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Effect of combined acid – thermal treatment of charcoal on clarification efficiency of sugar crystals affined water

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Abstract

The objective of this research was to study the clarification of affined water of sugar crystals. The clarification was carried out by different combined acid-thermal treatments of charcoal. In this work we analyzed the colour, total soluble solids (T.S.S), pH, titratable acidity, total sugars, reduced sugars, ash content, purity coefficient and ash ratio. During subsequent heating of the acid treated charcoal at different levels, the surface as well as interlayer water are dehydrated progressively with high temperature and leads to a collapse of the layer that become less accessible to adsorption of coloring pigments. The clarification of sugar crystals affined water showed that adding acid-thermal charcoal at different levels (5%, 10%, 15%, w/w) led to higher clarified affined water than those to standard clarifying agent (Tonsil). Highly significance ($P \geq 0.05$) was observed for total sugars, sucrose, reduced sugars and ash content in treatments; charcoal amounts, charcoal-acid ratios and heating temperature. Heating of the acid-treated charcoal leads to diminishing total sugars, sucrose, total ash, water soluble and insoluble ash as acid-thermal charcoal increased. On the other hand, both of total sugars, sucrose purity coefficient and ash ratios were increased with increasing charcoal-acid ratio. Meanwhile, total sugars and sucrose decreased with increasing the heating temperature degree of charcoal. The results illustrated that clarification greatly increased affined water purity and ash ratio. The clarified affined water of sugar crystals with Tonsil agent had slightly lower total sugars, sucrose and reduced sugars under the higher rates of acid-thermal charcoal. Also, the clarified affined water with Tonsil agent had slightly higher total ash, water soluble and insoluble ash. The colour, T.S.S, pH, total sugars, sucrose, reduced sugars, ash ratio and purity coefficient increased proportionally with increasing acid ratio to charcoal, with the reduced proportionally for T.A., total ash, water soluble and insoluble ash. With increased temperature degrees of acid –charcoal reduces comparatively of color, T.S.S., PH, total sugars, sucrose, ash ratio and purity coefficient, with relatively increase of T.A., reducing sugar, total ash, water soluble and insoluble ash. Additionally, the clarified affined water with clarifying agent had greatly lower purity coefficient and ash ratio. Our results suggest that affined water of sugar crystals treated with acid-thermal charcoal gave the best clarification process making it more suitable for reusing in raw sugar refining.

Keywords: affination process, affined water, clarification efficiency, sugar crystals.

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1. Introduction

Affined water of sugar crystals is the main syrup in sugar industry (Laksameethansan *et al*., 2012). Affination is the first process the milled raw sugar will go through. During affination milled raw sugar is mixed with hot concentrated syrup to help soften the hard molasses coating on the raw sugar crystals. This molasses coating, which contains many initial impurities, is dissolved into the syrup and then separated from the sugar crystals using a centrifugal machine. The syrup still has recoverable sugars and is typically processed in another area of the refinery to make molasses (Sugar Nutrition Resource Center, 2021). During the affination process, dry molasses film dilutes and separates from the crystals, allowing an average of 75 % of colour and 60 % of other impurities to be eliminated from crystals sugars, and retaining in affined water (Olbrich, 2006). Affined water after coarse affination of raw sugar contains soluble substances such as salt of acids, reducing sugars, sucrose, flavonoids and polyphenolics. The small amount of flavonoids, polyphenolics, reducing sugars and organic acid present in affined water of sugar crystals are contributed to dark brown colour (Thilagavathi and Hemalatha, 2016). These compounds should be removed or reduced in order to produce clear and light affined water of sugar crystals. Therefore, the clarification process is required to reduce particles in affined water before affination process utilization (Tawfeuk and Gomaa, 2017). In refining sugar industry, the clarification commonly uses lime in case of white sugar process and in case of molasses clarification, carbonation, sulfitation or activated carbon addition is followed (El-Geddawy 2013; El-Naggar, 2003; Seguí Gil *et al*., 2015). There are several studies on clarification and decolorization of sugar cane juice, sugar beet juice, date juice and dilute molasses treated with lime, activated carbon, bentonite, cationic homo and copolymers, microwave and ultra-sonication (El Geddawy, 2013; Fewllows *et al*., 2003; Miljana *et al*., 2018; Vu *et al*., 2019; Zia *et al*., 2019). The temperature of the magma may be varied according to the raw sugar impurities ranged from 60ºC to 75ºC (Samy El-Syiada *et al*., 2020). Fellows *et al*. (2003) suggested the treatment cane sugar juice with basic acidic reagents to give a pH of 6.7 – 6.9 adding a flocculating medium. Also, removing the insoluble, then, the product was passed through a bed of high porosity, highly-ionized sulphonated styrene divinely benzene copolymer resin to produce purified cane sugar juice. Miljana *et al*. (2019) uses cross-flow microfiltration coupled with bentonite treatment in sugar beet molasses purification. Up to the present time, little information is available in the literature concerning the clarification and physicochemical properties of affined water of sugar crystals. There are counted studies on clarification and decolorization of affined water of sugar crystals treated with lime, bentonite and activated carbon (Laksameethanasan *et al*., 2012). This accounts for the interest in studying the physicochemical properties of affined water of sugar crystals, as well as a need to clarify affined water using different acid-thermal charcoal.

2. Materials and methods

2.1 Materials

Affined water of sugar crystals was obtained from Hawamdia sugar factory during the 2019 operation season. Chemicals were obtained from Sigma Chemical Co. (St. Louis, Mo, and U. S. A).

2.2 Methods

The clarification was carried out by different combined acid-thermal treatments of grape wood charcoal. Primary acid activation was carried out on 30 g of the finely powdered raw charcoal added to 30 ml (1: 1), 60 ml (1: 2) or 90 ml (1: 3 w/v) of 0.5 N HCl. The charcoal-acid suspension was kept on a water bath at 65 – 70ºC with occasional stirring. This was followed by filtration, through washing, and drying till constant weight in a drying oven at 105ºC. Portions of each product for treatment were subjected to secondary thermal treatment for 3 hr at 110, 115 and 120ºC. The weight loss associated with each of these treatments was determined.

2.2.1 Clarification of sugar crystals affined water

These experiments were carried out on affined water of sugar crystals. 500 gm of affined water was made to one liter with distilled water. Affined water was clarified by acid-thermal charcoal concentrations of 5, 10 and 15% (w/w). In every test 5% of the charcoal was mixed with the formerly heated a diluted affined water of sugar crystals (at ca. 70ºC) and the temperature degree was raised to 90ºC and kept for 30 minutes, with stirring. The hot clarified affined water was subsequently centrifuged and filtered. The same technique was repeated whereas the percent of charcoal was varied to 10 and 15%, in addition to the above mentioned 5%. A standard German clarifying agent, Tonsil was used for the sake of comparison.

2.3 Analytical methods

Specific gravity (Sp. Gr) was determined according to AOAC (2005) at 20ºC and calculated as follow:

$$\text{Sp. Gr at } 20^{o}\text{C} = \frac{Weight\ of\ 100\ ml\ of\ sample}{Weight\ of\ 100\ ml\ of\ distilled\ water}\ g/cm^3$$

The pH was performed using a pH meter (Beckman pH meter) (Seguí Gil *et al*., 2015). A total soluble solid was measured by a hand refractometer. It was determined at 20ºC as described by Plews (1970). Colour was measured calorimetrically using a “Beckman – colorimeter” with a red filter and expressed as a % of transmission (T %) according to Plews (1970). Ash content, water soluble and insoluble ash was measured by the methods described in AOAC (2005). Total sugars content of samples was determined by the analytical method that carried out as described in EOS (1990) No. 358. Sucrose content of samples was determined by the Lane and Eynone volumetric method as described by AOAC (2005). Acid inversion was carried out as following; heating the treatment sample to 65°C, HCl (1.1029 specific gravity, 24.85° Brix) was added, and the solution was kept for thirty minutes.

$$\text{Sucrose \%} = \frac{\text{\% reducing sugars after inversion}}{\text{\% reducing sugars before inversion}} \times 0.95$$

Reducing sugars content of samples were determined by the Lane and Eynone volumetric method as described by AOAC (2005). Titratable acidity was measured by titration with 0.1 N NaOH to raise the pH of diluted affined water of sugar crystals to 8.3, using a pH meter to check the endpoint. Titratable acidity was calculated in terms of the number of milliliters 0.1 N. NaOH required to neutralize 100 gram of sample according to the method described by (Mathur, 1981). Purity coefficient was calculated by the following equation according to (Taha *et al*., 1994):

$$\text{Purity coefficient} = \frac{\text{Sucrose \%}}{\text{T.S.S}} \times 100$$

Where: T. S. S = total soluble solids.

Ash ratio was calculated by the following equation according to (Mathur, 1981):

$$\text{Ash ratio} = \frac{\text{Sucrose \%}}{\text{Ash \%}}$$

2.4 Statistical analysis

Data from physical and chemical analysis were analyzed statistically using analysis of t-test (L.S.D at 5%) as described by Snedecor and Cochran (1980). All determination reported were run in three replicates and the results was expressed as a mean of the three runs. A correlation and /or regression tests were applied to study the relationship between acid-thermal charcoal levels, heating temperature degrees of acid-charcoal, physical, and sugar properties, total ash, water soluble and insoluble ash, purity coefficient and ash ratio of clarified affined water of sugar crystals. Simple and multiple regression analysis were carried out using Excel program (Windows, 2010).

3. Results and Discussion

The clarification of affined water of sugar crystals was found to be quite difficult and tedious because of its high viscosity, high solid content, and deep turbid color. Thus, clarification was done on water diluted affined water of sugar crystals at the rate of 500 gm per liter. The increase in the amount of charcoal used in dilution resulted in better decolorization. Heating of the acid-treated charcoal leads to clarification efficiency. The differences obtained when using acid-thermal treated charcoal was high enough to justify the use of the acid-thermal treated charcoal in clarification processes.

3.1 Weight loss percent of grape wood charcoal during the combined acid-thermal treatments

It is evident from Table (1) that acid treatment at 70°C followed by drying at

105ºC leads to an increasing weight loss as function of an increasing acid: charcoal ratio. Subsequent, heating at 105-120ºC increases (P ≥ 0.5) the weight lost. The maximum weight loss through acid treatment is 0.16% and that attained at 120ºC is 25%. During subsequent heating of the acid- treated charcoal the surface as well as interlayer water is dehydrated progressively with temperature and leads to a collapse of the layers that become less and less accessible to adsorption of the sugar solution coloring pigments (Marta *et al.*, 2016).

Table (4): Weight loss percent of grape wood charcoal during the combined acid-thermal treatments.

Thermal treatments	Charcoal: 0.5 N HCl w/v			LSD 5%
	1: 1	1: 2	1: 3	
70	0.20	0.30	0.5	0.09
105	0.50	0.70	1.00	0.10
110	1.00	2.00	5.00	1.10
115	3.00	5.00	10.00	1.75
120	6.00	10.00	15.00	1.70

3.2 Changes in sugar crystals affined water quality properties

Table (2) indicated that acid treatment followed by drying at 105ºC leads to an increase in clarification efficiency as function of increased amounts of charcoal to the sugar crystals affined water and acid, to show a maximum at a ratio of 15% (charcoal) and 1: 3 (charcoal: 0.5 N HCl w/v). Heating of the acid-treated charcoal leads, in general, to the same clarification efficiency irrespective of the initial charcoal and acid amounts (from 5% up 15 % for charcoal and from 1: 1 ratio up 1: 3 for charcoal-acid ratio); the lower ratio (5% charcoal and 1: 1 charcoal-acid ratio) usually furnished charcoal of slightly higher. On the other side the higher ratio (15% charcoal and 1: 3 charcoal-acid ratios) usually leads charcoal of higher activity. However, heating of acid-treatment charcoal exhibit a linear decrease in clarification efficiency with increasing of temperature degrees, comparison with a standard clarifying agent (Tonsil). The clarified affined water with Tonsil had slightly lower brix. The reactivity of acid-treated charcoal as clarifying agent invariably decreases continuously subsequent to thermal treatment at and beyond 110ºC. The 115ºC as well as the 120ºC, charcoal show tendencies to restricting values of adsorption that indicate saturation of the adsorbing surface. Tonsil exhibits the lowest adsorbing capacity especially, with comparison to charcoal at the higher ratio. Consequently, ratios of 15% of charcoal and 1: 3 charcoal-acid ratios with a temperature of 110o C was significantly (P ≥ 0.5) higher clarification efficiency compared with the same ratio, but with temperature degrees of 115 or 120ºC. the minimum clarification efficiency was for a ratio of 15% charcoal and 1: 3 charcoal-acid ratio with a temperature degree of 115ºC. As shown in Table (2), mean value of actual calculated amounts of refractometer readings were significantly lower ($P \geq 0.5$) when compared with corresponding value of standard clarifying agent (Tonsil). This result agrees with those of Miljana *et al*. (2019).

Table (2): Clarification of sugar crystals affined water by grape wood charcoal during the combined acid-thermal treatments.

Thermal treatments		Tonsil at level 15%	Charcoal: 0.5 N HCl w/v									LSD 5%
			1: 1			1: 2			1: 3			
			5	10	15	5	10	15	5	10	15	
	Volume cm^3	750	750	760	775	770	785	795	710	720	740	10.2
110	Ref. reading	15.2	15.2	14.21	13.41	15.32	14.52	13.83	15.77	15	14.05	1.10
	Actual ref.	57	57	54	52	59	57	55	56	54	52	1.5
	Volume cm^3	790	760	772	790	765	790	810	730	750	755	12.4
115	Ref. reading	12.15	13.42	12.69	12.15	13.59	12.65	12.09	13.97	13.06	12.45	0.85
	Actual ref.	48	51	49	48	52	50	47	51	49	47	1.3
	Volume cm^3	779	755	763	779	774	780	790	715	720	730	9.5
120	Ref. reading	12.06	12.7	12.05	12.06	11.88	11.02	10.37	6.32	11.66	10.68	0.73
	Actual ref.	47	48	46	43	46	40	38	45	41	38	1.95

3.3 Effect of clarification of sugar crystals affined water by grape wood charcoal during acid-thermal treatments on physical properties

The comparative results of sugar crystals affined water treated with standard clarifying agent (Tonsil) and acid- thermal charcoal at different levels were presented in Table (3). Higher amounts of acid-thermal charcoal resulted in affined water of sugar crystals with higher transmittance at 420 nm indicating more clarity and lighter color. The most significant colorants formed during sugar processing are melanoidin and melanin (Laksameethansan *et al*., 2012). The ability of acid-thermal charcoal to absorb organic substances is raised with increasing surface areas (Tawfeuk and Gomaa, 2017). Heating of the acid treated-charcoal leads to diminishing the percentage of T.S.S and specific gravity as function of increased amounts of charcoal to the affined water and acid used due to the adsorption phenomenon. The lower ratio (5% charcoal and 1: 1 charcoal-acid ratio) and 110ºC heating temperature usually furnished charcoal of slightly higher activity, while the higher ratio (15% charcoal, 1: 3 charcoal: acid ratio and 120ºC heating temperature) optimized charcoal of higher activity. Meanwhile, no significant ($P \leq 0.05$) variation was observed in pH values and titratable acidity by acid-treated charcoal amount, heating temperature and acid ratios when compared to the standard clarifying agent (Tonsil). The results obtained were nearly similar with those obtained by (El-Naggar, 2002; Samy El-Syiada, 2020; Seguí Gil *et al*., 2015;

Thilagavathi and Hemalatha, 2016). Concerning the influence of heating of the acid-treated charcoal on physical properties, it could be noticed that 15% charcoal and 1: 2 charcoal: acid ratio showed considerably increased colour efficiency. Thus, it seems possible to improve physical properties. Results illustrate the correlation between acid-thermal charcoal level and physical properties of sugar crystals diluted affined water. There were strong correlations (r=0.968, -0.907, and -0.637) between the acid-thermal charcoal levels and colour, T.S.S. and pH values, respectively at 115ºC of acid-charcoal. The linear regression equations for prediction the physical properties of clarification process were:

Y(color) = 2.43 - 0.1167 X + 0.074 X^2 (acid-thermal charcoal level) r = 0.968

Y(T.S.S)= 54 - 0.6 X + 0.0133 X^2 (acid-thermal charcoal level) r = - 0.907

Y(PH) = 5.53 + 0.0113 X- 0.0012 X^2 (acid-thermal charcoal level) r = -0.637

Y(T.A) = 0.49 - 0.002 X + 0.000026 X^2 (acid-thermal charcoal level) r = 0.234

Table (3): Effect of clarification of sugar crystals affined water (500 gm/L) by grape wood charcoal during the combined acid-thermal treatments on physical properties.

Thermal treatments	Physical properties	Affined water of sugar crystals	Charcoal: 0.5 N HCl w/v																		LSD 5%
			1: 1 w/v						1: 2 w/v						1: 3 w/v						
			5		10		15		5		10		15		5		10		15		
			Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	
110	Color	2.5	3.8	3.5	8.9	8.5	15.5	18.5	5.1	4.5	10.3	10.4	18.5	20.3	4.2	4.0	9.1	9.0	18.4	19	0.65
	T. S. S	60.0	56	57	55	54	50	52	60	59	56	57	53	56	57	56	54	54	50	52	0.91
	Sp. Gr	1.4255	1.4050	1.4045	1.4050	1.4040	1.4040	1.4030	1.404	1.405	1.4035	1.4045	1.4030	1.4040	1.403	1.4030	1.402	1.4020	1.403	1.4010	0.001
	pH	5.5	5.54	5.55	5.55	5.54	5.50	5.50	5.60	5.58	5.58	5.56	5.56	5.55	5.55	5.3	5.50	5.56	5.45	5.54	0.02
	T. A	0.60	0.56	0.52	0.53	0.54	0.50	0.55	0.49	0.45	0.48	0.47	0.46	0.49	0.54	0.55	0.56	0.55	0.65	0.55	0.01
115	Color	2.5	3.8	3.3	8.9	8.0	15.5	15.0	5.1	4.1	10.30	10.0	18.5	19.0	12	3.5	9.1	8.0	18.4	18.0	0.70
	T. S. S	60.0	56	51	55	49	50	48	60	52	56.0	50	53	49	57	51	54	49	50	47	0.83
	Sp. Gr	1.4255	1.4050	1.4050	1.4050	1.4045	1.4040	1.4040	1.4040	1.4055	1.4035	1.4051	1.403	1.4041	1.4030	1.4040	1.402	1.4038	1.403	1.4030	0.001
	pH	5.5	5.54	5.54	5.55	5.50	5.50	5.30	5.60	5.61	5.58	5.57	5.56	5.50	5.55	5.53	5.5	5.51	5.45	5.50	0.02
	T. A	0.60	0.56	0.55	0.53	0.56	0.50	0.58	0.49	0.41	0.48	0.42	0.46	0.45	0.54	0.50	0.56	0.51	0.65	0.53	0.01
120	Color	2.5	3.8	3	8.9	6	15.5	12	5.1	3.2	10.30	7	18.5	15	42	3.0	9.1	6.5	18.4	13	0.60
	T. S. S	60.0	56	48	55	45	50	47	60	46	56	43	53	41	57	45	54	42	50	39	0.80
	Sp. Gr	1.4255	1.4050	1.4045	1.4050	1.4040	1.4040	1.435	1.404	1.404	1.4035	1.4030	1.403	1.4025	1.4030	1.4040	1.402	1.4035	1.403	1.4030	0.001
	pH	5.5	5.54	5.62	5.55	5.60	5.5	5.59	5.60	5.6	5.58	5.58	5.56	5.56	5.55	5.65	5.50	5.61	5.45	5.58	0.01
	T. A	0.60	0.56	0.52	0.53	0.53	0.50	0.55	0.49	0.50	0.48	0.51	0.46	0.53	0.54	0.55	0.56	0.56	0.65	0. 58	0.02

The transmittance light percent (colour), T.S.S. and PH values increased proportionally with increasing acid ratio to charcoal, besides decreasing the titratable acidity. With continued addition of acid charcoal, the T.S.S. and pH values soften steadily, while the transmittance light percent and titratable acidity increase steadily. With increased temperature degrees of acid-treated charcoal reduce comparatively of transmittance light percent, T.S.S. and PH values with relatively increase of titratable acidity. The correlation coefficient between heating temperature degrees and colour was (r= -0.993), T.S.S. was (r= -0.997), pH was (r= 0.500), and titratable acidity was (r =0.444), and linear regression equation for prediction the physical properties were:

Y(color) = -17.4 + 0.41 X - 0.002 X^2 (heating temperature)

Y(T.S.S) = -113 + 3.7 X - 0.02 X^2 (heating temperature)

Y (PH) = 44.61 - 0.685 X + 0.003 X^2 (heating temperature)

Y(T.A.) = 36.99 - 0.64 X + 0.0028 X^2 (heating temperature)

The clarified affined water with amount of 10% of Tonsil had slightly darker color with acid-thermal charcoal, had slightly higher total soluble solids and slightly lowers specific gravity. On the other hand, there was no significant ($P \leq 0.05$) difference in both of pH and titratable acidity of affined water with different levels of acid-thermal charcoal. Charcoal was used in sugar cane manufacture to remove colour of juice molecules on charcoal reduced raw juice colour (Laksameethanasan *et al*., 2012). The reduction in colour should be observed for increasing levels of acid-thermal charcoal due to the greater adsorption surface.

3.4 Effect of clarification of sugar crystals affined water by grape wood charcoal during acid-thermal treatments on chemical properties

Clarification of sugar crystals affined water by grape wood charcoal during acid-thermal treatment on chemical properties is presented in Table (4). The changes in chemical properties were assessed for total sugars, sucrose, reducing sugars and ash content. Highly significance ($P \geq 0.05$) was noted for total sugars, sucrose and reducing sugars in the treatments, charcoal amounts, charcoal-acid ratios and temperature degrees. Heating of the acid-treated charcoal leads to diminishing the total sugars and sucrose content as charcoal amount increased. On the other side, both of total sugars and sucrose were increased with increasing charcoal-acid ratio. Meanwhile, total sugars and sucrose decreased with increasing the heating temperature of charcoal. Activated carbon removed large part of the polymeric caramels, alkaline degradation products and melanoidins. It is due to the formation of a mild links between the colorant amino groups, and carbonyl groups (Lakameethanasan *et al*., 2012). The aforementioned finding of the total sugars and sucrose are in line with those obtained by Marta *et al*. (2016). The lower ratio (5 % charcoal and 1: 1 charcoal-acid ratio) and 120°C usually optimized charcoal of total sugars and sucrose, while the higher ratio (15% charcoal and 1: 3 charcoal: acid ratio) and 120°C usually minimized charcoal of total sugars and sucrose. Conversely, heating of the charcoal-acid ratio increased. On the other hand, heating of the acid-treated charcoal leads to increasing the reduced sugars as acid-treated charcoal leads to decreasing the reduced sugars as charcoal amount increased. These results are too close to (El-Geddawy 2013). Thilagavathi and Hemalatha (2016) noticed that the total sugars and sucrose of affined water of sugar crystals increased with increasing charcoal-acid ratio. On the other side, Marta *et al*. (2016) noticed that reducing sugars decreased with increasing charcoal amount. Similarly to these results, El-Naggar (2002) and Laksameethanasan *et al*. (2012) studied the total sugars and sucrose of cane sugar molasses treated by activated charcoal, gradually decreased with increasing activated charcoal with increasing activated charcoal amount. Results of affined water of sugar crystals quality after addition acid-thermal charcoal and / or standard clarifying agent (Tonsil) were studied. The clarified affined water with amount of 10% of Tonsil had slightly lower total sugars, sucrose and reduced sugars especially, under the higher rates of acid-thermal charcoal. It is worthy to mention that a linear decrease with a significantly correlation between acid-thermal charcoal levels and sugar components. The correlation coefficient between acid-thermal charcoal levels and sugar components were (r= -0.759 for total sugar), (r= -0.748 for sucrose), and (r= -0.711 for reducing sugars) at 110^{o}C of acid-treated charcoal and linear regression equations for prediction the total sugar was:

Y(Total sugar) = 55.67 - 0.433 X + 0.0048 X^2 (acid-thermal charcoal level)

Y(red. sugars) = 20.697 - 0.173 X + 0.000134 X^2 (acid-thermal charcoal level)

Total sugars, sucrose and reducing increased proportionally with increasing acid ratio to charcoal. With raised temperature degrees of acid treated charcoal reduces comparatively of total sugars, sucrose and reducing sugars. The correlation coefficient between heating temperature degrees and total sugars was (r= -0.823), sucrose was (r= -0.803) and reducing sugars was (r= 0.787), and the linear regression equation for prediction the sugars components were:

Y(total sugar) = -864.83 +16.258 X - 0.072 X^2 (heating temperature degree)

Y(sucrose) = -2285.78 + 41.06 X - 0.1816 X^2 (heating temperature degree)

Y(reducing sugars) =1423.7 - 24.849 X + 0.1098 X^2 (heating temperature degree)

Table (4): Effect of clarification of sugar crystals affined water (500 gm/L) by grape wood charcoal during combined acid-thermal treatments on sugar components.

Thermal treatments	Sugar components	Affined water of sugar crystals	Charcoal: 0.5 N HCl w/v																		LSD 5%
			1: 1 w/v						1: 2 w/v						1: 3 w/v						
			5		10		15		5		10		15		5		10		15		
			Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	
110	Total sugars	60.0	52.5	51.99	50.8	50.17	50.0	48.75	54.5	54.48	51.75	52.35	50.3	50.92	54.2	54.43	52.65	53.01	50.5	54.4	1.54
	sucrose	41.29	33.40	33.11	32.6	32.04	30.4	31.33	33.50	34.49	32.6	33.46	31.8	32.75	31.65	33.78	31.3	33.07	30.70	34.3	2.04
	Reducing sugars	18.70	19.0	18.88	18.65	18.13	17.30	17.42	19.95	19.99	19.0	18.89	18.3	8.15	19.61	20.64	18.5	19.93	18.6	20.10	1.24
115	Total sugars	60.0	52.5	51.94	50.8	50.18	50.0	49.48	54.5	53.69	51.75	52.64	50.50	51.94	54.2	55.45	52.6	53.69	51.5	53.90	1.41
	sucrose	41.29	33.40	34.46	32.6	33.41	30.4	34.06	33.50	36.16	32.6	34.46	32.80	33.11	31.65	34.51	31.3	33.81	32.0	33.91	2.00
	Reducing sugars	18.70	19.0	17.48	18.65	16.17	17.30	15.42	19.95	18.52	19.0	18.17	17.7	18.82	19.61	20.93	18.5	19.87	19.5	20.0	1.14
120	Total sugars	60.0	52.5	52.0	50.8	50.48	50.0	48.95	54.5	52.76	51.75	49.33	50.5	47.43	54.20	51.62	52.6	48.57	51.7	53.20	1.45
	sucrose	41.29	33.40	29.81	32.60	29.05	30.4	27.52	33.5	29.05	32.6	26.38	31.8	25.24	31.65	27.14	31.5	25.24	30.4	32.5	2.10
	Reducing sugars	18.70	19.0	22.18	18.65	21.43	17.30	21.42	19.95	23.71	19.0	22.94	18.32	22.19	19.61	24.47	18.5	23.33	21.3	20.7	1.25

3.5 Effect of clarification of sugar crystals affined water by grape wood charcoal during thermal-acid treatments on ash content

Results of affined water of sugar crystals after addition acid-thermal charcoal and / or standard clarifying agent (Tonsil) were presented in Table (5). The changes in ash content were assessed for total ash, water soluble and insoluble ash. Highly significance (P ≥ 0.05) was noticed for total ash, water soluble and insoluble ash in the treatments, charcoal amount, charcoal: acid ratios and temperature degrees. The total ash content of sugar crystals affined water is considerably slightly large. Results revealed that the clarification of diluted affined water resulted in a considerable diminution in the total ash content. Heating of the acid-thermal treated charcoal leads to diminishing total ash, water soluble and insoluble ash content as acid-thermal charcoal increased. Meanwhile, total ash, water soluble and insoluble ash content was increased with increasing the heating temperature of charcoal. Concerning the influence of acid thermal charcoal on the total ash, water soluble and insoluble ash of affined water of sugar crystals (AWSC), it could be noticed that acid-thermal charcoal showed considerably reduced. The negative correlation coefficient between the acid-thermal charcoal levels and total ash, water soluble and insoluble ash were high (r= -0.758, -0.875 and -0.609, respectively) at 120ºC of acid treated charcoal. The linear regression equation for prediction the ash components of affined water of sugar crystals were:

Y(total ash) = 1.13 - 0.025 X (acid-thermal charcoal)

Y(sol. Ash) = 0.753-0.024 X+ 0.000534 $X2$ (acid-thermal charcoal)

Y(insol. ash) = 0.377-0.00067 X - 0.00053 $X2$ (acid-thermal charcoal)

This explains the advantage of clarification by charcoal during acid-thermal treatments in diminution affined water from ash content making it more suitable for refining uses. The clarified affined water of sugar crystals with amount of 10 % of Tonsil had slightly higher total ash, water soluble and insoluble ash content especially, under the higher rates of acid-thermal charcoal. Similar results are very close to those reported by (El-Geddawy, 2013; El-Naggar, 2002; Laskameethansan *et al*., 2012; Miljana *et al*., 2019; Tawfeuk and Gomaa 2017).

Table (5): Effect of clarification of sugar crystals affined water (500 gm/L) by grape wood charcoal during combined acid-thermal treatments on ash components.

Thermal treatments	Ash components	Affined water of sugar crystals	Charcoal: 0.5 N HCl w/v																		LSD 5%
			1: 1 w/v						1: 2 w/v						1: 3 w/v						
			5		10		15		5		10		15		5		10		15		
			Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	
110	Total ash	1.55	0.80	0.85	0.75	0.77	0.72	0.74	0.74	0.75	0.70	0.70	0.70	0.69	0.59	0.60	0.57	0.55	0.53	0.50	0.04
	Water sol. Ash (g/100 mL)	0.95	0.50	0.52	0.49	0.50	0.46	0.45	0.46	0.45	0.43	0.44	0.44	0.44	0.32	0.33	0.31	0.30	0.21	0.22	0.01
	Water insol. ash	0.60	0.30	0.33	0.26	0.27	0.26	0.29	0.28	0.30	0.27	0.26	0.26	0.25	0.27	0.27	0.26	0.25	0.32	0.28	0.02
115	Total ash	1.55	0.80	0.90	0.75	0.81	0.72	0.80	0.74	0.76	0.70	0.73	0.70	0.68	0.59	0.58	0.57	0.55	0.53	0.50	0.03
	Water sol. Ash (g/100 mL)	0.95	0.50	0.55	0.49	0.51	0.46	0.50	0.46	0.43	0.43	0.40	0.44	0.37	0.32	0.33	0.31	0.35	0.21	0.30	0.01
	Water insol. ash	0.60	0.30	0.35	0.26	0.30	0.26	0.30	0.28	0.33	0.27	0.33	0.26	0.31	0.27	0.25	0.26	0.20	0.32	0.20	0.04
120	Total ash	1.55	0.80	1.10	0.75	1.00	0.72	0.90	0.74	0.97	0.70	0.85	0.70	0.73	0.59	0.95	0.37	0.80	0.53	0.65	0.10
	Water sol. Ash (g/100 mL)	0.95	0.50	0.69	0.49	0.60	0.46	0.55	0.46	0.62	0.43	0.55	0.44	0.52	0.32	0.63	0.31	0.55	0.21	0.47	0.12
	Water insol. ash	0.60	0.30	0.41	0.26	0.40	0.26	0.35	0.28	0.35	0.27	0.30	0.26	0.21	0.27	0.32	0.26	0.25	0.32	0.18	0.08

3.5 Effect of clarification of sugar crystals affined water by grape wood charcoal during thermal-acid treatments on true purity and ash ratio

Results of affined water after addition

acid-thermal charcoal and / or standard clarifying agent (Tonsil) were presented in Table (6). Highly significance ($P \geq 0.05$) was observed for purity coefficient and ash ratio in the treatments, charcoal amounts, charcoal-acid ratios and heating temperature degrees. Results revealed that the clarification of sugar crystals affined water resulted in a considerable elevation in both of purity coefficient and ash ratio. Purity percent and ash ratio were calculated from the equations:

$$Purity\,\% = \frac{\%\,Sucrose}{T.S.S} \times 100$$

$$Ash\,ratio = \frac{\%\,Sucrose}{\%\,Ash}$$

Sucrose elevation with increasing charcoal-acid ratio had slightly higher than total soluble solid and / or ash content. The result illustrated that clarification greatly increased affined water of sugar crystals purity and ash ratio. Heating of the acid-treated charcoal leads to elevation purity coefficient and ash ratio as acid-thermal charcoal increased. Meanwhile, purity coefficient and ash ratio were increased with increasing the heating temperature degrees of charcoal up to 115°C, then shrinkage the purity coefficient and ash ratio elevation over 115°C. The clarified affined water of sugar crystals with amount of 10% of Tonsil had greatly lower purity coefficient and ash ratio. The results obtained were nearly similar with those obtained by (El-Geddawy, 2013; El-Naggar, 2002; Miljana *et al.*, 2019). It is worthy to mention that a linear correlation between purity coefficient and sucrose ($r = -0.588$) and T.S.S. ($r = -0.786$). The linear regression equations for prediction the purity coefficient was:

$$Y(\text{purity coefficient}) = -300.68 + 22.772\,X - 0.34995\,X^2\ (\text{Sucrose})$$

$$Y(\text{purity coefficient}) = -33.2144 + 4.753\,X - 0.05423\,X^2\ (\text{TSS})$$

Table (6): True purity and ash ratio of sugar crystals affined water (500 gm / L) treated by grape wood charcoal during combined acid-thermal treatments.

Thermal treatments	Purity and ash ratio components	Affined water of sugar crystals	Charcoal: 0.5 N HCl w/v																		LSD 5%
			1: 1 w/v						1: 2 w/v						1: 3 w/v						
			5		10		15		5		10		15		5		10		15		
			Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	Tonsil	Char.	
110	Purity coefficient	55.5	58.59	58.08	60.37	59.25	58.46	60.25	56.77	58.45	57.19	58.70	57.81	59.54	56.78	60.33	57.96	61.24	59.03	62.88	1.54
	Ash ratio	26.63	41.75	38.28	43.46	41.24	42.22	42.57	45.27	46.26	46.57	45.34	45.42	48.69	53.64	59.50	54.91	61.47	57.92	65.52	1.65
115	Purity coefficient	--	58.59	67.56	60.37	62.18	58.46	70.95	56.77	67.51	57.19	68.92	57.81	70.44	56.78	67.66	57.96	69.00	59.03	69.70	1.59
	Ash ratio	--	41.75	38.95	43.46	41.61	42.22	42.33	45.27	45.98	46.57	47.80	45.42	47.46	53.64	56.30	54.91	61.12	57.92	64.80	1.70
120	Purity coefficient	--	58.59	62.10	60.37	63.15	58.46	64.00	56.77	63.15	57.19	65.95	57.81	66.42	56.78	60.31	57.96	61.56	59.03	61.39	1.50
	Ash ratio	--	41.75	27.10	43.46	29.05	42.22	30.57	45.27	29.95	46.57	31.03	45.42	34.57	53.64	28.56	54.91	31.55	57.92	35.89	1.60

On the other hand, the negative correlation coefficient between ash ratio and sucrose was weak ($r= -0.386$) and total ash was very high ($r= -0.983$). The linear regression equations for prediction the ash ratio was:

$$Y(\text{ash ratio}) = 5125.23 - 294.96\,X + 4.283\,X^2\ (\text{sucrose})$$

$$Y(\text{ash ratio}) = 1.5963 - 0.024\,X + 0.000114\,X^2\ (\text{T.S.S.})$$

Hereby, transmittance light percent, T.S.S., PH, titratable acidity, total sugars, sucrose, reducing sugars, total ash, water soluble and insoluble ash estimate carefully clarification of affined water of sugar crystals. This explains the advantage of clarification by grape wood charcoal during thermal-acid treatments in elevation affined water purity making it more suitable for reused in sugar refining.

4. Conclusion

The clarification of sugar crystals affined water by different combined thermal-acid treatments of charcoal showed that adding thermal-acid charcoal led to high clarified of sugar crystals affined water than those to standard clarifying agent. Further clarification by higher ratio of thermal-acid charcoal (15 charcoal and 1: 3 charcoal-acid ratio) indicated that the in colour and clarification. Highly significance ($P \geq 0.05$) was observed for total sugars, sucrose, reduced sugars, and ash content in treatments, charcoal amount charcoal-acid ratios and heating temperature. The clarified affined water with Tonsil had slightly lower total sugars, sucrose and reduced sugars under the higher ratio of thermal-acid charcoal. The clarified affined water with Tonsil has slightly higher total ash, water soluble and in soluble ash content. Also, clarified affined water with Tonsil had greatly lower purity coefficient and ash ratio.

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