Research Article

Preparation and characterization of waste papers based composites and their applications in leather industries

Mahmood M. Barbooti, Rola A.K. Abbas, Hajer A. Ali
Department of Applied Science, University of Technology, IRAQ
*Correspondent author email: rafigyehya@yahoo.com

Introduction
The accumulation of waste materials is associated with a lot of problems. The international community is working on developing efficient methods for dealing with such expanding task. The recently accepted 4R, reduce, reuse, recover, and recycle, approach receiving wide acceptance to limit the...
extensive consumption of the natural resources in addition to energy and water. Any of these options is linked with the type, composition, and quantity of the wastes [1]. Recovery allows the extraction of valuable materials from industrial waste materials employing simple low energy consuming methods [2]. Wastewater is treated to suite the same purposes or directed for other important uses like irrigation [3]. The recycle of waste materials will contribute much to the reduction of disposal and in the same time provides a new resource of raw materials for the industry. Thus, the recycle is a promising approach in this respect.

Recycling include a series of operations to change the used materials into usable products. The shortage of resources was the main reason to develop operations that fulfill such shortages. Meanwhile, the recycling processes of metals, papers, rubber and other products lowered the need to construct more landfills for waste materials and hence participated in the environmental production [4].

Materials and Methods

Apparatus

Forging of the specimens were performed on Lab two roll mill Machine model XK-100 supplied from China. Thickness was measured with a digital micrometer mode IDM-D0007 from Australia. The compressibility measurements were carried out on Leybold Harris Equipment Model N.36110 of British origin. Both shrinkage and moisture content were determined with hot air sterilizer model YCO-010 supplied from a German-Taiwan company. Water absorption measurements were performed on a homemade kit in accordance with the leather industry specification a steel hollow cylinder 8-cm diameter was used as a die to cut the specimen. The surface was allowed to be in contact with water for 60 and 120 minutes and the gain in weight was recorded to evaluate the percentage water absorption of the specimen and expressed as g per unit area.

Materials

Used papers were collected and used in the preparation of pulp as the main component of the desired composite. The papers were shredded into small pieces and immersed in water in a bucket for several hours to facilitate the mixing. The soaked material were then mechanically stirred at moderate speed for 2-3 hrs to end up with homogenous pulp. The wet pulp is dehydrated by manual pressing inside a cotton cloth and left to dry at room temperature to be stored at a low temperature to prevent any biological degradation.

Latex: this was a commercially available adhesive emulsion usually used by the shoemakers.

Paraffin wax: This was a product of a local refinery and has a melting point of 66 °C, congealing point of 63 °C, oil content, 4.4 wt%, penetration, 21 mm, and ash content of 0.17 wt% [5].

Woven acrylic fibers: A woven fabric was employed for the reinforcement of the pulp composite. It is available within the market and used in curtains. The material was identified by infrared absorption spectrum [6].

Paperboard preparation

The pressing of the pulp allows keeping some of its water content. The solid content of the pulp was determined during the preparation of each specimen. This was carried out by placing a known weight of the pulp in an oven and heats it until constant weight is reached. The pulp was mixed for few minutes with twice of its weight of water in a steel container. Wooden frames with 20×20 cm dimensions were prepared for the shaping of the pulp composite. The frame is composed of two identical parts to place a cotton cloth in between and to allow the draining of excessive water content. The mixture was placed within the frame and allowed to drain by pressing with a piece of cloth to ensure uniform surface. Ceramic tiles were then placed on the upper surface to aid further drainage. The piece was removed from the frame and partially dried in an oven to reduce the water content. The material was enclosed between two cloth fabrics and
manually pressed with hot iron to aid both drying and packing. The product was a piece of paperboard similar the ones available for book binding.

**Preparation of composite specimens**

The pulp and water was placed with the mixing container and the calculated amount of the latex was added during stirring. Stirring was continued for enough time (usually 60 min) to ensure the intimate mixing and homogenization. The procedure was completed as for the paperboard preparation above.

**Wax introduction:** To improve the water absorption of the specimens the boards need to be reinforced with water repellent additive. Wax is considered a good candidate for such property being also a cost effective material. Increasing amounts of 1, 2, 4, 5, 6 and 10 g of wax were added to pulp-latex composites (50, 20 g, respectively). The insertion of wax required some prior process to ensure the homogeneous distribution of the wax within the fabric. The water repellency of wax necessitated the addition of molten wax to the latex to utilize the presence of the emulsifier in the latex. Both components were added to the water – pulp mixture as a single additive and reasonably homogenous mixture could be achieved.

The third group of samples was those with the acrylic woven fibers. The woven fibers were placed in the wooden frame and the prepared pulp composites were added to ensure the inclusion of the fibers within the specimen. The same procedure was employed for the finishing. The overall composition of the prepared specimens is given in Table 1.

**Forging:** The manual pressing of the specimens is still inadequate for ensuring the required homogenization and compaction of the materials and consequently the achievement of the desired properties of the product. Forging is an important step during the preparation of the specimens to ensure enough packing and hence intimate mixing of the components. The sample passes between solid cylindrical rolls to reduce the thickness of the specimens and give acceptable shape of the specimens. The process may be done on hot or cold rolls of Hitop Lab Two Roll Mill Machine.

**Physical tests**

Some of the physical tests were carried out at the State Company for Leather Industries, Ministry of Industry and Minerals, National Center for Packing Research.

**Result and Discussion**

The experiments were designed to allow the improvement of the properties of the product to approach those of the commercially available insole product. The start-up was the study of the effects of the contents of the main components, namely, the pulp and latex, on the basic properties.

**Compressibility Test**

A series of specimens were prepared with various pulp and latex weight ratios, 3:1, 2.5:1, and 2:1, respectively, and subjected to a variety of physical tests. The 3:1 weight ratio sample showed relatively the highest compressibility values among the studied specimens which was also the closest to the behavior of the commercially available insole material. The increase in compression resulted in crushing of the specimens (Figure 1). The inclusion of latex results in lower crushing tendency than normal cardboard as a result of the improved strength [7]. The behavior was lower to comparable to that of commercial Dixon. This test is dependent on the contents of the reinforcing materials. The high density of the commercial specimens showed higher compressibility. The specimen of the highest density among the specimens gave the best compressibility results [8] by reference to (Table1). The bulk density values of the prepared samples (Table 2) are relatively smaller than that of the commercial Dixon. This reflects the significance of the preparation technology on the properties and consequently the behavior of the material.
### Table 1: The overall composition of the prepared specimens is given in

| NO. | Percentage of Acrylic fibers (g) | Percentage of paraffin wax (g) | Percentage of Rubber (latex) (g) | Percentage of pulp paper (g) | Components of the sample |
|-----|---------------------------------|-------------------------------|---------------------------------|------------------------------|--------------------------|
| 1   | Pulp paper                       | 100                           | --                              | --                           | --                       |
| 2   | Pulp paper +Rubber (latex)       | 40                            | 20                              | --                           | --                       |
| 3   | Pulp paper +Rubber (latex)       | 50                            | 20                              | --                           | --                       |
| 4   | Pulp paper +Rubber (latex)       | 60                            | 20                              | --                           | --                       |
| 5   | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 20 | 1 | -- |
| 6   | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 20 | 2 | -- |
| 7   | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 20 | 4 | -- |
| 8   | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 20 | 5 | -- |
| 9   | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 20 | 6 | -- |
| 10  | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 20 | 10 | -- |
| 11  | Pulp paper +Rubber (latex) + Paraffin wax | 60 | 20 | 1 | -- |
| 12  | Pulp paper +Rubber (latex) + Paraffin wax | 60 | 20 | 2 | -- |
| 13  | Pulp paper +Rubber (latex) + Paraffin wax | 60 | 20 | 4 | -- |
| 14  | Pulp paper +Rubber (latex) + Paraffin wax | 60 | 20 | 5 | -- |
| 15  | Pulp paper +Rubber (latex) + Paraffin wax | 60 | 20 | 6 | -- |
| 16  | Pulp paper +Rubber (latex) + Paraffin wax | 60 | 20 | 10 | -- |
| 17  | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 20 | 1 | 0.7 |
| 18  | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 20 | 2 | 0.7 |
| 19  | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 20 | 4 | 0.7 |
| 20  | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 20 | 5 | 0.7 |
| 21  | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 20 | 6 | 0.7 |
| 22  | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 20 | 10 | 0.7 |
| 23  | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 15 | 2 | 0.7 |
| 24  | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 25 | 2 | 0.7 |
| 25  | Pulp paper +Rubber (latex) + Paraffin wax | 50 | 30 | 2 | 0.7 |
| 26  | Pulp paper +Rubber (latex) + Paraffin wax | 60 | 20 | 1 | 0.7 |
| 27  | Pulp paper +Rubber (latex) + Paraffin wax | 60 | 20 | 2 | 0.7 |
| 28  | Pulp paper +Rubber (latex) + Paraffin wax | 60 | 20 | 4 | 0.7 |
| 29  | Pulp paper +Rubber (latex) + Paraffin wax | 60 | 20 | 5 | 0.7 |
| 30  | Pulp paper +Rubber (latex) + Paraffin wax | 60 | 20 | 6 | 0.7 |
| 31  | Pulp paper +Rubber (latex) + Paraffin wax | 60 | 20 | 10 | 0.7 |
| 32  | Cartoon                          | --                            | --                              | --                           | --                       |
| 33  | Dixon                            | --                            | --                              | --                           | --                       |

The introduction of wax into the composite resulted in improved properties. Fig 2 show the relation between the stress and the strain for specimens of 50 and 60 g pulp reinforced with latex and wax at various levels. The specimens with 50 g pulp, 20 g latex and 1 g wax exhibited comparable compressibility to commercial Dixon. Similarly, the specimen with 60 g pulp, 20 g latex and 2 g wax showed the highest compressibility among the prepared samples. The lowest level of tensile strength recorded was 3.0 Mpa for specimens with 50 g pulp content. The values could be raised up to 7.58 Mpa by the reinforcement with latex and insertion of wax and stands as the highest value for specimens with 60 g of pulp. This may be attributed to the lower mechanical properties of the wax [9]. Thus, the increase of pulp content overwhelmed the worsening effects of the wax.
Figure 1: The effect of pulp content on the crushing resistance as compared with Dixon.

Table 2: Bulk density for the prepared specimens

| Sample                          | Bulk Density (g/cm³) |
|---------------------------------|----------------------|
| Car.Lab.mi.(100g)              | 1.41                 |
| Re.Car.fa.                     | 1.74                 |
| Car.Lab.mi.(40g)+Lat.          | 1.27                 |
| Car.Lab.mi.(50g)+Lat.          | 1.29                 |
| Car.Lab.mi.(60g)+Lat.          | 1.41                 |
| Dixon                           | 1.96                 |

Figure 2: The effect of wax introduction on compressibility of pulp-latex composites with 50 g pulp content. The compression elasticity modulus was calculated for the wax inserted samples and the values are compared to that of the commercial Dixon as shown in Figure 3. As can be seen in Figure 4, the increase in the pulp content (60 g) caused a clear improvement in the compression resistance for the specimens inserted with paraffin wax. The compression resistance increased from 11.8 MPa as a minimum for specimens with 50 g pulp content up to a maximum value of 20.6 MPa for specimens with 60 g pulp content reinforced with latex.
and wax. Any defects during the preparation, that influence the distribution or concentration stress spots within the sample, will play an important role in the failure of the samples [10].

![Graph](image1)

**Figure 3:** The calculated compression elasticity modulus values for the wax-inserted samples as compared to commercial Dixon.

![Graph](image2)

**Figure 4:** Representative results compression resistance for wax treated specimens with 60 g pulp contents as compared with commercial Dixon.

Regarding the crushing resistance, the pulp composites reinforced with latex and wax showed higher values as the wax introduction decrease and its very close to Dixon (Figure 5). The insertion of woven synthetic fibers into the pulp-latex composites improved the behavior to match that of the commercial Dixon as indicated in Figure 6.
Figure 5: The crushing resistance of pulp composites reinforced with latex and wax.

Figure 6: The effect of woven fiber insertion on the compressibility behavior of pulp-latex composites at various wax contents.

The compression resistance values in the presence of the woven fibers acceptable in comparison with the commercial Dixon for wax contents of 1-2g (Figure 7). Water content and shrinkage on heating values of the specimens treated with wax were evaluated for the prepared specimens and compared with those of the Dixon. for two levels of pulp contents, 50 and 60 g. There was no correlation of the recorded shrinkage values with the wax content. This indicates only the effects of processing methods rather than wax effect. Generally, the water content values were in the range of 1.8-2.6 and showed similarity with the Dixon water content (3.1). The percentage shrinkage on heating for the composites treated with wax was in the range
of 1.8-4.0% in comparison with the Dixon value of 1.8%. The choice of the acrylic fibers for the reinforcement was based on the ability to keep the strength even after wetting [11] and this is one of the main characteristics of the insole products. Following the insertion of woven fibers, the water content values were in the range of 1.7-2.6 which were close to that of the Dixon water content (3.1). The percentage shrinkage on heating for the composites treated with wax was in the range of 1.8-3.8% in comparison with the Dixon value of 1.8%.

Figure 7: The compression resistance values in the presence of the woven fibers for pulp-latex composites with 60 g pulp content and various wax contents.

Water absorption Test
Water absorption is one of the defects in shoe lining material (insole) because it limits the service life of the material. Normal card board absorbs much water and hence fails in test as an insole material [12]. The main function of wax insertion in the composite was to improve the water absorption resistance. Wax contents up to 2g did not improve the resistance and the major improvement was noticed with specimens containing 4g wax and up. Figure 8 shows the comparison of the performance of pulp-latex composites, of 50 g pulp content, inserted with increasing wax contents with commercial Dixon. The behavior of specimens with 6g and more exceeded the commercial Dixon, the choice will be limited to the range of 4-5g on basis of the other mechanical properties of the wax containing samples. Similar behavior could be obtained for composites with 60 g pulp contents.

Effect of latex content
After establishing the various components effects on the physical properties and performance of the composite, a concluding study was carried out to optimize the latex content in the presence of all other components studied. The pulp content was fixed at 50 g, the wax at 2.0 g and woven fiber to suite the area of the specimen. Three latex contents are used of 15, 25 and 30g. To compensate for the wax content the water absorption values were measure for composites with small wax contents and increasing latex contents. The increase of latex in the composites improved the resistance to water absorption. Thus, a compromise was reached to restore the mechanical properties of the composite by limiting wax content and slightly increasing the latex in the mixture.
Conclusions

Paper pulp can be prepared from used papers to be utilized as the basis of various composite materials to fit the various uses. Rubber latex is a typical binder material that can reinforce the paper board and improve its strength and flexibility. The water absorption is an undesired property of the paper board and must be overcome to produce water resistant boards. Wax could be inserted within the paper board structure by incorporating it within the latex utilizing the presence of the emulsifier. The inclusion of paraffin wax at high rates improves the resistance to water absorption but harm to the mechanical properties. To improve the flexibility of the product and water absorption woven acrylic fibers were inserted within the pulp–latex during the molding process. Forging highly improved the homogeneity and characteristics of the products. The optimum performance of the specimens as a replacement of the commercial insole material was 50 g pulp, 20 g, latex, 2-4 g wax and about 1 g woven synthetic fibers. The production technology is highly effective for successful preparation of insole-like material.

References

[1] S.M. Al-Haggar, Basics and mechanisms of sustainable development, Dar Fikr Arabi, 1st Edn., Cairo, 2014.
[2] M. M. Barbooti, M. A. Zablouk, and U. A. Al-Zubaidi, Recovery of Chromium From Waste Tanning Liquors by Magnesium Oxide, Internat. J. Indus. Chem., 1 (2010) 29-38.
[3] A.H.M.J. Allobaidy, M. A. Al-Sameraiy, A. J. Kadhem, and A. Al-Mashhady, Evaluation of Treated Municipal Wastewater Quality for Irrigation, J. Environ. Protect., 1 (2010) 216-225.
[4] F. Abdulrahman, "Reduce, Reuse, Recycle, Alternative for Waste Management", Guide G-314, NM University, Las Cruces, 2014.
[5] A. S. Abbas, S. K. A. Baro, "DE-Aromatization of Paraffin wax", J. Engineering, 11(4), (2005), 715-722.
[6] ISU MatE453/MSE 553 - Lab 3 - FTIR, https://sites.google.com/site/isumate453lab3group8/data/polymethyl-methacrylate-pmma
[7] R. Lituri, J. Deforte, "The Design Guide for Bonding Rubber and Thermoplastic Elastomers", U.S.A.,Vol 2, (2005).
[8] N.E.Marcovich &M.A.Villar, “Journal of Applied Polymer since” Vol.90, PP.2775-2784 (2003).
[9] Speight, J.G., and Ozum, B. 2002. Petroleum Refining Processes, Marcel Dekker, New York.
[10] D. Askeland "The science and Engineering of Material" Second Edition Chapman and Hall London, (1995).
[11] Gary J.Capone, James C.Masson, "Acrylic fibers-Encyclopedia of Polymer Science and Technology", DOI:10.1002/0471440264.pst008, by John Wiley & Sns, Inc, (2004).
[12] A. Wilson, M. A. Carter and W. D., "Materials and Structures and water absorption tests: A critical evaluation", Vol. 32. October 1999, pp 571o578 British Standard M. Hoff Department of Building Engineering, UMIST, P 0 Box 88, Manchester, M60 1QD, UK.