Egyptian J. Anim. Prod., 59, Suppl. Issue, January (2022):97-104

USING LEATHER CHROME SHAVINGS IN PRODUCING GYPSUM BOARD FOR ACHIEVING SUSTAINABILITY

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SUMMARY

Animal skin, as a byproduct of farm animals, is the main raw material for leather tanning industry that contributes to optimizing farm economic revenues. Creating new usage of the skins, residuals and wastes of manufacture processes insures extra income to tanning industry and reduces the contaminants which may be resulted during hides' production. About three million tons of leather chrome shavings (LCS) are generated annually from leather tanning containing trivalent chromium. The utilization of LCS is a way to prevent environmental hazards and avoid economic loss of leather tanning. In this regard, the present study aimed to utilize LSC in different proportions (0%, 10%, 15%, 20% and 25%) to manufacture gypsum boards, and in turn, to evaluate their physical-mechanical characteristics. The pH value and contents of moisture, ash and chroumium salt of LCS, as well as, bulk density, compressive and flexural strengths, thermal conductivity and construction morphology via electron microscopy scanning (EMS) of prepared LCS composites mixtures were determined. The homogeneity and good interactions among fibers of LCS and gypsum were observed in EMS micrographs. The increment of LCS content decreased (P<0.01) density, compressive strength, flexural strength and thermal conductivity in prepared gypsum composites. The results pointed to the possibility of using the LCS in making gypsum boards due to its homogeneous structure. Moreover, the gypsum board with high content of LCS is suitable for using as a filling and separation substance in buildings to increase the thermal insulation, and reduce the construction costs.

Keyword: Leather tanning industry, physical properties and thermal conductivity

INTRODUCTION

Animal production generates meat, milk, wool and hair as main products, additionally hides as byproducts (FAO, 2011). Leather tanning is the way to convert the animal hides or skins into genuine leather to be utilized in a wide range of usages and maximizing their economic value (Husen *et al*., 2016).

During tanning, different mechanical and chemical processes are applied and different liquid, solid, and gaseous contaminants are produced (Gudro, 2011). Solids form the greatest amounts among total leather tanning wastes, accounting for about 80% of raw skins or hides weight (Bank, 1999). Whilst chrome tanning is the most widely used method worldwide (Agrawal and Kumar, 2006), leather chrome shavings (LCS) are the greatest solid waste of leather chrome tanning industry (Kolomaznik *et al.*, 2003). LCS are the output from the shaving process to adjust the desired pelt thickness to be suitable for end uses of finished leathers. Thus, tremendous quantities of LCS are generated, estimated as about 10% of wet salted hide or skin weight that ranks between 35-40% of the total tannery solid wastes (Rao et al., 2002). That quantity was estimated at 3 million tons/year (FAOSTAT, 2017).

The chemical structure of LCS is mainly a collagenous protein cross-linked with chromium salts (Saravanabhavan *et al*., 2005). Depending on tanning process, LCS contains 2.5- 5% chromium mainly in the form of trivalent chromium (Tahiri *et al*., 2007). Globally, the traditional major ways of LCS waste management are landfill or incineration, which creates environmental threats due to the toxicity and carcinogenic effect of chromium salts when converts into the hexavalent form (Pati *et al*., 2014).

Previous investigations aimed to increase the economic value of LCS while decreasing its environmental impact, without damage to the sustainability of leather tanning industry. The utilization of LCS was by using it directly for producing leather boards or treating it for modifying its chemical structure for producing eco-friendly products that can be used in other useful proposes (Sathish Kumar and Vijayaravind, 2015; Eylem and Pere, 2017 and Ponsubbiah *et al*., 2018).

Gypsum board is a panel made of calcium sulfate dehydrates with or without additives, used in the construction of interior walls and ceilings. The panel properties differ according to components. Fibers such as fiberglass, paper, or a combination of them are commonly mixed when producing panels to reinforce them and improve their physical properties (Li and Ren, 2011). Few preliminary investigation, utilized LCS in producing cementitious mortars but no evidence of using it to manufacture gypsum board was detected.

This study aimed to produce LCS-gypsum boards by adding LCS in increasing proportions during gypsum panel manufacture and evaluate the properties and microstructure of the composite. Achieving the maximum utilization of LCS will reduce the environmental hazards, increase the added value of LCS and, consequently ensure the sustainability of leather tanning industry.

MATERIALS AND METHODS

Materials:

Leather chrome shavings (LCS):

LCS was collected from El-Shafei Sons tannery located in El-Max district, Alexandria, Egypt and was naturally dried in an open shaded area for five days. Figure (1) shows the natural fresh LCS without any chemical or physical treatments changes.

Figure 1. Leather chrome shaving waste (LCS).

Characteristics of LCS were evaluated in the chemistry laboratory of Mariout Research Station, Desert Research Center, according to the standard procedures (ASTM, 2014). The studied characteristics were moisture, ash, chromium, fat and protein contents in addition to pH values and bulk density which was calculated as the ratio of weight to volume for each specimen.

Water saturation and water loss for LCS in 48 hrs were determined as

$$R = (W_t - W_0) / W_0 \times 100 \qquad (1)$$

where R is the water saturation or water loss ratio, Wt is the wet specimen weight at a specific time and W0 is the dry specimen constant weight**.**

Gypsum:

The utilized gypsum was produced in Sinai Gypsum Factory and consisted of 95% calcium sulfate semi hydrate ($CaSO_4$. 0.5 H_2O). The chemical composition of the utilized gypsum (Table1) was determined by X-ray fluorescence technique in the Faculty of Science - Alexandria University Laboratories.

Water:

A clean tap water was used for these experiments.

Test method:

Composites preparation:

An attempt was made to use LCS for the production of LCS-gypsum composite mixtures. These mixtures were prepared by hand mixing of gypsum, water and LCS in increasing proportions of 0, 5, 10, 15, 20 and 25% (Table 2). The formulated composites were denoted as LCSGC0, LCSGC5, LCSGC10, LCSGC15, LCSGC20 and LCSGC25, respectively. Water was added to obtain a homogeneous mixture for good workability.

Physico-mechanical properties:

Compressive and flexural strengths:

Wooden molds (4 × 4 × 16 cm) were used for preparing three prismatic test specimens for each studied mixture. The molds were filled with the studied composite mixtures and then left at room temperature for 24 hrs. Thereafter, compressive and flexural strengths were measured according to DIN (2005) at days 1, 7 and 28 of molds filling. Equations (2) and (3) were used for calculating compressive and flexural strengths, respectively as:

$$C_s = F_c / B^2 \qquad (2)$$

where C_s is the compressive strength, F_c is the compressive failure load and B is the sample width.

$$F_s = (3 \times P_f \times L) / (2 \times B \times T^2) \qquad (3)$$

where F_s is the flexural strength, P_f is the flexural failure load , L is the sample length, B is the sample width and T is the sample thickness.

Figure (2) shows specimens after flexural strength test.

Table 1. Basic chemical composition of gypsum

Element (%)	Fe_2O_3	SiO_2	CaO	Al_2O_3	MgO	SO_3	Na_2O	Traces
Gypsum	0.04	0.23	32.7	0.09	0.42	43.68	0.03	22.81

Table 2. Proportions of different leather chrome shaving gypsum composite (LCSGC)

Mixture	Gypsum, g	CTLS, g	Water, g
LCSGC0	500	0	200
LCSGC5	475	25	215
LCSGC10	450	50	230
LCSGC15	425	75	245
LCSGC20	400	100	260
LCSGC25	375	125	275

Figure 2. Specimens after the flexural strength test.

Bulk density:

Density was measured for 3 prisms of dimensions (4 × 4 × 16 cm) for each formulated composite at days 1,3,7 and 28 after filling molds. After drying samples at 105± 5 °C and reaching a constant weight, the weight and volume of each sample were recorded and the bulk density was calculated using equation (4).

$D = W_d / (L \times B \times T)$ (4)

where D is the bulk density, W_d is the dry sample weight, L is the sample length, B is the sample width and T is the sample thickness.

Thermal conductivity test:

Thermal conductivity was measured according to ASTM-D5930 by placing specimen between hot and cold plates. The measurements were made on each of 3 prisms of dimensions 4 × 4 × 16 cm for each specimen at days 1, 7 and 28 after filling molds. The thermal conductivity was calculated using Fourier's law (5):

$K= (Q \times T) / [(T_h - T_c) \times A]$ (5)

where K is the thermal conductivity, Q is the steady state conducted heat transfer across the sample, T is the sample thickness, T_h is the temperature of the hot plate, T_c is the temperature of the cold plate, and A is the heat transfer area of the sample.

Electron microscopy scanning (EMS)

An electron microscopy of JEOL model JSM-5300 with accelerating voltage of 25 kV in the Laboratory of the Faculty of Science, Alexandria University was used to capture electron micrographs for each composite mixture.

Statistical analysis:

Data were analysed using general linear model (GLM) procedure of SAS program for analysis of variance by adopting the fixed effects model:

$$Y_{ijk} = \mu + C_i + A_j + CAij + e_{ijk}$$

Where Y_{ijk} is the observation taken, μ is the overall mean, C_i is a fixed effect of the i^{th} LCS proportion, A_j is a fixed effect of the j^{th} age of specimen, CA_{ij} is the interaction effect between LCS proportion and specimen age, and e_{ijk} is a random error assumed to be normally distributed with mean=0 and variance=σ 2 e. Means were significantly separated using Duncan’s multiple range tests.

RESULTS

Leather chrome shavings (LCS) characteristics:

The normal shapes of fresh LCS pieces were variable in length between less than 1 mm to about 5 cm (Figure 1). The scanning electron micrographs show the fibrous structure of LCS at 1500x and 5000x (Figure 3). The micrographs revealed that collagen fibers emerged as close bundles with diameters lower than 10µm with air gaps appearing among the bundles. Moreover, the smooth surface predominates in the appearance of these fibrous bundles.

Bulk density and chemical properties of LCS are shown in Table (3). The bulk density value being 0.1027 g/cm^3, showed that LCS is a light weight material that can furnish large space. The chemical properties showed that LCS has low moisture content (10.24%) but high contents of trivalent chromium salt (2.51%) in total ash content (12.37%). Additionally, pH of LCS is strong acidic (3.32ml mol/L) when soaked in water.

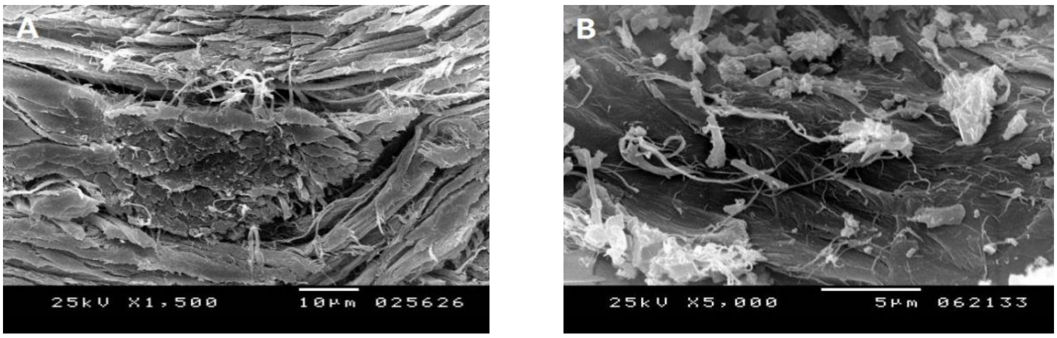

Figure 3. Electron micrograph of leather chrome shaving (LCS), (A) at 1500x and (B) at 5000x.

Table 3. Leather chrome shaving (LCS) characteristics

Parameter	ASTM	Value
Moisture content	D-6403	10.24%
Ash content	D-2617	12.37%
Chromium content	D-6714	2.51%
pH Value	D-2810	3.32ml mol/L
Bulk density	D-6683	0.1027 gm/cm^3

Figure (4) shows the capacity of LCS for water saturation and water loss in 48 hrs. The saturation curve displayed that LCS absorbed water at the ratio of 150-170% of its weight in the first 3 minutes after soaking but absorption continued slower to reach 190% of CLS weight after 60 minutes of soaking. Thereafter, absorption fluctuated slower to reach 192% after 48 hrs of soaking. Regarding the water loss curve, LCS lost water in equal rates until reaching initial dry weight after 24 hrs.

From the saturation curve, LCS absorbed about 150 -175% water during first 3 minutes of soaking, while the water absorption was increased slightly till fixed after 60 min of soaking at 190%. Thereafter, water absorption fluctuated in a narrow range between 190 and 192% till 48 hrs of soaking. Regarding to the loss curve, LCS lost water at a similar rate until it returned to its dry weight again within 24 hrs of taken it out of water.

Characteristics LCSGC:

Flexural strength test showed that the distribution of LCS fibers was perfect in all specimens of the prepared LCSGC (Figure 2) and Figure (5) shows the difference in scanned electron micrographs between both of LCSGC0 and LCSGC25 composites. Gypsum crystals of LCSGC0 showed distinctive needle shape with smooth surface and many overlapping points among crystals, and the air gaps among crystals were small, while for LCSGC25, the gypsum crystal shape and air gaps volume were larger. Adding LCS to composites caused gypsum particles to appear stuck to the collagen fibers and the volume of crystal needles was higher. Thus, the composite between LCS and gypsum particles formed a complex of homogenous structure.

The effects of LCS addition, specimen age and interaction between them on the physical properties of the prepared LCSGC are shown in Table (4). The increment of LCS addition decreased ($P<0.01$) the physical properties of density, compressive and flexural strengths and thermal conductivity and therefore the panels became weaker by increasing the percentage of LCS, but thermal insulation property improved. The density values ranged between 1.25 and 1.98 t/m^3 with continuous decrease by increasing LCS proportion up to 15% then remained constant. Thereafter, other physical properties showed wide ranges of changes with more significant differences among different LCS concentrations.

On the other hand, all physical properties of LCSGCs were not affected by age of specimen except thermal insulation, which was increased ($P<0.05$) from day 7 of composites formulation. A slight decrease in the bulk density was found when specimens' age increased, but an inverse trend was found for compressive and flexural strength values.

Moreover, the effect of interaction between LCS proportions and specimen age was highly significant ($P<0.01$) for all physical properties (Table 4). The changes of physical parameters due to the interaction are shown in Figure (6). The bulk density decreased by the increment of LCS proportion and age of formulated composites, whilst the properties of compressive and flexural strengths and heat insulation were improved. The positive correlations between bulk density and compressive strength, flexural strength and thermal conductivity were strong as shown in Figure (7). R^2 values were 0.973, 0.968 and 0.971 for the correlation between bulk density with compressive strength, flexural strength and thermal conductivity, respectively.

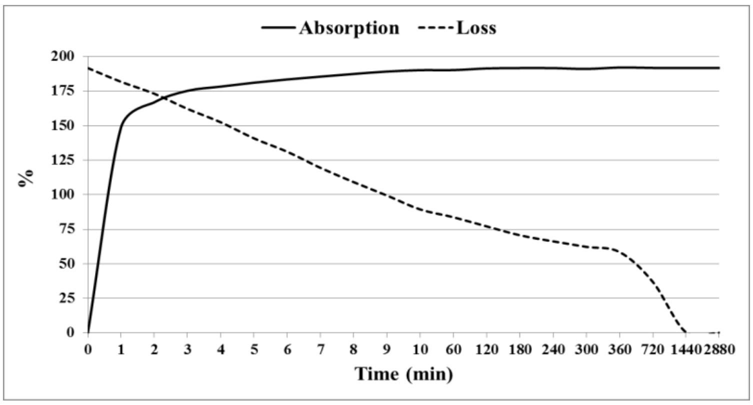

Figure 4. Water imbibition and water loss of leather chrome shaving (LCS).

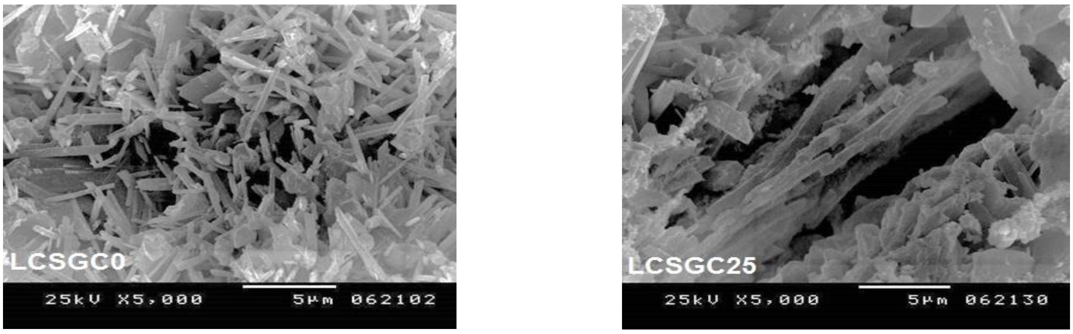

Figure 5. Electron micrographs scanning of LCSGC0 and LCSG25 at 5000X after 28 days of formulation.

Table 4. Means ± SEM of physical properties for LCSGC as affected by LCS content, specimen age and their interactions

Parameter	Density (t/m^3)	Compressive strength (MPa)	Flexural strength (MPa)	Thermal conductivity (W/m°C)
Leather chrome shaving, % by weight (C)	**	**	**	**
LCSGC0	1.98^a	6.91^a	4.36^a	0.599^a
LCSGC5	1.72^b	5.66^b	3.82^b	0.526^b
LCSGC10	1.47^c	4.42^c	3.11^c	0.441^c
LCSGC15	1.34^d	3.62^d	2.73^d	0.392^{cd}
LCSGC20	1.26^d	2.64^e	2.27^e	0.339^{de}
LCSGC25	1.25^d	2.39^e	2.08^f	0.311^e
Age (A)	**ns**	**ns**	**ns**	*
1 day	1.63	3.88	2.89	0.500^a
7 days	1.45	4.33	3.07	0.407^b
28 days	1.43	4.61	3.22	0.397^b
(C) × (A)	**	**	**	**
Mean	1.50	4.27	3.06	0.435
SEM	0.04	0.22	0.12	0.017

Means in the same column having different superscripts are significantly different ($P<0.05$). ns: not significance; * $P< 0.05$; **$P<0.01$; SEM standard error of mean; LCSGC leather chrome shaving gypsum composite. LCS leather chrome shaving

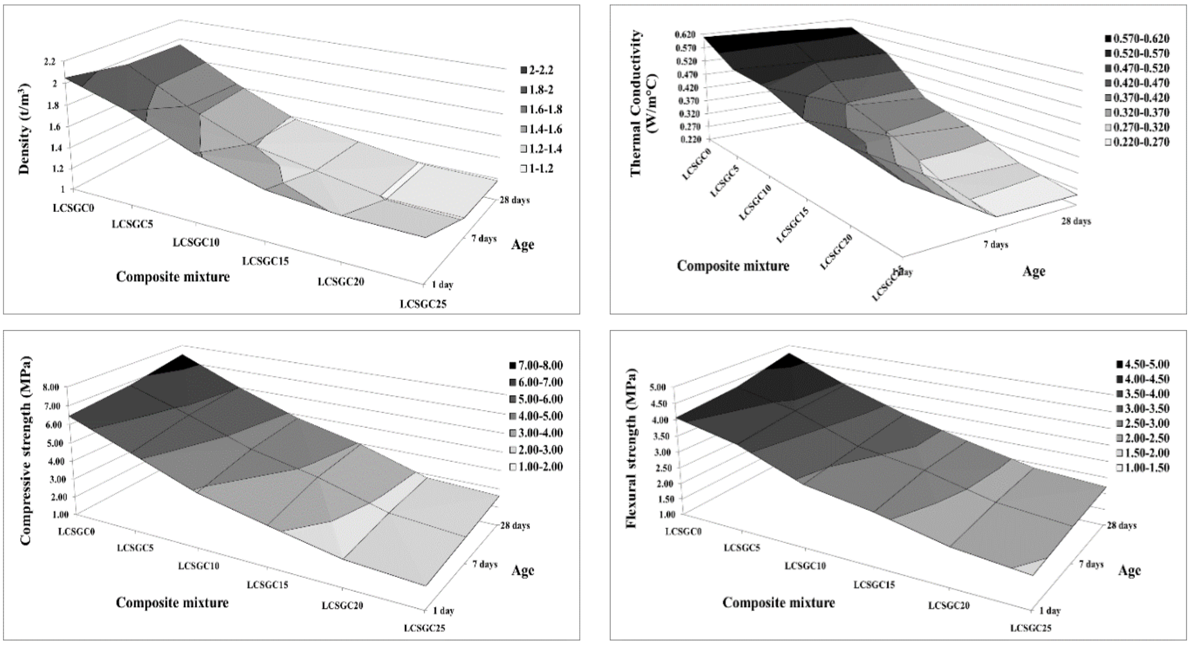

Figure 6. Changes in physical properties of formulated composites due to the interaction between leather chrome shaving proportion and specimens' age.

Table (5) shows the differences in production costs among formulated LCSGC. Addition of LCS resulted in reduction in production costs. This reduction increased gradually by increasing LCS proportion to reach the maximum for LCSGC25. The cost savings rate was 1% for each 1% increase in LCS content.

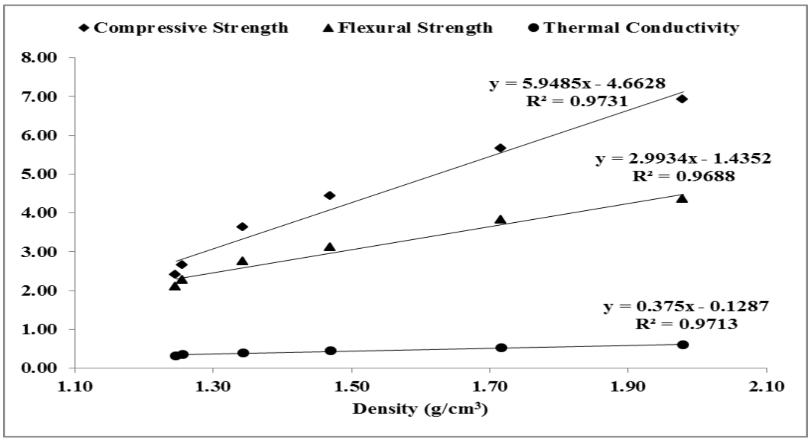

Figure 7. Correlations among bulk density values with compressive strength, flexural strength and thermal conductivity.

Table 5. Production cost of formulated leather chrome shaving gypsum composites (LCSGC)

Group	Gypsum		LCS		Water		Total cost (L.E)	Reduction (%)*
	Quantity (kg)	Cost (L.E)	Quantity (kg)	Cost (L.E)	Quantity (litre)	Cost (L.E)		
LCSGC0	1000	650	0	0	400	2.4	652.40	0.00
LCSGC5	950	617.5	50	0	430	2.58	620.08	4.95
LCSGC10	900	585	100	0	460	2.76	587.76	9.91
LCSGC15	850	552.5	150	0	490	2.94	555.44	14.86
LCSGC20	800	520	200	0	520	3.12	523.12	19.82
LCSGC25	750	487.5	250	0	550	3.3	490.80	24.77

* Reduction percentage calculated based on the cost of LCSG0

DISCUSSION

Based on the present data, the chemical properties of LCS were similar to those of tanned leather (Sethuraman *et al*., 2013; Putshaka *et al*., 2015 and Abebaw and Abate, 2018). After getting rid of some organic substances such as fats and globular proteins during pre-tanning steps (BASF, 2007 and Dutta, 2008), LCS turned into a collagenous material having the expectedly obtained moisture, ash, chromium content and pH value.

Though, the acidity effect of LCS on gypsum setting is unknown, the bonding between gypsum crystals and LCS was strong as shown in Figure (5). Moreover, collagen fibers in LCS were not subjected to deterioration and the hemihydrate reaction of gypsum had no effect on LCS particles, probably due to the strong bond between collagen fibers and chromium salts such as in chrome tanned leathers that is characterized by stability under high temperatures up to 110°C (Covington, 2011).

From the physical point of view, LCS can absorb nearly twice its weight of water in the first three minutes of soaking (Figure 4). That explained why the need for water increased with increasing LCS proportion in the composite (Table 2). Also, water might help gypsum particle to stick better on the collagen fiber due to the high absorbability of LCS and the smoothness of its surfaces. However, LCS loses absorbent water gradually until it is completely lost within 24 hours (Figure 4), which may improve the reaction conditions of gypsum by absorbing excessive water in LCS particles during first 30 minutes until completing the final setting (Li and Ren, 2011).

Moreover, the low bulk density of LCS has (0.1027 gm/cm^3) was in agreement with the findings of Rao *et al*. (2002). It is even lower than that of tanned leathers which ranged between 0.59 and 0.67 gm/cm^3 (Nasr *et al*., 2017). That was probably due to its irregular shapes and dimensions (Figure 2), as well as the presence of internal air gaps among collagen fibers (Figure 3).

The naturally low bulk density of LCS caused decrease in the final bulk density of the formulated LCSGCs depending on the proportion of LCS in the composite. Such decrease in density plays an important role in altering the characteristics of the produced LCSGCs as indicated from the correlation between density and other physical properties (Figure 7).

The decrease in compressive strength, flexural strength and heat conductivity by increasing LCS proportion in the composite were because of the extra porosity induced by more LCS fibers and the nature of LCS porous. Adding different types of fibers when formulating composite including leather wastes showed the same trend (Lakrafli *et al*., 2012; Abuh and Umoh, 2015; Reddy *et al*., 2016; Zhua Cong *et al*., 2018 and Selamat *et al*., 2019).

Although the effects were insignificant (Table 4), specimen age brought about an improvement in the physical properties of LCSGCs. By time, water is lost by evaporation and the gaps within LCSGC composites become full of air instead of water, which lead to decrease in the values of bulk density and thermal conductivity. On the other hand, the values of compressive and flexural strengths were improved due to increased gypsum hardness (Li and Ren, 2011).

The highly significant interaction effect between LCS proportions and specimen age on characteristics of LCSGCs was beneficial. The strength of LCSGC composites improved by time pass and by lowering LCS contents in gypsum composites, but the opposite trend was noticed for heat insulation property. Therefore, the gypsum composite LCSGC0 was the strongest but the lowest for heat insulation. Also, LCSGC25, which contained the highest LCS proportion, was the weakest but the highest for heat insulation. Consequently, to reach a compromise LCSGC5 and LCSGC10 composites may improve the thermal insulation properties with a marginal saving of about 5% and 10% in the production costs, respectively (Table 5).

CONCLUSION

The present study indicated that the utilization of chrome shaving wastes in the construction field will enhance the sustainability in leather tanning sector while achieving distinctive environmental and economic benefits. Adding leather chrome shaving when formulating gypsum composite enables its use as a construction decoration material, increases the void gaps among gypsum particles, enhances collecting crystal needles of gypsum on added fibers and more important enhances its utilization as thermal insulator of high durability. Adding 5-10% leather chrome shaving should improve heat insulation 12 - 25% but decreases the strengths 15 - 25%, while lowering the production cost 5 -10%.

Consequently, using chrome tanned leather wastes as insulation material not only presents a good solution to save energy but also resolve the problems of handling leather industry wastes.

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إستخدام مخلف سلاتة الكروم في إنتاج ألواح الجبس لتحقيق الاستدامة

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يعتبر جلد الحيوان منتج ثانوي للمزارع الحيوانية، وهو المادة الخام الرئيسية لصناعة دباغة الجلود. لذلك فإنه يساهم في تحسين الإيرادات الاقتصادية للحيوانات الزراعية. ويؤدي استخدام الجلود ومخلفاتها في عمليات التصنيع إلى ضمان المزيد من الدخل للمزارعين وكذلك تقليل الملوثات التي قد تنتج أثناء إنتاج الجلود. يقدر إنتاج مخلف سلاتة الكروم حوالي ٣ ملايين طن سنويًا وتحتوي على املاح الكروم الثلاثية. لذا فإن تدوير مخلف سلاتة الكروم يعتبر وسيلة لتجنب المخاطر البيئية والخسارة الاقتصادية الناتجة عن دباغة الجلود. وفي هذا الصدد استهدفت هذه الدراسة الاستفادة من هذا المخلف بنسب متفاوتة (٠٪ ، ١٠٪ ، ١٥٪ ، ٢٠٪ و ٢٥٪) في تصنيع ألواح الجبس وتقييم خصائصها الفيزيائية. حيث تم تقدير قيمة الأس الهيدروجيني ومحتويات الرطوبة والرماد وأملاح الكروم في مخلف السلاتة، علاوة علي تقدير الكثافة الظاهرية وقوة الانضغاط والانحناء ، والتوصيل الحراري و الفحص المجهري الإلكتروني لعينات ألواح الجبس المعدة من التجربة. وقد بينت النتائج وجود تجانس والتحام جيد بين ألياف سلاتة الكروم والجبس من خلال الصور المجهرية للميكرسكوب الاليكتروني. وأدت الزيادة في محتوى السلاتة بالعينات الى انخفاض الكثافة معنوياً وكذلك انخفاض قوة الانضغاط وقوة الانحناء والتوصيل الحراري. كما أشارت النتائج إلى صلاحية استخدام هذا المخلف في صناعة الألواح الجبسية بسبب تجانسها في التوزيع وتلاحمها مع الجبس. ولذلك فإن عمل ألواح الجبس ذات المحتوى العالي من مخلف سلاتة الكروم تبدو مناسبة للاستخدام كمادة فاصلة في المباني لزيادة العزل الحراري علاوة علي تقليل تكلفة الإنتاج.