowing the candy to change over time. Moisture content, water activity, crunchiness, colour diffusion, and moisture absorption were analysed throughout the experiment. ANOVA and ImageJ were used to analyse the data.

Promising outcomes were observed in formula 1 (9,8 DE) regarding reduced moisture absorption and improved stability of crunchiness and colour diffusion compared to the other formulations. A correlation between molecular weight sugars and moisture absorption was observed. The absence of hygroscopic low molecular weight sugars combined with the presence of high molecular weight sugars mitigates moisture absorption. This saccharide composition also increases the stability of colour diffusion and crunchiness, as in formula 1 (9,8 DE). The moisture absorption of all formulations showed an initial increase, implying rapid moisture absorption at the surface. Followed by a slower increase due to the decreasing driving force. Then, an exponential increase in moisture absorption is observed with a high calculated diffusion coefficient. This data indicates a phase transition from a glassy to a rubbery state with high mobility, resulting in a higher moisture migration rate. Formulas 1 (9,8 DE) and 2 (13 DE), with a small amount of low molecular weight saccharides, show lower diffusion coefficients, indicating a delayed transition to the rubbery state and better retention of crunchiness.

Popular science summary

Hard candy readily picks up moisture from the air, resulting in a soft and sticky texture causing an end of shelf life. This research investigates the influence of different sugar compositions on the moisture uptake of hard candy when exposed to a humid environment.

Five different hard candy recipes were exposed to a humid environment for 8 days, allowing the candy to change over time. Several measurements were performed during the 8-day storage experiment.

Formula 1 (9,8 DE) showed the most promising results based on reduced moisture uptake and increased stability, which indicates an extended shelf life. Formula 2 (13 DE) had almost the same effectiveness. In contrast, formula 4 (27 DE) showed the highest moisture uptake. A relation between moisture uptake and the sugar composition of the hard candy was observed. The moisture uptake of all recipes showed first an increase, indicating a rapid moisture uptake at the surface. Followed by a slower increase in moisture uptake due to the decreasing difference between the water activity of air and candy. Then, a steep increase in moisture uptake is observed which indicates a decreased viscosity. As moisture uptake increases with decreasing viscosity.

Further research is recommended to understand better the impact of different hard candy recipes regarding processability and physical characteristics such as hardness and sweetness. The low DE hard candy recipes, such as formula 1 (9.8 DE), might decrease the processability and sweetness and increase hardness which may be unacceptable to customers.

Acknowledgements

Table of Contents

List of abbreviations

Abbreviation	Definition
ANOVA	Analysis of variance
aw	Water activity
DW	Dry weight
DE	Dextrose equivalent
DP	Degree of polymerisation
DS	Dry substance
MC	Moisture content
n	Number of observations
RH	Relative humidity
SEM	Standard error of the mean
T	Time
Tg	Glass transition temperature
U	Water uptake
W	Water content
WW	Wet weight

1. Introduction

OV Sweden – Candeco is a confectionery company under the Orkla Group, which specialises in the production of a diverse range of sugar-based products, including candies, sprinkles, and sugar pearls. These products are used as decoration or inclusion in ice cream, bakery and chocolate applications. As experts in the field, OV Sweden - Candeco recognises that these sugar-based products, particularly hard candies, exhibit high hygroscopicity, which implies that those products readily absorb moisture from the environment. This hygroscopicity negatively affects product quality and shelf life. (OV group, n.d.)

Hard candy becomes sticky when exposed to a humid environment, for instance in the summer. It loses its crunchy texture, and the colour diffuses out of the product. Addressing the hygroscopicity of hard candy is crucial to enhance product stability and extend shelf life. Altering the sugar composition of hard candy offers a promising approach to mitigate these challenges.

2. Aim

The aim of this thesis is to investigate the impact of different sugar compositions on the moisture absorption of hard candy when exposed to a humid environment.

This thesis is conducted in cooperation with OV Sweden – Candeco and Lund University. OV Sweden – Candeco currently produces hard candy on the batch and continuous line but aims to improve the hard candy on the continuous line as it is more prone to moisture absorption. Thus, this thesis focuses on hard candy formulations that can run on a continuous line process as crystallisation is a higher risk on a continuous line. Therefore, more than 30% (dry weight) of glucose syrup must be added to prevent crystallisation (Hartel et al., 2018).

3. Theoretical background

3.1. The composition

Hard candy is a subcooled, supersaturated sugar solution with most water removed by boiling (moisture content approximately 1-3%) (Ergun et al., 2010). Hard candy is cooled below the glass transition temperature (Tg), where it transforms into an amorphous sugar glass (Hartel, 2002). The tremendously high viscosity of the viscous liquid results in solid-like properties when held below the Tg, meaning that the hard candy will not flow for an extended period. (Hartel et al., 2018; Netramai et al., 2018)

The base of hard candy is typically a mixture of glucose syrup and sucrose with minor ingredients such as flavour, colour and acids (Netramai et al., 2018). The sucrose-glucose syrup ratio is chosen according to the desired characteristics of the final product. A high glucose syrup content (at least 30% dry weight) inhibits the crystallisation (graininess) of sucrose during storage (Hartel et al., 2018). Nevertheless, hard candy with increased amounts of glucose syrup results in a higher degree of stickiness and lower sweetness (Netramai et al., 2018). In addition, the type of glucose syrup also affects the desired properties. Those syrups are categorised with dextrose equivalent (DE) value. The most common one in hard candy is 42 DE glucose syrup (Hartel et al., 2018). A higher DE syrup contains many low molecular weight sugars leading to a lower Tg resulting in a product which is more prone to stickiness (Spanemberg et al., 2019). Thus, the physical characteristics of hard candy strongly depend on the amount and composition of glucose syrup.

3.2. The process

Hard candy is made in various ways. The first step in the process is to mix sucrose, glucose and water. The sucrose must be completely dissolved at the end of the process to avoid crystals in the final glassy product. As the syrup boils, the water content decreases, and the boiling point increases. This phenomenon is called boiling point elevation, where the boiling point increases as more solids are in the solution. The final boiling temperature correlates with the final water content of the confectionery (Hartel & Nowakowski, 2017). Each sugar mixture has its own boiling point elevation curve, which depends on the sucrose-glucose syrup ratio. (Ergun et al., 2010; Hartel et al., 2018)

At atmospheric pressure, very high temperatures (around 146-155°C) are reached to establish a water content of 1-3%. Chemical changes such as sucrose inversion, reversion or discolouration can occur more rapidly at high temperatures. Therefore, vacuum cookers are used in the last step of the boiling process to achieve the final water content without negative chemical changes. Under vacuum, the boiling point is lower than at atmospheric pressure. As a result, an identical moisture content can be achieved at lower temperatures. In addition, the stirring blades continuously sweep the surface to ensure rapid heating to prevent chemical changes. (Hartel et al., 2018)

After boiling, the candy is cooled quickly to prevent sugar crystallisation (Hartel et al., 2011). Colours, flavours and acids are usually added at this stage to prevent chemical changes caused by extreme temperatures (Hartel & Nowakowski, 2017). Cooling continues until the mass reaches around 80-88°C and can be formed into the desired shape. However, this temperature range depends on the recipe. Forming temperatures are higher when the Tg of the candy is higher. The candy is more prone to crystallisation at the forming temperature due to supersaturation. However, the inhibiting properties of glucose syrup and its high viscosity slow down the rate of crystal nucleation, allowing the candy to be formed and preventing crystallisation. (Hartel et al., 2018)

3.3. Physicochemical properties of sugars

As mentioned above, sucrose and various glucose syrups are used in hard candies. Glucose syrups are obtained through the hydrolysis of starch. These sugars are characterised by a DE value (Dokic et al., 2004). This value indicates the degree of starch hydrolysis in the glucose syrup (Nowakowski & Hartel, 2002).

Glucose (monosaccharide) has a DE value of 100, and maltose (disaccharide) has a DE value of 58 (Hartel et al., 2018). The DE value of the saccharide decreases as the degree of hydrolysis decreases. Low molecular weight saccharides contribute significantly to the total DE value of the glucose syrup. Therefore, a high DE glucose syrup contains noticeably more low molecular weight molecules, and a low DE glucose syrup contains many high molecular weight molecules. Moreover, high DE glucose syrups are more hygroscopic and absorb moisture quickly due to the high amount of low molecular weight sugars (Ergun et al., 2010).

However, it should be acknowledged that DE values of glucose syrups can be the same but with different saccharide compositions (Hartel et al., 2011). Furthermore, the DE value of the overall candy can also be calculated. It is worth noting that sucrose does not contribute to the DE value as it has a value of 0. Therefore, the presence of sucrose significantly reduces the overall DE value.

3.3.1. Boiling point elevation

The final boiling point of the process controls the final water content of candy through the boiling point elevation. This parameter is critical in the confectionery industry as the final moisture content is linked to product quality, texture and shelf life. (Hartel et al., 2011).

Several factors influence the boiling point of a sugar solution. This boiling point is reached when the vapour pressure of the liquid equals the ambient pressure. One of these factors is the molecular weight of the sugar. Low molecular weight saccharides, such as glucose, raise the boiling point. In addition, the concentration of sugars in the solution also affects the boiling point. As the sugar syrup starts to

boil, water evaporates, and the concentration of solutes increases. The boiling point increases due to the higher concentration of sugar. The interaction between the water and the sugar molecules causes the vapour pressure to decrease and the boiling point to increase. The sugar solution must be heated to higher temperatures to achieve equilibrium between vapour and ambient pressure. (Ergun et al., 2010; Hartel et al., 2018)

Another factor is the pressure during the boiling process. The boiling temperature depends on the ambient pressure. The equilibrium between vapour and ambient pressure is reached more quickly at low ambient pressure. Therefore, pressure below atmospheric pressure reduces the boiling temperature because less energy is required to boil. (Roos & Drusch, 2016b)

A suitable boiling point elevation curve is required to determine the final boiling temperature for the specific sugar mixture. The boiling point of combined sugars mainly ranges between those two sugars. The weight percentage of each sugar determines the actual value of the boiling point. Hard candies with high DE glucose syrup have higher boiling points due to their low molecular weight. However, glucose syrup with the same DE-value could have noticeably different saccharide profiles, resulting in a different boiling point. A change in moisture content may occur when a confectionary manufacturer switches between glucose syrups with the same DE-value. (Hartel et al., 2011)

3.3.2. Glass transition temperature

The glass transition can be described as the transformation of a viscous liquid into a solid-like glass due to a change in temperature and/or water content (Spanemberg et al., 2019). The Tg is not a specific temperature point, but the transition takes place in a temperature range (Hartel & Nowakowski, 2017).

The Tg can indicate shelf stability and texture. A significant increase or decrease in Tg will affect those factors. Hard candy is relatively stable as the storage temperature stays above the Tg and is not exposed to moisture. Tg values between 40-45°C are generally the most stable hard candies, and Tg values near room temperature are less stable and prone to stickiness (Hartel & Nowakowski, 2017). Tg values higher than 50°C are quite stable but would cause negative physical characteristics. The candy becomes too brittle and hard with sharp edges. Simply increasing the Tg is not the solution to increase the shelf life. A balance is required between the stability and physical characteristics of hard candy to determine an appropriate Tg. (Ergun et al., 2010; Hartel et al., 2018)

The Tg depends on the molecular weight and moisture content of the candy (Šmídová et al., 2003). The Tg increases when many high molecular weight sugars are added. Table 1 illustrates different compounds and their T_g.

Table 1: Glass transition of different compounds

Compound	T_g (°C)
Glucose	31
Maltose	87
Sucrose	62-70
42 DE corn syrup solids	79
20 DE corn syrup solids	139

Note. Adapted from Ergun et al., 2010

Table 2 illustrates the effect of the Tg on mixed sucrose -corn syrup. The mixture of sucrose and 42 DE corn syrup only changes the Tg noticeably until about 50% of the solution is 42 DE corn syrup. The 20 DE corn syrup, with substantially higher molecular weight, leads to a significant rise in Tg already at low concentrations. (Ergun et al., 2010)

Table 2: Glass transition temperature of sucrose-corn syrup (dry weight)

Additional levels of corn syrup (%)	20 DE	42 DE
0	63.9 ± 1.4	63.9 ± 1.4
10	66.8 ± 0.4	64.1 ± 2.2
20	65.8 ± 2.6	66.9 ± 0.7
50	73.6 ± 3.0	69.1 ± 2.0
75	91.6 ± 1.3	70.9 ± 1.6
100	139.3 ± 0.8	79.0 ± 2.9

Note. From Ergun et al., 2010

Thus, the additional 42 or 20 DE corn syrup increases the Tg. Conversely, sugar mixtures with inverted sugars or higher DE glucose syrups substantially decrease the Tg due to the molecular weight. (Ergun et al., 2010; Hartel et al., 2018)

Moreover, the water content in the sugar solution extensively affects the Tg due to the plasticizing effect (Kawai et al., 2019). Minor changes in the final water content can already substantially impact the Tg (table 3) (Ergun et al., 2010). The water content of hard candy is between 1-3% (Ergun et al., 2010). As seen in Table 3 this can change the Tg substantially. Therefore, it is crucial to control the water content of the candy during manufacturing (Hartel et al., 2018).

Table 3: Glass transition temperature of sucrose in relation to water content

Water content (%)	T_g (°C)
0	69
1	61
2	50
3	42

Note. Adapted from Ergun et al., 2010

3.4. Instabilities and shelf life of hard candy

Hard candy is a relatively stable product if stored below its Tg, it could be stored for up to a few years even though it is in a metastable, nonequilibrium state (Ergun et al., 2010). When the storage temperature is below its Tg, the chemical and physical kinetics of phase changes are extremely slow, allowing it to be stored for long periods (Hartel et al., 2018). Microbial spoilage during storage is also not a problem, as the aw is usually less than 0.3 (Ergun et al., 2010).

3.4.1. Moisture absorption

The primary cause for the end of shelf life is moisture absorption (Hartel & Nowakowski, 2017). Hard candy has hygroscopic characteristics and readily absorbs moisture from a humid environment. Storage at high temperatures or RH results in moisture absorption, causing crystallisation, softening and stickiness (Kawai et al., 2019). The equilibrium RH of hard candy is around 20-30%, where no moisture absorption or desorption occurs. A higher RH of 30% causes already moisture absorption (Hartel et al., 2018). (Ergun et al., 2010; Roos & Drusch, 2016b)

Moisture absorption of candy includes two stages. First, water molecules adsorb to the surface by hydrogen bond interactions causing stickiness (Hartel & Nowakowski, 2017). Then, the moisture migrates extremely slowly into the centre. If hard candy is exposed to humid air, it can take more than a month for the moisture content to reach equilibrium throughout the whole candy. Nevertheless, the end of shelf life is reached earlier due to changes at the surface, well before equilibrium is reached. (Ergun et al., 2010; Hartel et al., 2018)

The thermodynamic driving force influences the rate of moisture migration. The driving force is the difference between water activities inside the candy (surface-centre), and between the air and the candy. A greater difference between the RH of the air and the aw of candy increases the driving force to absorb water from the air. Therefore, candies with a low aw are susceptible to moisture absorption due to the high driving force. Eventually, the rate of moisture migration decreases during moisture absorption due to the decreasing driving force. This migration continues till an equilibrium between the aws has been reached. (Ergun et al., 2010; Hartel et al., 2018)

In addition, the rate of moisture migration in hard candy is influenced by molecular mobility, which refers to the ability of water to move through the candy. Moisture absorption at the surface causes a decrease in Tg and, subsequently a decrease in surface viscosity. This change in viscosity increases molecular mobility, resulting in an increase in the moisture migration rate. However, it is important to note that the centre of the candy still retains a sugar glass structure, which limits molecular mobility and slows down moisture migration. In the glassy state, molecular mobility is limited, whereas in the rubbery state it is relatively higher. This difference in molecular mobility and driving forces explains

the variation in the rate of moisture migration throughout the candy. (Ergun et al., 2010; Hartel et al., 2018)

3.4.2. Stickiness

The rapid moisture absorption at the surface could have a negative impact on the shelf life due to stickiness. The surface is sticky, and the interior candy is still in a glassy state (Ergun et al., 2010). Candy becomes sticky when they absorb water or the storage temperature rises (Bund & Hartel, 2010). The confectionery is above the Tg value in both cases, causing a decrease in viscosity, which leads to stickiness (Hartel, 2002).

Hard candies with a high concentration of glucose syrup are typically more prone to stickiness, even though hard candy with a high amount of glucose syrup may have a higher Tg (Netramai et al., 2018). The moisture absorption is mainly promoted by the low molecular weight molecules (glucose) due to their hygroscopic behaviour causing them to be more susceptible to stickiness (Hartel et al., 2018).

3.4.3. Crystallisation

In hard candy, sucrose crystallisation is undesirable as it leads to a soft and granular texture (Ergun et al., 2010). Therefore, crystallisation must be prevented. Crystallisation involves two main steps: nucleation (crystal formation) and propagation (crystal growth) (Roos & Drusch, 2016a). Crystals can be formed and continue growing, if the candy is in a supersaturated state with sufficient molecular mobility. The sugars rearrange into a crystalline structure, and undesirable characteristics appear. (Bund & Hartel, 2010)

Several factors can inhibit the crystallisation rate. Reduced molecular mobility caused by the high viscosity generally prevents crystallisation (Šmídová et al., 2003). When hard candy is in a glassy state, molecular mobility is extremely reduced (Bund & Hartel, 2010). The sugar molecules cannot move freely to form a crystalline structure. Moreover, adding glucose syrup (at least 30%) affects the crystallisation rate (Liang et al., 2007). This addition increases the Tg, which reduces molecular mobility, resulting in a lower crystallisation rate. (Netramai et al., 2018)

3.4.3.1. Production

During the production of hard candy, sugar crystals can be integrated into the glassy state due to incorrect candy handling. Therefore, it is crucial to cool rapidly because supersaturation makes the candy mass susceptible to crystallisation. During cooling, the molecular mobility decreases, which reduces the crystallisation rate. Moreover, hard candy produced on the continuous line is more prone to

crystallisation due to the high shear force on the supersaturated candy mass. The high shear force can disrupt the amorphous structure and promote the formation of crystals (Richard & Speck, 2015) Therefore, a higher concentration of glucose syrup is required to inhibit crystallisation during production. (Hartel et al., 2018)

Commercial hard candies show around 1-3% crystallised sucrose in their products (Šmídová et al., 2003). These sucrose crystals are stable. They do not grow if the temperature of the hard candy is kept below the Tg. Although, as the temperature rises above the Tg, the crystals begin to grow due to the increased molecular mobility (Šmídová et al., 2003). This crystallisation process is referred to as ´internal´ graining because crystals can grow without absorbing moisture at the surface. (Bund & Hartel, 2010)

3.4.3.2. Storage

During moisture absorption, sugar crystallisation is a more significant concern than stickiness in high sucrose candies (above approximately 70%). The limited amount of glucose syrup in these candies causes the sucrose to crystallise as moisture is absorbed into the surface. The study by Makower & Dye (1956) showed that storage at low RH (<12%) prevented the graining of pure sucrose glasses for almost two years. At this low RH, the amount of moisture uptake was not increasing the molecular mobility sufficiently to cause crystallisation. In other words, the degree of moisture uptake was insufficient to lower the Tg value of the surface below storage temperature. Storage at elevated RH resulted in a higher crystallisation rate. With an RH of 30%, crystallisation occurred after a couple of days. (Hartel et al., 2018)

Initially, the crystallisation occurs at the candy's surface (Liang et al., 2007). The Tg is lowered at the surface due to moisture absorption (Šmídová et al., 2003). The molecular mobility increases, allowing sugars to rearrange into a crystalline structure, and crystals start to form (nucleation). The high moisture content promotes the formation and growth of new crystals, which start at the surface and gradually spread to the centre. The significant difference in moisture content accelerates moisture diffusion to the centre due to the crystallisation (Hartel et al., 2018). Although, the occurrence of crystallisation in candy can eventually decrease the moisture uptake (Netramai et al., 2018). The water is pushed out of the candy because the Aw at the surface is higher than in the air. (Ergun et al., 2010)

4.   Material and methods

4.4.   Sample preparation

A total of five different hard candy formulations were prepared, a reference and four other hard candies with an altered sugar composition (Table 4). The glucose syrups were provided by Cargill. The formulations were based on a sucrose-glucose syrup ratio of 56:44 (dry basis) with 0.02% colourant (sodium copper chlorophyllin, E141ii). All formulations had an initial dry substance (DS) of 72%. Formulas 1 to 3 contained different glucose syrups, and formula 4 contained a higher concentration of glucose syrup 44 DE, namely a sucrose-glucose syrup ratio of 40:60 (dry basis). The chemical composition of the syrups is given in Table 5.

Table 4: Hard candy formulations

Ingredients (%)	Reference (19 DE)	Formula 1 (9.8 DE)	Formula 2 (13 DE)	Formula 3 (20 DE)	Formula 4 (27 DE)
44 DE glucose syrup	40				54
22 DE[1] glucose syrup		34			
29 DE glucose syrup			44		
High-maltose glucose syrup				40	
Sucrose	40	40	40	40	29
Water	20	26	16	20	17
Colourant (E141ii)	0.02	0.02	0.02	0.02	0.02
[1]Dried glucose					

Table 5: Average chemical composition of glucose syrups

	44 DE glucose syrup	22 DE[1] glucose syrup	29 DE glucose syrup	High-maltose glucose syrup (45 DE*)
DP1 (%)	20	2.0	3.0	3.0
DP2 (%)	17	7.0	12	49
DP3 (%)	13	10	16	20
DP4+ (%)	51	81	70	28

Note: DP= degree of polymerisation
[1]Dried glucose
*DE-value is calculated. See Appendix 1 for complete calculation.

All hard candy formulations were produced in two batches of 3,6 kilograms each. The production process of hard candy was standardised, and the same procedure was applied to each sample. The candy boiler (Bosch, GA0006) used steam to heat the sugar solution.

Initially, the stirrer was turned on, and the sucrose and water were boiled up to 110°C. Subsequently, glucose was added, and the boiling process continued till the candy mass reached a temperature of 130°C. At this stage, the colourant was added. The mass was then further heated to 138°C. Next, the

steam is turned off and the vacuum process starts. The reference (19 DE) was cooked under vacuum till a temperature of 125°C with continuous stirring. Batch 1 of formulas 1 (9.8 DE) and 2 (13 DE) were treated similarly but without stirring. The remaining hard candy formulations underwent a 1-minute vacuum treatment without stirring.

After boiling, the candy mass was cooled on a cooling table. The mass was then shaped into drops with a diameter of 4 millimetres using a drop roller. Lastly, the candy was separated into individual drops and slightly tumbled to curve of the edges.

4.5. Storage experiment

The humid environment was simulated in a desiccator containing a saturated salt solution of sodium chloride. This salt solution provides an RH of 75%. The candy was placed in the humid environment at room temperature. Throughout the 8-day storage experiment, samples were removed from the humid environment, and analyses were performed. Temperature and RH were recorded with data loggers.

4.6. Determination of moisture content

The hard candy's moisture content (MC) was determined before the storage experiment (day 0) using the vacuum oven method. The aluminium pans were oven dried at 100°C for at least 1 hour and cooled down in a desiccator for at least 30 minutes. Next, the candy was ground to powder in an analytical grinder (AKA, A11 basic). For each candy formula, three samples per batch were analysed. The samples contained grams of candy and were dried in a vacuum oven at 70°C for 25 hours. Lastly, the samples were cooled down in a desiccator for at least 30 minutes and weighed. The moisture content was calculated by the formula below.

$$MC = \frac{WW - DW}{WW} \cdot 100\%$$

WW: wet weight   DW: weight after drying

4.7. Determination of colour diffusion

The colour diffusion was analysed on day 0 of the storage experiment. For this measurement, a white filter paper was saturated with a sugar solution containing 60% sucrose and 40% water. The saturated filter paper was placed in a petri dish with a piece of candy on top. This measurement was performed in duplicate, resulting in two samples per batch. The colour diffusion was captured by taking a snapshot

with a digital camera (Nikon DX AF-S NIKKOR 18-55 mm 1:3 5,0-5,6GII ED) every hour for eight consecutive hours. The photos were taken in a dark light box with four fluorescent tubes positioned above the product to provide consistent lighting. The camera settings were set to: maximum aperture (F): f/6,3-7,1, camera sensitivity: ISO 400, shutter speed: 1/25 s, focal length: 28 mm and exposure value (EV): 0 EV. In-between measurements, the petri dish was covered. Finally, the data was analysed using ImageJ. The area of the colour diffusion was measured. The default colour threshold method was used with HSB colour space to mark the area of colour diffusion.

4.8. Determination of moisture uptake

The moisture uptake was determined by the difference in weight before and after the storage experiment. Aluminium pans were oven dried at 100°C for at least 1 hour and cooled down in a desiccator for at least 30 minutes before the storage experiment. Samples of 8 grams were weighed and placed in the desiccators with a saturated salt solution at room temperature. Throughout the multi-day storage experiment, 2 samples per batch were weighed on days 1,3, 6 and 8. The moisture uptake is expressed as a percentage.

4.9. Determination of water activity

The aw was analysed with the aw meter (Aqualab, series 3) on days 0, 1, 3, 6 and 8 of the storage experiment. The measurements were performed on the whole candy and the crushed candy. The candy is crushed into coarse fragments using a mortar and pestle. For each formula, two batches were analysed in duplicate. For this measurement, the equipment protocol of the aw meter was followed.

4.10. Determination of crunchiness

The crunchiness was examined by dropping a 200-gram weight 3 cm above a piece of hard candy. The results of this test were colour ranked. Green indicated crunchiness, the candy was still predominantly in a glassy state. The product cracked visibly and made an audible cracking sound. While red indicated that hard candy was not crunchy, the candy was primarily in a rubbery state. No visual or audible cracking of the candy was observed. Orange indicated that the product was moving towards the rubbery state but still showed some signs of a glassy material. Only a few splinters were observed around the product, with a slight cracking sound. Appendix 2 shows an example of the colour classification of the reference (19 DE). 2 samples per batch were measured on days 0, 1, 3, 6 and 8 of the storage experiment. Each sample was analysed in triplicate.

4.11. Statistics

The data were expressed as mean values ± standard error of the mean (SEM). All graphs were plotted in GraphPad Prism. Possible significant differences were determined by the one-way ANOVA with the alpha-level set to 5%. A Shapiro-Wilk test was performed to confirm the normality of data, and the variance within each formulation was assumed to be equal.

5.   Results

The total chemical composition of the hard candy formulations is shown in Table 6. The reference (19 DE) and formula 4 (27 DE) were high in DP1 (monosaccharides), while formula 1 (9.8 DE) had the lowest number of monosaccharides. The number of disaccharides (DP2) was low in formula 4 (27 DE) due to the lower addition of sucrose. Furthermore, formula 3 (20 DE) had a low concentration of DP4+ compared to the other hard candy formulations.

Table 6: Average chemical composition of the hard candy formulations (sucrose and glucose syrup).

	Reference (19 DE)	Formula 1 (9.8 DE)	Formula 2 (13 DE)	Formula 3 (20 DE)	Formula 4 (27 DE)
DP1 (%)	8.9	0.9	1.3	1.3	12
DP2 (%)	63	59	61	77	50
DP3 (%)	5.6	4.4	7.1	8.9	7.5
DP4+ (%)	22	36	31	12	31
DE[1]	19.4	9.8	12.9	20.4	26.6

Note: DP= Degree of Polymerisation
[1] DE-value of formulations are calculated. See Appendix 3 for complete calculation

The final vacuum temperatures are shown in Table 7. The temperature- and time-controlled vacuum process showed a large variance in the final vacuum temperature between batches for formulas 1 (9.8 DE) and 2 (13 DE). The non-stirred temperature-controlled batches are excluded from further analysis.

Table 7: The final vacuum temperature

	Reference (19 DE)	Formula 1 (9.8 DE)	Formula 2 (13 DE)	Formula 3 (20 DE)	Formula 4 (27 DE)
Batch 1 (°C)	125[1]	125[2]	125[2]	±125[3]	±128[3]
Batch 2 (°C)	125[1]	±130[3]	±115[3]	±128[3]	±128[3]

[1]With stirring – temperature controlled (t=1 minute)
[2]Without stirring – temperature controlled (t=unknown)
[3]Without stirring – time controlled (t=1 minute)

Batch 2 of formula 2 (13 DE) had a lower final vacuum temperature than the reference (19 DE). On the other hand, batch 2 of formula 1 (9.8 DE) showed the highest final vacuum temperature.

5.4.   Initial moisture content

The initial moisture content before the storage experiment can be found in Table 5. The hard candy formulas showed a moisture content between 1-3%. The average moisture content of the reference (19 DE) was significantly lower than the other formulations. Reference (19 DE) had around 1% moisture, and the other formulations had approximately 2-3%. Minor differences were observed between the

batches of all formulations. Although, a major difference was observed between the batches of formulas 1 (9.8 DE) and 2 (13 DE).

Table 5: Initial moisture content (%) of hard candy formulations

	Moisture content (%)				
	Reference (19 DE)	Formula 1 (9.8 DE)	Formula 2 (13 DE)	Formula 3 (20 DE)	Formula 4 (27 DE)
Batch 1	1.15 ± 0.005	0.96 ± 0.022	1.69 ± 0.007	2.44 ± 0.023	2.13 ± 0.018
Batch 2	0.93 ± 0.008	2.35 ± 0.023	2.69 ± 0.057	2.12 ± 0.033	2.39 ± 0.027
Average	1.04 ± 0.049			2.28 ± 0.074	2.26 ± 0.060
The batch values are expressed as mean $\pm$ SEM (n=3)					

5.5. Moisture uptake

Figure 1 illustrates the moisture uptake at RH of 75%. In the first phase, moisture uptake increased rapidly and then more slowly. In the second phase, moisture uptake increased rapidly again. In general, the standard error of the mean was relatively minor in this measurement, causing precise results.
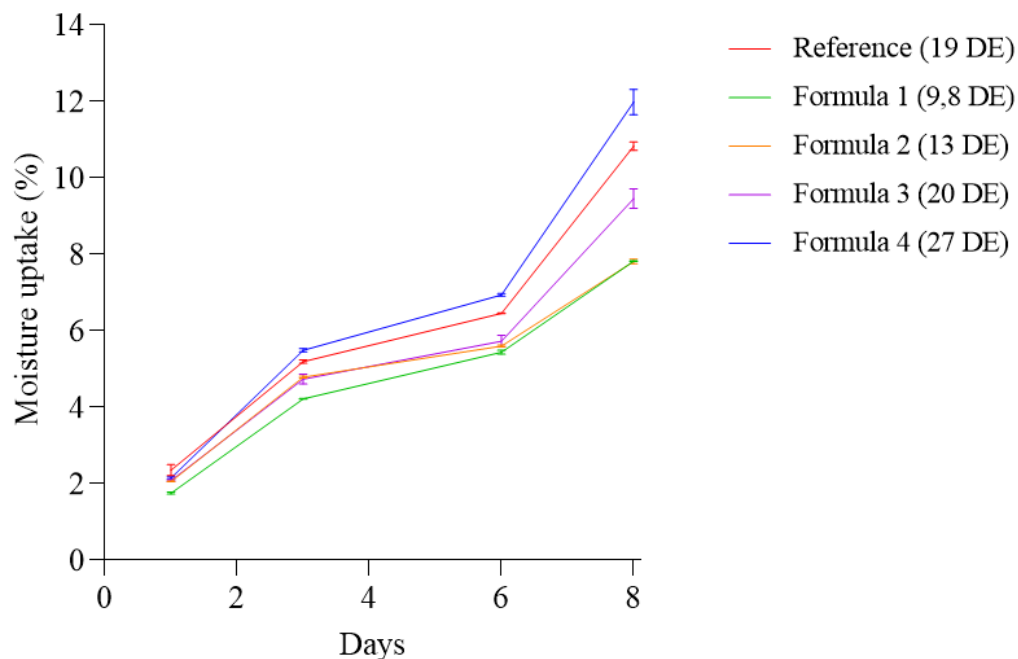

Figure 1: Moisture uptake at 75% RH, values are expressed as mean $\pm$ SEM (n=4). Batch 1 of Formula 1 and 2 are excluded (n=2)

On day 1, there was not a noticeable distinction between the formulations. Only formula 1 (9.8 DE) differed significantly from the reference (19 DE), which continued till the end of the storage experiment. This formula had the lowest moisture uptake on days 1 and 3. As the storage experiment continued, the moisture uptake of formula 1 (9.8 DE) was still considerably low. On day 3, formula 2 (13 DE) showed

no significant difference between the reference (19 DE) and formula 3 (20 DE). However, formula 2 (13 DE) and 3 (20 DE) increased more gradually between days 3 and 6 and converged towards the moisture uptake of formula 1 (9.8 DE). Formula 2 (13 DE) continued to follow the formula 1 (9.8 DE) trend line. However, the moisture uptake accelerated for formula 3 (20 DE) between days 6-8. Nevertheless, formulas 1 (9.8 DE), 2 (13 DE) and 3 (20 DE) remained significantly below the reference on days 6 and 8. On the other hand, the highest moisture uptake was by formula 4 (27 DE), which had significantly higher moisture uptake on days 6 and 8 than the reference (19 DE). Table 6 illustrates the exact values of moisture uptake. The P-values can be found in Appendix 4.

Table 6: Exact values of moisture uptake at 75% RH, values

	Moisture uptake				
	Reference (19 DE)	Formula 1 (9.8 DE)	Formula 2 (13 DE)	Formula 3 (20 DE)	Formula 4 (27 DE)
Day 1	2.36 ± 0.148	$1.75^{1} \pm 0.023$	2.06 ± 0.018	2.09 ± 0.020	2.15 ± 0.025
Day 3	5.20 ± 0.049	$4.23^{1} \pm 0.014$	4.79 ± 0.032	$4.73^{1} \pm 0.128$	$5.49^{1} \pm 0.042$
Day 6	6.46 ± 0.010	$5.44^{1} \pm 0.057$	$5.60^{1} \pm 0.026$	$5.74^{1} \pm 0.152$	$6.94^{1} \pm 0.034$
Day 8	10.82 ± 0.110	$7.83^{1} \pm 0.015$	$7.82^{1} \pm 0.063$	$9.46^{1} \pm 0.258$	$11.98^{1} \pm 0.334$

Values are expressed as mean $\pm$ SEM (n=4).
Batch 1 of formulas 1 (9,8 DE) and 2 (13 DE) are excluded (n=2)
P-value can be found in Appendix 4
[1]Significant difference compared to the reference

5.6.   Moisture sorption isotherm

Figure 2 illustrates the moisture sorption isotherm of the hard candy formulations compared to the reference (19 DE). The aw generally elevated rapidly in the beginning, while the moisture content was still relatively low. The greatest increase in aw was within the first 24 hours. For formulas 1 (9.8 DE), 2 (13 DE) and 3 (20 DE), the aw was doubled within the first 24 hours. Only formula 4 (27 DE) did not double within the first 24 hours.
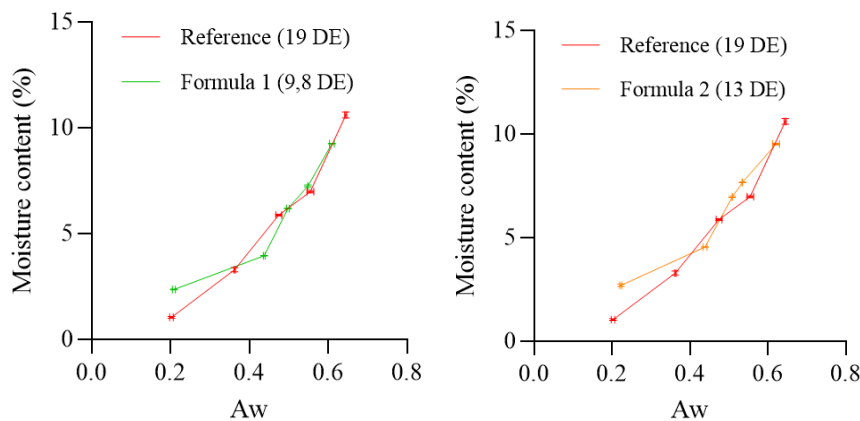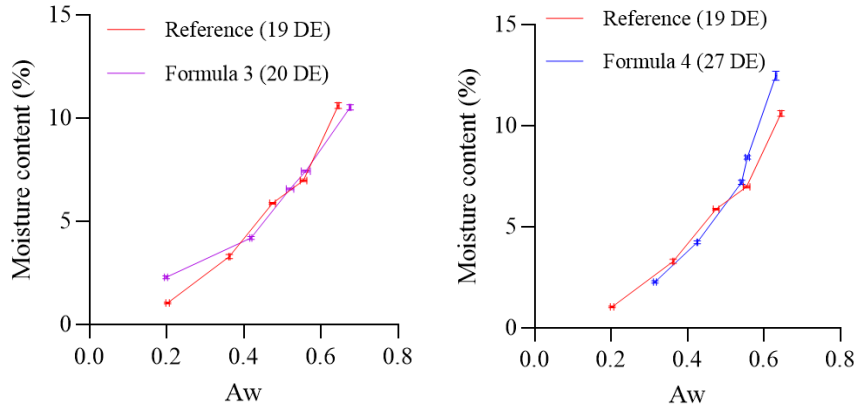
Figure 2: Moisture sorption isotherm of different formulations compared to the reference.

5.7. Water activity

Figure 3 shows the aw during the storage experiment. Before the candy was exposed to the humid environment, a significant difference in initial aw was observed between formula 4 (27 DE) and all the other hard candy formulations. Formula 4 (27 DE) had an initial aw of approximately 0.3, while the other formulations had an aw of around 0.2.

All formulations increased rapidly at first and then more slowly. The formulations show a relatively uniform trend with some exceptions. None of the formulations are in equilibrium with the ambient air of 75% RH. On day 1, the aw of the reference (19 DE) was significantly lower than the other formulations. While on day 3, only formulas 3 (20 DE) and 4 (27 DE) were significantly different from the reference (19 DE). On day 6, no significant difference between the formulations was observed. Formula 3 (20 DE) had a significantly higher aw on the final day than the other formulations.
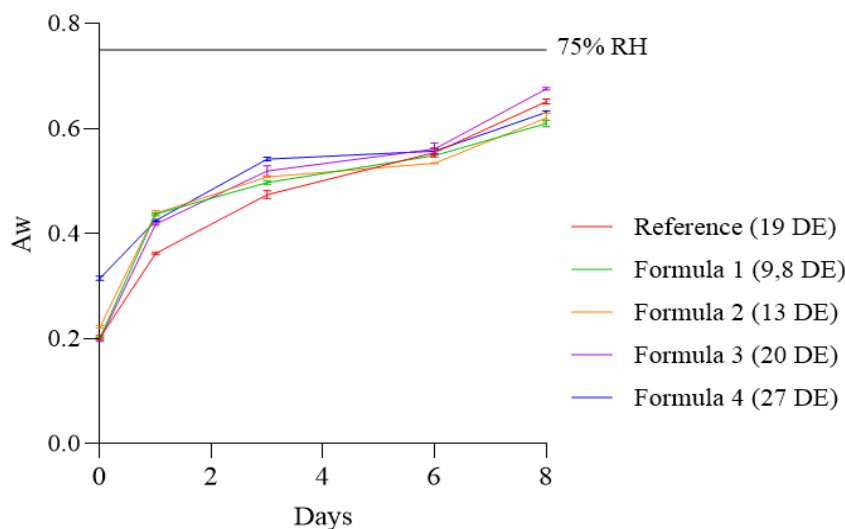

Figure 3: Water activity of all hard candy formulations. The horizontal line shows the water activity of the air during the storage experiment.

Figure 4 shows the aw of the different hard candy formulations, including the surface and average aw. First, the aw at the candy's surface increased more rapidly than the crushed aw. Subsequently, the different aw's were more converging towards each other. An equilibrium between the surface and the average aw emerged in formulas 3 (20 DE) and 4 (27 DE).
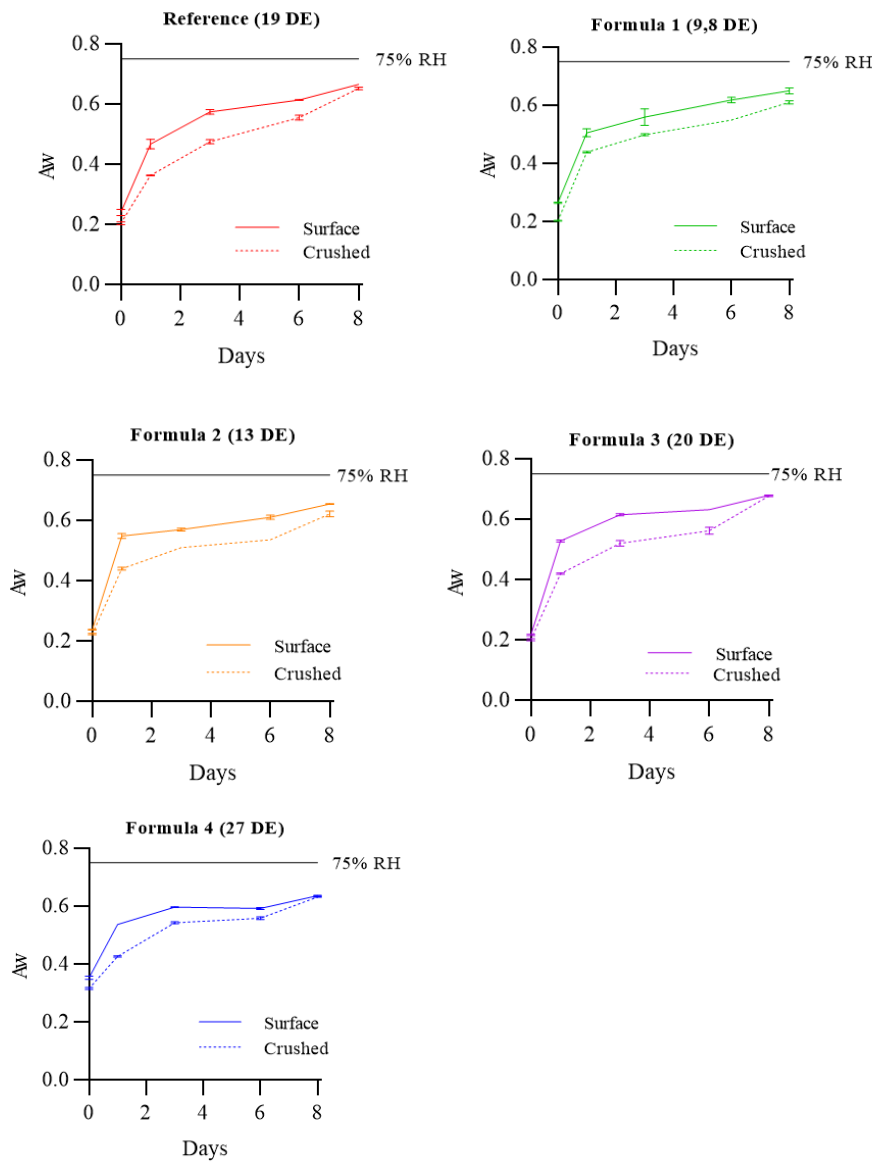

Figure 4: Surface and crushed water activity of different formulations. The horizontal line shows water activity of the air during the storage experiment.

5.8. Crunchiness

Table 7 illustrates the crunchiness when exposed to 75% RH. The reference (19 DE), formulas 3 (20 DE) and 4 (27 DE) were primarily in the rubbery state on day 6, while formulas 1 and 2 (13 DE) were at this stage on day 8. Formulas 1 (9.8 DE) and 2 (13 DE) remained crunchy for a prolonged period.

Table 7: Crunchiness at 75% RH

	Day 1	Day 3	Day 6	Day 8
Reference (19 DE)	✓		X	X
Formula 1 (9.8 DE)	✓	✓	✓	X
Formula 2 (13 DE)	✓	✓		X
Formula 3 (20 DE)	✓		X	X
Formula 4 (27 DE)	✓		X	X

5.9. Colour diffusion

Figure 5 illustrates the colour diffusion of different formulations for five consecutive hours. A representative picture of the colour diffusion experiment is given in Appendix 5, showing the observed colour diffusion of formula 1 (9.8 DE) after 7 hours. A similar trend in formulations 1 till 4 can be observed, a near-linear increase. Although, the reference (19 DE) was approaching a plateau. At hours 4 and 5, the reference was significantly above formula 3 (20 DE), but the reference was significantly below formula 3 (20 DE) after 8 hours.
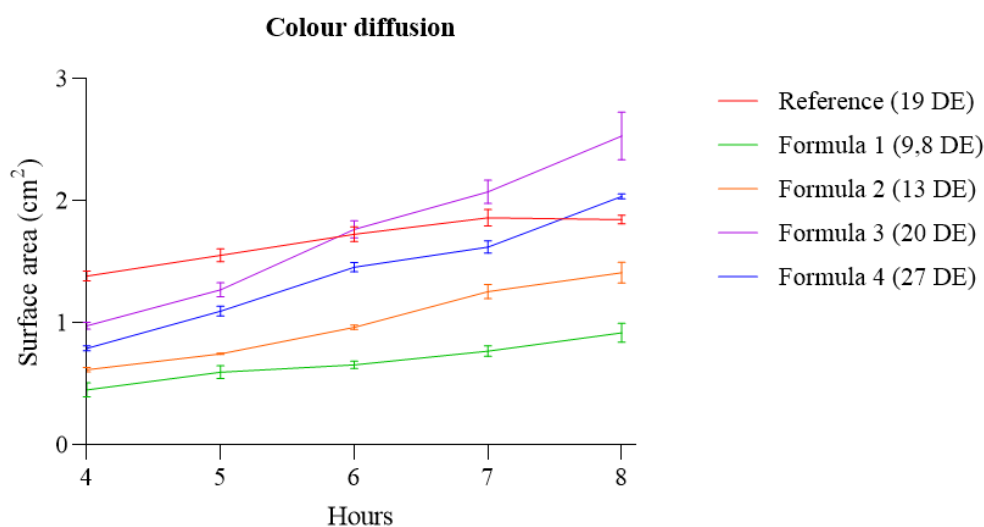

Figure 5: Colour diffusion of different hard candy formulations

After 8 hours, the surface area of formula 3 (20 DE) was significantly larger than the other formulations. In contrast, the surface area of formulas 1 (9.8 DE) and 2 (13 DE) was significantly smaller. No significant difference was observed between the reference (19 DE) and formulas 2 (13 DE) and 4 (27 DE) after 8 hours.

6. Discussion

The outcome of this research has provided insights into the behaviour of different hard candy compositions during prolonged exposure to a humid environment. Nonetheless, results should be interpreted cautiously because of the current study's limitations. This chapter provides a comprehensive analysis and interpretation of the results. The limitations and possible consequences of the method are discussed. At the end of the chapter, some recommendations for the future are given.

Hard candy is hygroscopic, implying that hard candy readily absorbs moisture from the environment, resulting in a change in the texture and appearance of the candy. This thesis aims to investigate the impact of different sugar compositions on the moisture absorption of hard candy when exposed to a humid environment. The results indicate that moisture absorption is significantly reduced by using altered glucose syrups. On the other hand, hard candy formulations with a greater quantity of reference glucose syrup, such as formula 4 (27 DE), indicate a significant increase in moisture uptake. Formula 1 (9.8 DE) showed the most promising outcomes, not only in terms of moisture uptake throughout the multi-day storage experiment. It also showed improved performance in terms of colour diffusion and crunchiness.

6.4. Influence of the vacuum process

Throughout the experiments, the vacuum process has changed. The process was initially temperature-controlled till a temperature of 125°C and was later changed to a time-controlled process of 1 minute without stirring. The high viscosity of the formulations led to the failure of the stirrer during the vacuum process. The vacuum process could no longer be controlled by temperature. All formulations should be time-controlled to standardise the process. Therefore, the stirred vacuum process of the reference (19 DE) was measured in time to establish a comparative baseline. The reference (19 DE) required 1 minute under vacuum, with continuous stirring to reach a temperature of 125°C. The remaining hard candy formulations were treated in an identical timeframe but without stirring. The non-stirred temperature-controlled batches were excluded to avoid misleading conclusions and enhance reliable and reproducible outcomes.

Batch 1 of formulas 1 (9.8 DE) and 2 (13 DE) were still controlled by temperature, causing a lower moisture content. This deviation suggests that the vacuum process may have been prolonged. The highly viscous mass and lack of stirring reduced heat transfer (Fleer et al., 2021; Zhang et al., 2020). The candy mass needs to be vacuumed longer to drop to a temperature of 125°C. In addition, the temperature on the thermometer probably shows an unrepresentative average temperature. The lack of stirring causes temperature variations throughout the candy mass due to reduced heat transfer (Zhang et al., 2022). The

surface is cooled down sufficiently, while the centre is above 125°C. All these factors cause unrepresentative results of the moisture content of formulas 1 (9.8 DE) and 2 (13 DE) compared to the other formulations.

Despite excluding temperature-controlled batches, the standardised time-controlled process without stirring may face similar difficulties. Deviations in the final vacuum temperature are still observed. The time is standardised, but the heat is still unevenly distributed due to the reduced heat transfer causing unreliable final vacuum temperatures (Zhang et al., 2022). The reduced heat transfer could also cool the mass more slowly, resulting in different final vacuum temperatures due to the different viscosities of the formulations (Fleer et al., 2021). The results suggest a relation between the final vacuum temperature and the viscosity of the glucose syrups. The viscosity of formula 1 (9,8 DE) was the highest due to the low DE-value (Castro et al., 2016), which could cause the higher final vacuum temperature.

6.5.   Initial moisture content

The stirred reference (19 DE) shows a significantly lower moisture content. Stirring helps expose more of the candy surface to the vacuum, which will likely increase the evaporation rate resulting in lower moisture content. Therefore, the other non-stirred formulations might show a higher moisture content than the reference (19 DE) due to the reduced surface area. In addition, the moisture content could differ due to the boiling point of the formulations. Hartel et al. (2011) state that different glucose syrups have different boiling point elevations, resulting in different moisture content when using the same boiling process. In this study, the same boiling point is used for all formulations. Thus, the moisture content of formula 4 (27 DE) should be the lowest due to the high amount of lo molecular weight sugars (glucose), resulting in a higher boiling point (Ergun et al., 2010). However, the data in this study does not confirm the theory that a lower moisture content is observed due to the high boiling point.

The moisture content influences the T_g of the candy extensively (Ergun et al., 2010). The reference (19 DE) might have a significantly higher T_g than the other formulations due to the low moisture content— a high T_g results in a more stable product (Nowakowski & Hartel, 2002). Although, the excluded batches of formulas 1 (9,8 DE) and 2 (13 DE) with a low initial moisture content did not show remarkable differences in moisture uptake throughout the storage experiment. The potential impact of this significantly lower moisture content on moisture absorption remains uncertain.

6.6.   Initial aw

The initial aw is relatively similar to each other. However, formula 4 (27 DE) with a higher aw is the outlier. The initial aw of formula 4 (27 DE) does not fit the theory that the most significant aw reduction is achieved using ingredients with low molecular weight and high solubility (Ergun et al., 2010). Formula 4 (27 DE) has the highest number of monosaccharides (low molecular weight molecules) due to the increased concentration of glucose syrup. This high amount of glucose should result in a significant decrease in aw rather than an increase. On the other hand, the solubility in formula 4 (27 DE) may be low due to the high amount of glucose and low amount of sucrose, as sucrose has a higher solubility than glucose. Also, the solubility of sucrose decreases when glucose syrups are added, which could cause an even lower solubility of sucrose for formula 4 (27 DE) due to the high amount of glucose syrup in this formula (Hartel et al., 2011). Although, limited research exists on the impact of various glucose syrups on sucrose solubility. Nevertheless, the total amount of dissolved solids is increasing despite the decreased solubility of sucrose. Therefore, the solubility of the ingredients in the formulations is uncertain and definitive conclusions about initial aw cannot be drawn. (Hartel et al., 2018)

A higher initial aw ensures a lower driving force for moisture migration. The difference between relative humidity and the aw of the candy is smaller for formula 4 (27 DE). The decreased driving force, results in a lower rate of moisture migration (Ergun et al., 2010). Therefore, an initial aw similar to the other formulations (aw=0.2) could result in even higher moisture absorption for formula 4 (27 DE).

6.7.   Moisture uptake trend

The hard candy's moisture uptake trend is relatively similar for all formulations. Also, the moisture sorption isotherm of different formulations is relatively similar. Therefore, no interpretation in relation to the kinetic model is possible, and the main focus is on moisture uptake. The moisture uptake of hard candies is expected to have a logarithmic trend since the driving force is decreasing. The moisture migration rate depends on the driving force and continues till the water activities are in equilibrium (Hartel et al., 2018). However, there is a discrepancy between predicted and observed results, with experimental results showing a rapid increase twice.

The first phase of the moisture uptake graph is in line with the existing literature, which suggests a significant increase and then a slower increase in moisture uptake due to the decreasing driving force. The first steep increase in moisture uptake may be caused by the surface absorbing moisture. This area quickly absorbs moisture from the air. The data obtained provides empirical evidence that supports this theory, as a rapid increase in surface aw can be observed during the first day of the storage experiment

in Figure 4. Additionally, a high driving force is observed, which stimulates moisture uptake (Figure 6) (Ergun et al., 2010). Then, the moisture uptake slows down. The moisture absorption at the surface may be saturated, and moisture migrates slowly into the interior, slowing the overall moisture uptake (Ergun et al., 2010). The driving force also slows down, which inhibits the moisture absorption rate (Figure 6).
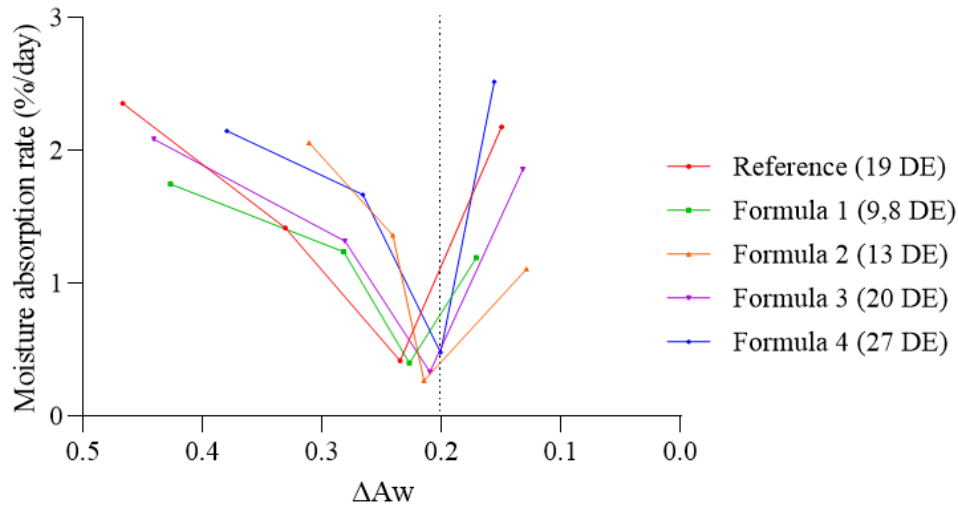

Figure 6: Additional graph to demonstrate the rate of moisture uptake per day and the driving force between the candy and the air. The line indicates the second phase of the moisture uptake. ΔAw= driving force = RH ambient (0,75) - Aw_candy.

In the second phase, the driving force decreases even more, which should contribute to a decreasing moisture uptake rate (Ergun et al., 2010). However, the moisture absorption rate is noticeably increased. The data presents a conflicting perspective compared to the existing literature. Therefore, the diffusion coefficient is calculated using the following equation to understand this data better. The calculated diffusion coefficient indicates how quickly particles diffuse in the candy in relation to water content. The equation is a simplification of Fick's, where rough approximations are made. The geometrical constraints are not taken into account. (Coulson et al., 2001)

$$Qualitative\ diffusion\ coefficient \ \propto \frac{\left(\frac{\Delta U}{\Delta t}\right)^2 \cdot t}{\Delta Aw \cdot W}$$

U   Water uptake (%)
t   Time (days)
Aw   Water activity
W   Water content (%)

Figure 7 shows an exponential increase in the diffusion coefficient between days 6 and 8 of the storage experiment (the last data point on the graph). This result implies that the particles have a higher mobility.

Over time, moisture absorption causes the candy to drop below its Tg. The candy transitions from a glassy to a rubbery state, which increases mobility (Ergun et al., 2010; Šmídová et al., 2003). Therefore, this sudden increase in the second phase of the moisture uptake curve could be due to the phase transition. The higher mobility makes the product more susceptible to moisture absorption (Ergun et al., 2010).

There is a noticeable difference between the formulations. Formulas 1 (9.8 DE) and 2 (13 DE) have a significantly lower diffusion coefficient than the other formulations. The composition could affect the diffusion coefficient. Low molecular weight molecules significantly decrease the Tg of hard candy, which could accelerate the transition to the rubbery state (Spanemberg et al., 2019). The reference (19 DE) and formula 4 (27 DE) both have a high amount of glucose, which could accelerate the transition. However, formula 3 (20 DE) has similar amounts of glucose to formulas 1 (9.8 DE) and 2 (13 DE) but still shows a noticeable increase in the diffusion coefficient. Nevertheless, the Tg could be reduced due to the high amount of DP2 and low amount of DP4+. All three formulations could significantly reduce the Tg due to the composition, which results in an early transition from a glassy to a rubbery state.
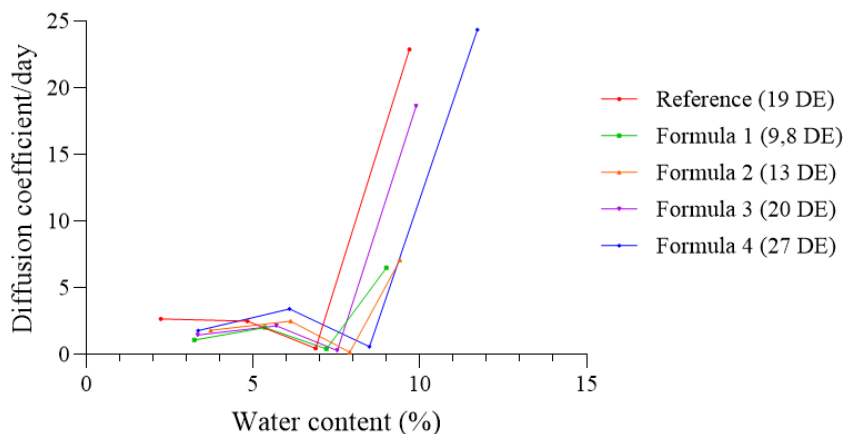

Figure 7: Additional graph to demonstrate the relation between diffusion coefficient and water content.

The diffusion coefficient shows a correlation with the crunchiness test. On day 6, formulas 1 (9.8 DE) and 2 (13 DE) still showed the characteristics of a glassy material. By contrast, the other formulations had already lost their crunchy texture. The diffusion coefficient of formulas 1 (9,8 DE) and 2 (13 DE) is relatively low between days 6-8 because these candies are still partly in the glassy state, as confirmed by the crunchiness test. However, the other formulations were predominately in the rubbery state between days 6-8, causing the high diffusion coefficient.

For the crunchiness test, it should be noted that the duration of the experiments was about eight weeks. The researcher may have subconsciously changed the rating of hard candy during this period. This experimental error should be considered. The classification of red was clear, no glassy material was

observed when dropping the weight which indicates that the candy is primarily in the rubbery state. Nevertheless, ranking green and orange was more challenging due to the different formulations with different behaviour. For instance, the surface became sticky and moved towards the rubbery state but the inside was still predominantly in the glassy state, this sample could be ranked as green or orange. The transition from green to orange was difficult to observe because of the lack of a clear-cut distinction in this classification. Therefore, the focus is on the red-classified areas.

6.8.   Influence of the composition on the formulations

There is a clear distinction between the moisture uptake of the different formulations. The results indicate that the moisture absorption is significantly reduced by using altered glucose syrups with a decreased level of monosaccharides. Formula 1 (9.8 DE) had the lowest moisture uptake throughout the multiday storage experiment. In contrast, formula 4 (27 DE) with more monosaccharides indicates a significant increase in moisture uptake. This study confirms a correlation between molecular weight and moisture uptake. Even though the Tg may be increased in formula 4 (27 DE) due to the high amount of glucose syrup, moisture uptake is mainly stimulated by the hygroscopic low molecular weight sugars (Hartel et al., 2018). Due to the absence of low molecular weight sugars in formula 1 (9.8 DE), the moisture uptake could be mitigated.

Formula 3 (20 DE) shows a decreased moisture uptake compared to the reference, but not as decreased as formulas 1 (9,8 DE) and 2 (13 DE) at the end of the storage experiment. However, in terms of composition, they all contain approximately the same number of monosaccharides. Although, as mentioned in the ´moisture uptake trend´ paragraph, formula 3 (20 DE) also contains the lowest amount of DP4+ and the highest amount of DP2, which reduces the total molecular weight. This difference could result in higher moisture uptake than formulas 1 (9,8 DE) and 2 (13 DE). The moisture uptake is not only decreased by the absence of monosaccharides alone, but in combination with high levels of DP4+. However, drawing conclusions from DP4+ levels alone is not reliable. For instance, formula 3 (20 DE) has the lowest amount of DP4+, which should result in higher moisture uptake than the reference, if only DP4+ is considered. In addition, formula 4 (27 DE) has the same amount of DP4+ as formula 2 (13 DE), although the moisture uptake of formula 4 is significantly higher. The precise value of high molecular weights (DP4+) remains uncertain due to the lack of definitive data. The amount of DP4+ may be low, but the molecular weight may be extremely high. The degree of polymerization in the DP4+ is not precisely defined.

Low levels of monosaccharides combined with high levels of DP4+ not only decrease moisture uptake but also improve stability of crunchiness and colour diffusion. Formula 1 (9.8 DE) shows the most

improved results when looking at these aspects. Formula 2 (13 DE) demonstrates almost comparable efficacy.

The DE value of the reference (19 DE) is comparable with formula 3 (20 DE). However, both formulations show different behaviour in moisture absorption. The reference had a higher moisture absorption due to more hygroscopic monosaccharides in the hard candy. The DE value can be a valuable tool for characterising glucose syrups. However, glucose syrups with the same DE value can still show differences in saccharide composition and subsequent properties. Therefore, the primary focus should be on the saccharide composition for a better understanding of the properties of the glucose syrup.

The moisture content and the composition of the formulations provide insights into the Tg. Low molecular weight molecules and high moisture content decrease the Tg and adversely affect stability (Šmídová et al., 2003). Formula 4 (27 DE) is expected to have the lowest initial Tg due to its high glucose content and increased moisture content compared to the reference (19 DE).

Moreover, crystallisation could affect the results of this study by affecting the texture and moisture absorption (Netramai et al., 2018). The high amount of glucose syrups should inhibit crystallisation, especially in formula 4 (27 DE) (Ergun et al., 2010). However, the high moisture content promotes crystallisation due to the lowered Tg, which increases the molecular mobility (Netramai et al., 2018). Therefore, formulations with high molecular weight are less suspectable for crystallisation, such as formulas 1 (9,8 DE) and 2 (13 DE), due to the high Tg. Crystallisation occurs more rapidly in highly humid environments. Therefore, crystallisation may have occurred in this study, resulting in decreased moisture uptake and consequently affecting texture.

6.9.   Colour diffusion method

The different formulations are expected to occupy the same position in the water uptake graph as in the colour diffusion graph. Because factors that reduce viscosity, such as water content, increase the diffusion rate, according to Ergun et al. (2010). Formulas 1 (9,8 DE) and 2 (13 DE) reflect the same results as the water uptake: a low water uptake and a low colour diffusion. However, the reference (19 DE) and formulas 3 (20 DE) and 4 (27 DE) do not show the same results as the water uptake. The data of formula 4 (27 DE) is contradictory, as the colour diffusion is relatively low. This formula should have a significant colour diffusion due to the high-water uptake.

The reliability of the test is uncertain. This measurement is a self-designed analysis, which could have many errors. For instance, the colour diffusion could be increased by adding more sugar solution to the filter paper. The colour diffusion is the same but leaks out more, resulting in a larger colour diffusion

area. In addition, the photographs were low quality due to environmental circumstances. The reflection was visible in the photographs, and the lighting conditions in the different photographs showed inconsistencies. All those variations could result in an incorrect determination of the colour threshold in ImageJ.

6.10. Limitations

The irregular shape and small size of the hard candy constrained the methodological choices. Specific analyses could not be performed due to the shape and size of the candy, such as stickiness and hardness.

Furthermore, the reliability of the data is impacted by the non-standardised vacuum process and the sample size. The candy process should be standardised throughout the whole process. Although, this was not the case which may have influenced the results. The boiling machine was unsuitable for the processing requirements of the viscous candy mass. The process had to be adapted to the limitations of the equipment. Also, the sample size is limited due to time and material limitations. The excluded batches of formulas 1 (9,8 DE) and 2 (13 DE) further reduce the reliability of the data.

Other material limitations were encountered in the colour diffusion analyses. The materials were limited to obtain high-quality photographs, which could affect the data analysis in ImageJ. The determined colour threshold may be less accurate due to the low-quality photographs.

6.11. Recommendations

Future studies should consider less extreme humid conditions for a prolonged period with a bigger sample size to obtain more reliable results. The differences between extreme and typical humid conditions should be investigated because a study by Castro et al. (2016) showed different hygroscopic behaviour of maltodextrins at different RH´s. The study should be conducted for an extended period due to the low moisture uptake rate. In addition, the candy-producing process should be standardised to a greater extent by using a high-performance machine that can stir highly viscous products.

Additionally, it is recommended to conduct further research to better understand the effects of different hard candy formulations on processability and physical characteristics, such as hardness and sweetness. The small amount of low molecular weight sugars and high amounts of high molecular weight sugars increase the Tg and hardness and decrease sweetness. The increased Tg might decrease processability due to the early transition to the glassy state, which might deal with difficulties in the process. The low sweetness and brittle texture may also be unacceptable to customers.

7. Conclusion

This research aimed to investigate the effect of different sugar compositions on the moisture absorption of hard candy when exposed to a humid environment. The results indicate that altering the sugar composition can reduce moisture absorption in hard candy and consequently improve the stability of colour diffusion and crunchiness. Formula 1 (9,8 DE) shows the most significant reduction in moisture absorption, while formula 4 (27 DE) shows the highest moisture absorption when exposed to a humid environment. A correlation between molecular weight and moisture uptake was observed. The absence of hygroscopic low molecular weight sugars combined with the presence of high molecular weight sugars mitigates moisture absorption, as in formula 1 (9,8 DE). Almost the same efficacy applies to formula 2 (13 DE).

Furthermore, the moisture absorption trend is relatively similar for all formulations. The first phase of moisture uptake follows the expected pattern, with an initial rapid increase followed by a slower increase due to the decreasing driving force. This steep increase could be due to the rapid moisture absorption at the surface. In the second phase, the driving force continues to decrease, which should lead to a decrease in the moisture absorption rate. However, an exponential increase in moisture uptake is observed with a high calculated diffusion coefficient in the candy mass. This increase in moisture absorption could be due to a phase transition from a glassy (low diffusion coefficient) to a rubbery state (high diffusion coefficient), resulting in higher mobility, which increases susceptibility to moisture absorption. Formulas 1 (9,8 DE) and 2 (13 DE) with small amounts of low molecular weight saccharides, such as glucose, show lower calculated diffusion coefficients in the candy mass, indicating a delayed transition to the rubbery state and better retention of crunchiness.

In summary, formula 1 (9.8 DE) showed promising outcomes regarding reduced moisture uptake and improved stabilities of crunchiness and colour diffusion compared to the other formulations due to the limited amount of low molecular weight saccharides combined with high amounts of high molecular weight saccharides. Further research is recommended to understand better the impact of different hard candies concerning processability and physical characteristics such as hardness and sweetness.

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Appendices

Appendix 1: DE-value calculation of glucose syrup

Calculation of DE value for high maltose glucose syrup

Component	Composition (%)	Observed DE	Minimal contribution to DE	Maximum contribution to DE
Monosaccharide	3	100	3	3
Disaccharide	49	58	28,4	28,4
Trisaccharide	20	39,5	7,9	7,9
Tetrasaccharide	0-28	29,8	0	8,3
Pentasaccharide		24,2		
Hexasaccharide		20,8		
Heptasaccharide and higher	0-28	10,2	2,9	0
Total	100		**42,2**	**47,6**

Average DE-value of the glucose syrup = 45 DE

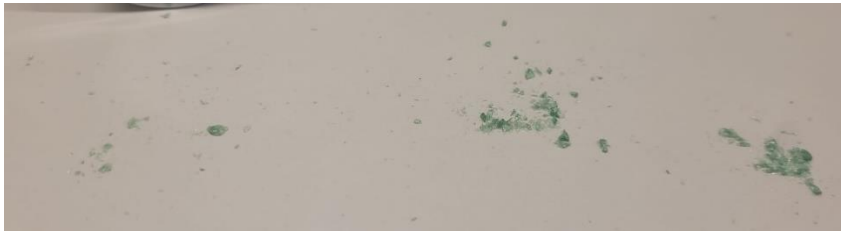Appendix 2: Example of crunchiness classification

Figure 8: Green classification of the reference (19 DE)

Figure 9: Orange classification of the reference (19 DE)

Figure 10: Red classification of the reference (19 DE)

Appendix 3: DE-value calculation of hard candy formulations

Reference hard candy

Component	Composition (%)	Observed DE	Contribution to DE
44 DE glucose syrup	44.4%	44 DE	19.4 DE
Sucrose	56%	0 DE	0 DE
Total	100%		**19.4 DE**

Formula 1 hard candy

Component	Composition (%)	Observed DE	Contribution to DE
22 DE glucose syrup	44.4%	22 DE	9.8 DE
Sucrose	56%	0 DE	0 DE
Total	100%		**9.8 DE**

Formula 2 hard candy

Component	Composition (%)	Observed DE	Contribution to DE
29 DE glucose syrup	44.4%	29 DE	12.9 DE
Sucrose	56%	0 DE	0 DE
Total	100%		**12.9 DE**

Hard candy formula 3

Component	Composition (%)	Observed DE	Contribution to DE
46 DE glucose syrup	44.4%	46 DE	20.4 DE
Sucrose	56%	0 DE	0 DE
Total	100%		**20.4 DE**

Hard candy formula 4

Component	Composition (%)	Observed DE	Contribution to DE
44 DE glucose syrup	60%	44 DE	26.6 DE
Sucrose	40%	0 DE	0 DE
Total	100%		**26.6 DE**

Appendix 4: P-value of moisture uptake

Day 1	P-value	Significant difference
Reference vs Formula 1	0,0076	Yes
Reference vs Formula 2	0,2658	No
Reference vs Formula 3	0,1821	No
Reference vs Formula 4	0,3901	No
Formula 1 vs Formula 2	0,3520	No
Formula 1 vs Formula 3	0,1797	No
Formula 1 vs Formula 4	0,0892	No
Formula 2 vs Formula 3	0,9997	No
Formula 2 vs Formula 4	0,9664	No
Formula 3 vs Formula 4	0,9792	No

Day 3	P-value	Significant difference
Reference vs Formula 1	<0,0001	Yes
Reference vs Formula 2	0,0549	No
Reference vs Formula 3	0,0074	Yes
Reference vs Formula 4	0,1085	No
Formula 1 vs Formula 2	0,0201	Yes
Formula 1 vs Formula 3	0,0155	Yes
Formula 1 vs Formula 4	<0,0001	Yes
Formula 2 vs Formula 3	0,9921	No
Formula 2 vs Formula 4	0,0016	Yes
Formula 3 vs Formula 4	0,0001	Yes

Day 6	P-value	Significant difference
Reference vs Formula 1	0,0002	Yes
Reference vs Formula 2	0,0007	Yes
Reference vs Formula 3	0,0005	Yes
Reference vs Formula 4	0,0118	Yes
Formula 1 vs Formula 2	0,8753	No
Formula 1 vs Formula 3	0,3206	No
Formula 1 vs Formula 4	<0,0001	Yes
Formula 2 vs Formula 3	0,8802	No
Formula 2 vs Formula 4	<0,0001	Yes
Formula 3 vs Formula 4	<0,0001	Yes

| Day 8	P-value	Significant difference
Reference vs Formula 1	<0,0001	Yes
Reference vs Formula 2	<0,0001	Yes
Reference vs Formula 3	0,0097	Yes
Reference vs Formula 4	0,0279	Yes
Formula 1 vs Formula 2	>0,9999	No
Formula 1 vs Formula 3	0,0112	Yes
Formula 1 vs Formula 4	<0,0001	Yes
Formula 2 vs Formula 3	0,0108	Yes
Formula 2 vs Formula 4	<0,0001	Yes
Formula 3 vs Formula 4	<0,0001	Yes

Appendix 5: Representative image of colour diffusion experiment
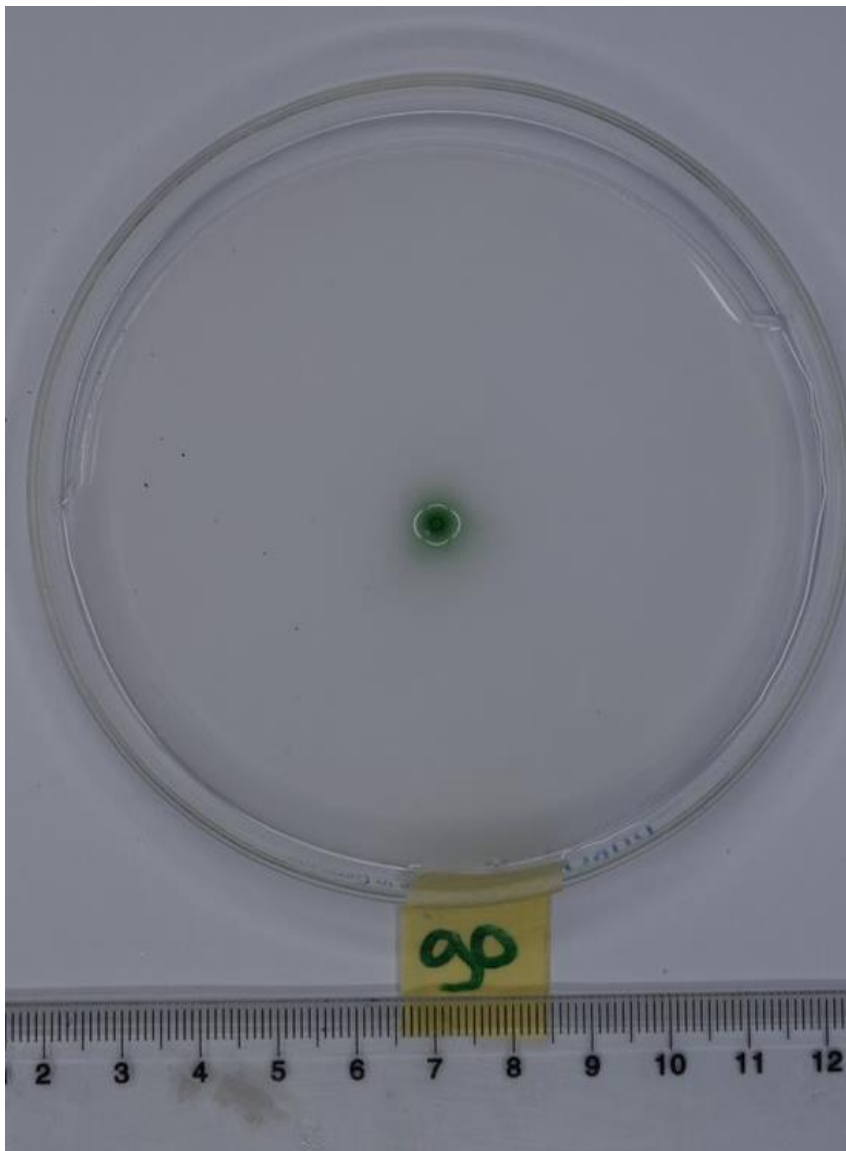
Figure 11: Image of colour diffusion experiment