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Effect of different Soaking media on the Efficiency of Carob Molasses Production

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***Abstract*— S*everal commercial products have been produced from carob pods ranging from food additives to ready-to-eat foods. Examples include carob gums as food thickening agents and carob molasses as traditional sweeteners in the Middle East. During processing, carob is cut into different sizes and soaked in different reagents to yield permeates that undergoes several processing in late production stages. It has been shown that soaking in basic media (Sodium Bicarbonate 1%) yields higher brix values followed by soaking in alcohol (20%), distilled water and acidic media (Citric Acid 1%) respectively. Her we show that powder form yields significantly the highest brix value followed by small, large, mixed and medium, which did not differ significantly from each other. Moreover, no significant difference is noticed between 2 hours, 4 hours or 6 hours of soaking. Sensory analysis shows that base powder was the most favorable reagent-size interaction among all samples.*
Keywords*— *carob; carob pods; carob molasses; brix; permeate; reagent-size interaction.

I. INTRODUCTION

Carob tree, scientifically named "Ceratonia siliqua L.", is cultivated in most Mediterranean countries mainly in mild and dry areas (Papaefstathiou, Agapiou, Giannopoulos, & Kokkinofta, 2018) (Tetik, Karhan, & Oziyci, 2010). It has been cultivated throughout the Mediterranean region for over 4000 years. Carob tree grows to a height of 12-15 meters with a productive life span of more than one hundred years (Karababa & Coşkuner, 2013). World production is estimated at about 160,000 tons per year produced from about 80000 hectares. Yield varies depending on different cultivars and farming practice (Goulas, Stylos, Chatziathanasiadou, Mavromoustakos, & Tzakos, 2016a). Spain is the largest producer of carob followed by Italy, Portugal, Morocco, Turkey, Greece, Cyprus and Lebanon (Papaefstathiou et al., 2018). Lebanon is one of the large consumers of carob products and especially carob molasses because of the Lebanese traditional sweet "carob molasses and tahini". According to FAO statistics (2017), Lebanon witnessed a remarkable decline in both: the production of Carob from around 7000 to 2226 tons/year and the area of harvest from around 700 to 243 hectares, between 1998 and 2017 (HAMADE, 2016).

The fruit is a dark brown pod with a straight, curved or twisted shape (D. Petit & M. Pinilla, 1995). The mass of the pod ranges between 5 and 30 grams with its length and thickness being up to 25 cm and 1.3 cm respectively. Carob bean consists mainly of two parts: the pulp and the seeds (El Batal. H et al., 2016). The fruit consists of: 48%-56% sugar (mainly sucrose, glucose and fructose), 3%-4% protein and 0.2%-0.6% fat content. Carob fruits are rich in dietary fibers and especially in the seeds (Haber, 2002). Pulp is composed of sugars, polyphenols (tannins, flavonoids, phenolic acids) and minerals like potassium, calcium, magnesium, sodium, copper, iron, manganese and zinc. However, seed contains proteins, dietary fibers, polyphenols and minerals (Goulas, Stylos, Chatziathanasiadou, Mavromoustakos, & Tzakos, 2016b). The fruit is rich in vitamins E, D, C, niacin, B6 and folic acid (Papaefstathiou et al., 2018). Carob fruit has many health benefits and is an excellent natural remedy (Azab, 2017). Not only does it act as an anti-diabetic drug, but also it enhances lipid metabolism. Carob lowers total and LDL cholesterol (Wursch, 1979). Moreover, carob relieves gastrointestinal complications and favors lipid oxidation (Rtibi et al., 2015). In addition, it has anti-cancer and anti-diarrheal effects (Goulas et al., 2016a).

Being a natural sweetener, carob can be used as a substitute for cocoa based on the fact that carob does not contain high levels of caffeine and theobromine as cocoa does (Yousif & Alghzawi, 2000). On the market's shelves, carob products are exposed with wide varieties including: powder, creams and molasses. Carob molasses with tahini is the Lebanese sweets signature (Tounsi, Kchaou, Chaker, Bredai, & Kechaou, 2019).

Molasses is produced from sugar rich fruit juices by boiling until achieving 70%-80% soluble dry matter content (Akbulut & Özcan, 2008). Fruit is cut into

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different sizes (small, medium, large and mixed) the mixed equally and soaked for several hours, keeping in mind that no significant difference is seen between brix values of one hour and three hours or between two hours and six hours or between three hours and nine hours (Dimassi, Fawaz, & Rached, 2019). Thus, there is a need to develop new methods to increase the yield of carob molasses via processing modifications without the addition of sucrose or glucose syrup (Dimassi, Rached, Fawaz, & Akiki, 2019).

Based on the fact that carob molasses are done by boiling for few hours after cutting and soaking carob pods in water (Dimassi, Fawaz, et al., 2019). It is interesting to study whether soaking carob pods with different reagents and starting from different sizes of pods can affect the yield of the product or not. Another important point to pinpoint in this study is the sensory evaluation and the product acceptance of the consumers.

Aim is to study the effect of sizes, soaking media and their interaction on the permeate Brix value. Furthermore, sensory evaluation and consumer's acceptance for the acceptance of products resulting from each soaking media and size would be tested using sensory analysis.

II. MATERIAL AND METHOD

2.1. Materials

The materials used in this study are carob pods, which were broken into small, medium and large pieces at the carob production facility at Tier Felsay, South district, Lebanon. Furthermore, the soaking media were distilled water, acidic media using 1% citric acid, basic media using 1% sodium Bicarbonate, 20% alcohol soaking media.

Brix Value Analysis: Brix Value was measured using Portable hand held RFM700 refractometer (Bellingham and Stanley LTD. United Kingdom).

Weight determination: Weight was measured using Portable electronic balance Model 727 was used to measure the weight with an accuracy of ±1 gr (Jata Hogar).

pH analysis: Microcomputer based pH/conductivity/TDS/salinity and temperature pocket meter Model pH/EC80 was used to measure the pH (Jenco VisionP).

Powder Production: Powder was attained by using Moulinex uno 2-in-1 blender of 350 W power and stainless steel blade model LM2211 (SEB group, Ecully, France)

2.2. Methods

2.2.1 Effect of different ground carob sizes and soaking reagents on brix value of permeate

To study the effect of different ground carob sizes, carob pods were cut into four different sizes (small, medium, large and powder). Small, medium and large sizes were attained using mechanical carob pods cutter of 0.1 cm, 1 cm (rounded), and 1 cm (squared) sieves respectively.

To study the effect of different soaking reagents, citric acid), sodium bicarbonate, 95% ethanol alcohol and distilled water were used as acidic, basic, alcoholic and control mediums. Acid and base were added to prepared mediums as 1% by mass while Alcohol is diluted to 20% by volume.

Cups were used to soak ground carob with assigned reagents. For each reagent, tubes are prepared using different sizes. For acid, 3 tubes were put in a group as triplicate samples to be measured at 2 hrs for each size (small, medium, large and mixed). Another 3 tubes were prepared to be measured at 4 hrs and an additional 3 tubes were prepared to be measured at 6 hrs. Procedure is repeated for each reagent (base, alcohol and control).

For small, medium large, and powder sizes, 10 grams of ground carob is weighed and added to each tube with 30 ml distilled water. Mixture is stirred and covered.

For mixed size, 10 ± 1 grams were weighed (3x3x3) from small, medium and large ground carob and added to the tube with 30 ml distilled water. Mixture is stirred and covered.

2.2.1 Effect of different ground carob sizes and soaking reagents on sensory analysis.

Seven samples were prepared to undergo testing. First ample is prepared by the addition of distilled water to commercial carob molasses bought from local producer. Second and third samples were prepared using powder size and mixed size soaked in distilled water. Fourth and fifth samples were prepared from powder and from mixed sizes soaked in acidic media. The sixth and seventh samples were prepared from powder and from mixed sizes soaked in a basic media. Mixtures were soaked and stirred intermittently. Brix value was measured recurrently until a value of 20 °B is attained. In addition to that the commercial carob molasses attained from the carob production facility was diluted till a 20 °B was reached.

The sensory attributes include: color intensity with 0 being the brightest and 5 being the darkest one, aroma with 0 being the unpleasant and 5 being the most pleasant one, bitterness with 0 having no bitterness and 5 having the highest bitterness sweetness with 0 having no sweetness and 5 having highest sweetness, sourness with 0 having no sourness and 5 having the highest sourness, mouth feel with 0 being very thin and 5 being very thick, non-mouth feel with 0 having no grittiness and 5 having the most grittiness. Due to cultural reasons, integrating alcohol in our sensory evaluation was avoided.

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2.3. Statistical analysis

All tests and analysis were run in triplicates and averaged. General linear repeated measure model performed via SPSS (statistical Package for the Social Sciences, version 17.0) was used to study the treatment effect (size as covariate) and size effect (treatment as covariate).

Furthermore, general linear model was used to study the effect of source of carob molasses on sensory evaluation taking soaking media as covariate. The general linear model was also used to study the soaking media effect on the scores of the sensory attributes taking source of carb molasses as covariate.

III. RESULTS

3.1. Effect of soaking time on permeate brix value

Average brix value of different soaking medium at 2, 4, and 6 hrs is presented in figure 1. No significant effect is seen with increased soaking time (Fig. 1). There is a significant difference between acid and base treatment where the brix of permeate from soaking in a basic medium is significantly higher than Brix of permeate resulting from soaking in water and basic medium and tended to be higher, although not significant, than the Brix resulted from soaking in alcohol.

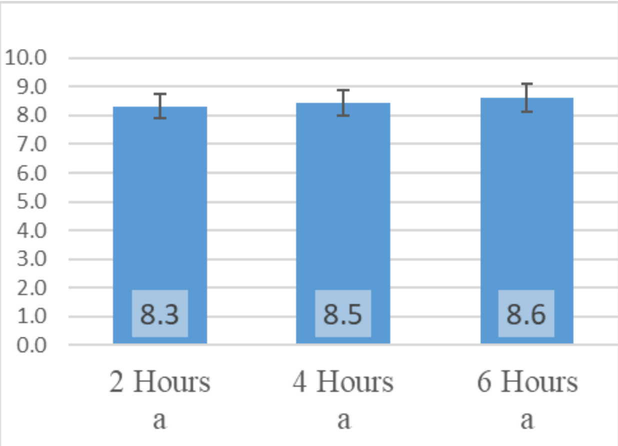

Fig.1 Effect of Soaking Time on Brix of Permeate

The Brix resulting from soaking in alcohol was significantly higher than that resulting from soaking in acidic medium, being the lowest, and tended to be higher than that resulting from soaking in water. The brix resulting from soaking in water did not differ significantly from that of Brix resulting from soaking in acidic medium

3.2. Effect of different forms of ground carob on permeate brix value.

There is no significant difference between brix of permeate resulting from soaking small, medium, large and mixed forms. Powder is the only form that showed the significantly highest brix compared to the Brix resulting from the soaking of the different forms, which did not differ significantly. Brix values from soaking Powder is higher by a factor of 2, 2.96, 2.42 and 2.54 compared to Brix of permeate resulted from soaking small, medium, large and mixed forms respectively. (Fig. 3).

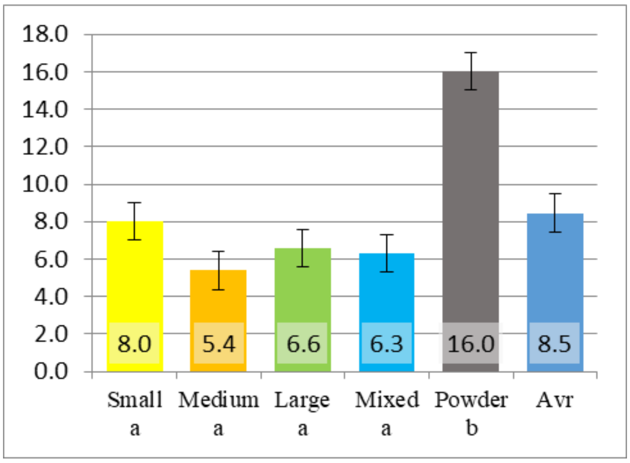

Fig.2 Effect of ground pod form on permeate brix

3.3. Effect of different forms of ground carob on permeate brix value.

The permeate brix value obtained from soaking in base and alcohol medium was significantly the highest followed by the those obtained from soaking in water and acidic media which did not differ significantly (Fig. 2)

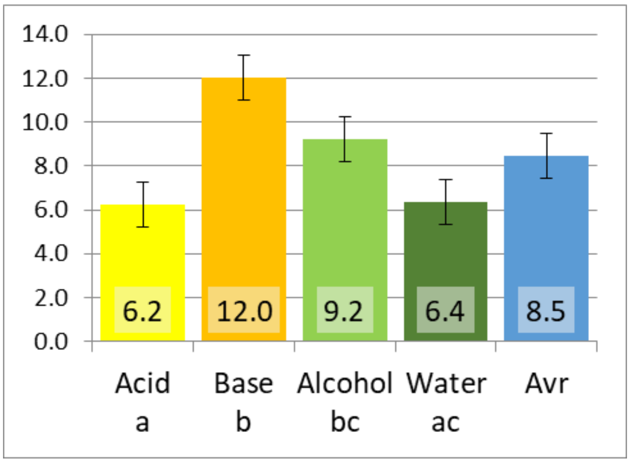

Fig.3 Effect of Soaking Medium on Permeate Brix

3.4. Sensory analysis.

Panelist of n=37 aged between 16 and 55 years old with 37 participants underwent scoring process (0 to 5 scoring). When running sensory evaluation, different parameters were studied.

3.4.1. Effect of source of carob molasses on sensory attributes

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Carob molasses drink produced from soaking powder in acid had the significantly highest score compared to molasses drink produced from acid mixed, base powder, base mixed, water powder and water mixed (Table 1). The first word being the soaking medium te second word being the form of ground carob pods. The carob score of the carob molasses drink resulting from the commercial product scored significantly the lowest and only comparable to the carob molasses drink resulted from water mixed method, which is a simulation of what is done in the carob producing facility (Table 1).

Concerning the color the carob molasses drink resulting from the acid and base powder methods scored significantly the highest. While carob molasses drink from the acid mixed method scored significantly the lowest followed by the carob molasses drink from the commercial and the water mixed source (Table 1).

Aroma followed the a similar pattern where carob molasses drink resulting from the acid powder source possessed the significantly highest value followed by the carob drink resulting from the base powder, base mixed and commercial source and ended by the drinks resulting from the acid mixed, water powder and water mixed sources (Table 1).

Table 1 Score, Color and Aroma of carob molasses from different sources

Carob Drink Source*	Score			Color			Aroma		
	Mean		SE	Mean		SE	Mean		SE
Acid Mixed	1.86	a	0.10	1.16	a	0.19	1.46	a	0.24
Acid Powder	3.19	b	0.10	4.11	b	0.19	4.00	b	0.24
Base Mixed	2.22	c	0.10	3.54	c	0.19	2.49	c	0.24
Base Powder	2.73	d	0.10	4.24	b	0.19	3.24	d	0.24
Commercial	1.57	e	0.10	2.22	d	0.19	2.65	cd	0.24
Water Mixed	1.78	ae	0.10	1.95	d	0.19	0.86	a	0.24
Water Powder	2.81	d	0.10	3.51	c	0.19	1.30	a	0.24

Means with different letters among columns are significantly different

*: First word refers to the soaking medium the second word to the form.

Commercial is diluted carb molasses attained from the carob production facility.

The second sets of tested sensory attributes were bitterness, sourness and sweetness. As for bitterness the carob molasses drink from Acid Mixed and Acid Powder were significantly the highest followed base mixed, water powder, then by the scores of the base powder, water mixed and ended by the score of the drink from the commercial molasses, which is the lowest (Table 2).

The sourness scores of the drinks from the Acid Powder, Acid mixed and water mixed were significantly the highest followed by the water powder which was significantly higher than the carob drinks originating from the base mixed and ended by the drinks resulting from the base powder and commercial sources being the significantly lowest (Table 2).

As for the sweetness the drink obtained from the commercial source scored significantly the highest, followed by the molasses drink from the water powder, base mixed and base powder and ended by the drinks from acid powder and acid mixed being the significantly lowest (Table 2).

Table 2 Bitterness, Sourness and Sweetness of carob molasses from different sources

Carob Drink Source*	Bitterness			Sourness			Sweetness		
	Mean		SE	Mean		SE	Mean		SE
Acid Mixed	2.84	a	0.30	4.70	a	0.19	0.54	a	0.20
Acid Powder	2.03	ab	0.30	4.73	a	0.19	0.81	ab	0.20
Base Mixed	1.68	bc	0.30	2.43	b	0.19	1.65	ce	0.20
Base Powder	1.19	ce	0.30	1.49	c	0.19	1.54	ce	0.20
Commercial	0.68	de	0.30	1.38	c	0.19	3.30	d	0.20
Water Mixed	1.08	ce	0.30	4.46	a	0.19	1.14	be	0.20
Water Powder	1.41	bce	0.30	3.30	d	0.19	1.78	c	0.20

Means with different letters among columns are significantly different

*: First word refers to the soaking medium the second word to the form.

Commercial is diluted carb molasses attained from the carob production facility.

The third sets of sensory attributes were the Mouth-Feel and the non-mouth-feel. The first sensory attribute the Mouth-Feel the molasses drink from water Powder is significantly the highest followed by the drinks originating from base powder, acid powder and base mixed then by the scores of the drink obtained from the water mixed and the acid mixed and ended by the score originating from the commercial molasses being significantly the lowest (Table 3).

As for the non-mouth-feel the molasses drink done from water powder and base powder source were significantly the highest, followed by the drinks obtained from acid powder source and consequently the molasses from the water mixed, base mixed and the acid mixed, ending with the drinks from the commercial source, which is the significantly lowest (Table 3).

Table 3 Mouth Feel and Non-Mouth Feel of carob molasses from different sources

Carob Drink Source*	Mouth-feel		Non-Mouth-Feel	
	Mean	SE	Mean	SE
Acid Mixed	1.11 a	0.18	1.03 a	0.16
Acid Powder	3.05 b	0.18	3.24 b	0.16
Base Mixed	2.57 b	0.18	1.08 a	0.16
Base Powder	3.51 bd	0.18	4.27 c	0.16
Commercial	0.51 c	0.18	0.14 d	0.16
Water Mixed	1.59 a	0.18	1.35 a	0.16
Water Powder	3.73 d	0.18	4.35 c	0.16

Means with different letters among columns are significantly different

*: First word refers to the soaking medium the second word to the form.

Commercial is diluted carb molasses attained from the carob production facility.

3.4.2. Effect of soaking reagents on sensory evaluation

The score, aroma and non-mouth-feel of molasses drinks soaked in acidic and basic media were significantly higher than those drinks done by soaking in water. The color and mouth feel of molasses drinks soaked in acidic medium is significantly higher than those obtained from soaking in water and basic medium. As for the bitterness score of drinks soaked in acidic media were significantly the highest compared to those soaked in water and basic medium which did not differ significantly. As for the sweetness score of drinks soaked in acidic media were significantly the lowest compared to those soaked in water and basic medium which did not differ significantly. Different from each other but they are both significantly different than base. As for the sourness score of a drink coming from soaking in an acidic medium was significantly the highest and that of the drink coming from soaking in basic medium is significantly the lowest.

Table 4 Sensory Attribute of Molasses from Different Soaking Medium

Sensory Attribute	Water		Acidic		Basic	
	Mean	SE	Mean	SE	Mean	SE
Score	1.99 a	0.08	2.58 b	0.09	2.52 b	0.09
Color	2.36 a	0.14	2.79 a	0.16	4.04 b	0.16
bitterness	1.15 a	0.18	2.36 b	0.21	1.36 a	0.21
Sweetness	1.86 a	0.13	0.84 b	0.15	1.76 a	0.15
Sourness	3.38 a	0.12	4.46 b	0.14	1.71 c	0.14
Aroma	1.28 a	0.15	2.98 b	0.18	3.11 b	0.18
Mouth-feel	1.91 a	0.15	2.11 a	0.18	3.07 b	0.18
Non-Mouth Feel	1.80 a	0.18	2.24 ab	0.22	2.78 b	0.22

Attributes with different letters among rows are significantly different

IV. DISCUSSION

As for the soaking time it showed no significant difference in the permeate brix values between 2 hours, 4 hours and 6 hours. This is in accordance with the results found by Dimassi el. 2019 (Dimassi, Fawaz, et al., 2019).

Brix values from soaking Powder is higher by a factor of 2, 2.96, 2.42 and 2.54 compared to Brix of permeate resulted from soaking small, medium, large and mixed forms respectively. Thus making powdered carob to be used in food processing may be useful, although a formation of clumps may pose a major problem. This might be solved by doing two stages mixing. As for the small, medium and large showed no significant difference which is not in accordance with Dimassi et al. 2019 where the medium and the mixed ground carob pods form tended to possess the highest permeate brix. This might be explained by the fact that the sample size in this study was much less than that of the later study and thus could not capture the difference. In addition to that, powdered carob pods were proven to high levels of dietary fibers, brown color and water/oil retention capacity compared to molasses done from soaking ground pods which were characterized by reducing sugars content, dark color and functional properties with high antioxidant activity and emulsifying capacity. Thus, it would be interesting to study if soaking using the powder form would combine the benefits of both (Leila Tounsi, Sirine Karra, Héla Kechaou, & Kechaou, 2017).

Furthermore, soaking in basic and alcohol base medium resulted in significantly highest brix values. As for the basic medium it is done by sodium bicarbonate which is available in the market and is relatively used at low concentration. The only concern is the pH value which would make the product a low acid food thus increasing its constrain by considering it a safe food according to the FDA and thus the end product should have a water activity lower than 0.6. Furthermore, one of the uses of carob molasses is as a sweetener in ice cream (Leila Tounsi et al., 2017). If it is to be used as such a basic medium is beneficial since it will make the molasses pH value nearer to the normal milk value and away from the milk proteins isoelectric point.

As for the sensory attributes the acidic medium and the powder form scored the highest. The alcohol as a soaking medium was not done due to cultural reasons. And all the forms and soaking medium sources possessed higher overall score compared to the drinks obtained from the commercial and the water mixed.

V. CONCLUSION

The results of this study suggested the fixation of soaking time to 2 hours and thus lowering the operation times needed in the carob molasses production. Furthermore, basic and alcoholic as a soaking media will increase the brix of permeate thus would increase the efficiency of carp molasses production. The main problem is with the alcoholic soaking medium, which would affect the consumption of carob molasses in Lebanon and Arab world due to cultural reasons. Furthermore, soaking using the powder form should be seriously considered, but care should be taken as not to have development of clumps.

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REFERENCES

[1] Akbulut, M., & Özcan, M. M. (2008). Some Physical, Chemical, and Rheological Properties of Sweet Sorghum (Sorghum Bicolor (L) Moench) Pekmez (Molasses). *International Journal of Food Properties, 11*(1), 79-91. doi:10.1080/10942910701233389

[2] Azab, A. (2017). *CAROB (Ceratonia siliqua): HEALTH, MEDICINE AND CHEMISTRY* (Vol. 2017).

[3] D. Petit, M., & M. Pinilla, J. (1995). *Production and purification of a sugar syrup from carob pods* (Vol. 28).

[4] Dimassi, O., Fawaz, R., & Rached, M. (2019). Effect of soaking time, interval, temperature and and ground carob size on carob permeate Brix value. *International journal of Science, Environment and Technology, 8*(3), 472 – 481.

[5] Dimassi, O., Rached, M., Fawaz, R., & Akiki, R. (2019). Polarimetry and Spectrophotometry to detect adulteration in commercial carob molasses in Lebanon. *International journal of Science, Environment and Technology, 8*(2), 345-357

[6] El Batal. H, A. Hasib. A, F. Dehbi. F, Zaki. N, Ouatmane. A, & Boulli. A. (2016). Assessment of nutritional composition of Carob pulp (Ceratonia Siliqua L.) collected from various locations in Morocco. *J. Mater. Environ. Sci., 7*(9), 3278-3285.

[7] Goulas, V., Stylos, E., Chatziathanasiadou, M. V., Mavromoustakos, T., & Tzakos, A. G. (2016a). Functional Components of Carob Fruit: Linking the Chemical and Biological Space. *International journal of molecular sciences, 17*(11), 1875. doi:10.3390/ijms17111875

[8] Goulas, V., Stylos, E., Chatziathanasiadou, M. V., Mavromoustakos, T., & Tzakos, A. G. (2016b). Functional Components of Carob Fruit: Linking the Chemical and Biological Space. *17*(11), 1875.

[9] Haber, B. (2002). *Carob fiber benefits and applications* (Vol. 47).

[10] HAMADE, K. (2016). *NON-WOOD FOREST PRODUCT, VALUE CHAINS IN LEBANON*. Retrieved from Beirut, Lebanon:

[11] Karababa, E., & Coşkuner, Y. (2013). Physical properties of carob bean (Ceratonia siliqua L.): An industrial gum yielding crop. *Industrial Crops and Products, 42*, 440-446. doi:https://doi.org/10.1016/j.indcrop.2012.05.006

[12] Leila Tounsi, Sirine Karra, Héla Kechaou, & Kechaou, N. (2017). Processing, physico-chemical and functional properties of carob molasses and powders. *Journal of Food Measurement and Characterization, 11*(3), 1440.

[13] Papaefstathiou, E., Agapiou, A., Giannopoulos, S., & Kokkinofta, R. (2018). Nutritional characterization of carobs and traditional carob products. *Food science & nutrition, 6*(8), 2151-2161. doi:10.1002/fsn3.776

[14] Rtibi, K., Jabri, M. A., Selmi, S., Souli, A., Sebai, H., El-Benna, J., . . . Marzouki, L. (2015). Gastroprotective effect of carob (Ceratonia siliqua L.) against ethanol-induced oxidative stress in rat. *BMC complementary and alternative medicine, 15*, 292-292. doi:10.1186/s12906-015-0819-9

[15] Tetik, N., Karhan, M., & Oziyci, H. R. (2010). Characterization of , and 5-hydroxymethylfurfural concentration in carob pekmez. *GIDA, 35* (6), 417-422

[16] Tounsi, L., Kchaou, H., Chaker, F., Bredai, S., & Kechaou, N. (2019). *Effect of adding carob molasses on physical and nutritional quality parameters of sesame paste* (Vol. 56).

[17] Wursch, P. (1979). Influence of tannin-rich carob pod fiber on the cholesterol metabolism in the rat. *J Nutr, 109*(4), 685-692. doi:10.1093/jn/109.4.685

[18] Yousif, A. K., & Alghzawi, H. M. (2000). Processing and characterization of carob powder. *Food Chemistry, 69*(3), 283-287. doi:https://doi.org/10.1016/S0308-8146(99)00265-4