UMYU Scientifica, Vol. 4 NO. 3, September 2025, Pp 232 – 241
https://doi.org/10.56919/usci.2543.023
ISSN: 2955 – 1145 (print); 2955 – 1153 (online)
A periodical of the Faculty of Natural and Applied Sciences, UMYU, Katsina

ORIGINAL RESEARCH ARTICLE

Effect of Acrylic Polymer Dispersions on Water Vapour Permeability and other Physical Properties of Finished Leather

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ABSTRACT
The research investigates the effect of acrylic polymer dispersions on the water vapour permeability of finished leather. An acrylic-based commercial binder, AE 558 Nycil, has been characterized, and its effect when applied in a finish formulation of originally retanned leathers. The binder intrinsic viscosity was found to be 227 dL/g, with of viscosity molecular weight (Mv) of $4.03x10^5$ at solution viscosity measurement of the solid polymer toluene at 25 °C, melting temperature of the solid binder was found to be at the range of 361.7°C - 370 °C, the Formulations for leather finishing were prepared containing the binder at varied proportions of 125 g, 150 g and 175 g was applied on the leather substrates corresponding to samples A1, A2 and A3 respectively they physical properties of coated samples conducted. The water vapour permeability of the originally retanned (uncoated) leathers was measured using the Ostwald method, using the same viscometer reduction was noted significantly after the finish was applied. A1 shows the lowest permeability at 125 g, and A3 has the highest at 175. A2 recorded the highest Shore A value at 150g of the binder in the formulation, while A3 with the lowest Shore A value at 175 g of the binder in the formulation. Distension and Bursting strength of the uncoated leathers were improved after the leathers were coated there was no particular trend in effect as the quantity of the binder in the finish formulation increased. In conclusion, the result of the study showed that the binder is a high polymer material that qualifies it for use in leather finishing, with sample A3 having the best resistance to wet rub action

ARTICLE HISTORY
Received February 19, 2025
Accepted September 12, 2025
Published September 30, 2025

KEYWORDS
Acrylic polymer, Water vapour permeability, finished leathers, Shore A Hardness Tester, Lastometer test

INTRODUCTION
Polymer binders are the main components of aqueous finishing preparations, and the three chemically different synthetic types of binders widely used in leather finishing include acrylates, butadiene, and urethanes. Acrylic polymers have found extensive use in leather finishing as a result of their basic properties such as softness, heat and light stability, and favourable economics (Mamza and Folaranmi, 2023). Acrylic polymers contain hydroxyl groups that are utilized in base coats as binders which bind the pigments together by a catalyzed cross-linking mechanism with a polyisocyanate hardener providing a chip-resistant coating these coatings are usually two-component systems, this explain that the binder and cross-linker are stored separately and mixed to form a pot mix prior to application also the binder are synthesized by emulsion polymerization (Eid *et al.,* 2017).

The physical properties of acrylics (such as gloss, hardness, adhesion, and flexibility) can be modified by altering the composition of the monomer mixture used in the polymerisation process. Acrylics are used in a wide range of industries, and the list below is simply a selection of some of the more common examples: Adhesives, textile industry (e.g. making sponge fill used in padded jackets), paper coatings, paint industry particularly in paints used for road markings (Mamza and Folaranmi, 2023).

Acrylic polymers, when applied as coatings or treatments to substrates like leather, textiles, or films, influence their properties in various ways, knowing that leather is a durable and versatile material that holds a significant place in human history and culture (Hirose *et al.,* 2019).

Leather is derived from the tanning and processing of animal hides or skins, resulting in a material with remarkable properties and applications. Leather has been utilized for various purposes, from clothing and accessories to upholstery and industrial goods. Its distinct attributes of strength, texture, and aesthetics have

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How to cite: Abdulkadir, S. A., Zakari, Y. I., Okpanachi, C. B., Mohammed, I. A., Yusuf, O. L, & Ojobo, L. O. (2025). Effect of Acrylic Polymer Dispersions on Water Vapour Permeability and other Physical Properties of Finished Leather. *UMYU Scientifica*, 4(3), 232 – 241. https://doi.org/10.56919/usci.2543.023

contributed to its enduring popularity (Richard et al., 2015). The properties of leather depend on the origin of the raw materials, how the pelt is prepared for chemical modification, how that modification is conferred chemically, how the leather is lubricated, and finally, how the surfaces are prepared. Leather can be made as stiff and tough as wood, soft and flexible as cloth, and anything in between. It is the traditional art and craft of the leather technologist to control the parameters and variables of processing to make leathers with defined desired or required properties. It is from the creativity of the leather scientists that the range of leathers that can be made is continually widening (Keyong et al., 2023).

The effect of acrylic polymer dispersion on water vapor permeability was measure using the Ostwald method is a simple method for the measurement of viscosity, in which viscosity of liquid is measured by comparing the flow times of two liquids of equal volumes using same viscometer and other physical properties such as hardness, rub fastness, and lastometer performance can be a complex and significant aspect of material development (Abd El-Ghaffar, *et al.,* 2017).

Mamza and Folaranmi (2023) successfully determined the molecular weight of polymer blend systems by comparing their viscosities using the solution viscometric method. The result of their experiment confirmed that the versatility of viscometric techniques and density measurement is not affected by the choice of solvent. The melting temperature is the temperature at which the crystallites of a polymer melt and the polymer becomes a viscous liquid. The process of melting involves the separation of chains in crystalline regions, so that melting points will inevitably depend on interchain forces.

Ibrahim and Gawad (2024) reported that small particle size binders have better penetration properties. These are good for impregnation, but less suitable for covering grain defects, and not good for embossing. The acrylic binders are very useful for tightening the grain. The harder a product is based on the same chemistry, the better the grain tightens. That means that an acrylate with a shore A3 hardness of 65 -70 $^{o}_{c}$ would be better than the one with 25 ^{o}C. On the other hand, the leather becomes slightly firmer. The shore A3 hardness recorded in this article is higher than the reported ones

Due to the increasing incidence of worn shoe complaints involving lack of finish fastness, and other performance defects, thus leading to the investigation of the influence of finish components and impregnating resins as it was difficult to investigate a comprehensive range of resin dispersions, thus simple formulations of acrylic-based binders have been selected.

Water vapour permeability and wet rub fastness of finished leathers are two opposing physical properties. Therefore, any attempt to improve one could jeopardize the performance of the other. Hence, the leather finisher would have to decide on the type of finish and formulation of the mix to strike a balance for optimal performance. And talking of choice, acrylics are used in top coats, mostly in admixture with urethanes, indicating an ongoing interest in their further development as a versatile resin binder. There is a number of aqueous dispersed resin binders based on acrylics, polyurethanes, and synthetic rubbers available commercially in the market. After selecting the proper binders, the finish formulation is worked out according to requirements (El-Nahass *et al.,* 2017).

MATERIALS AND METHODS

2.1 Materials

The Leather made from the skin of a goat was used for this research. It was obtained from the Nigerian Institute of Leather and Science Technology (NILEST), Samara, Zaria.

Finishing Consumables

Acrylic resin binder (Nycil, AE 558), Wax (Lepton-Wax A, Basf), Penetrating agent (EE 8044, Pixel Colour), Liquid Syntan (Syntan-Re, Smit-zoom), Powdered Syntan (Syntan-SA, Smit Zoom), *Bagaruwa* Vegetable Tannin), Wet sheep skins, and Toluene (99% Sigma Aldrich, UK).

2.2 Methods

A reversible reaction between an acrylic acid and an alcohol prepared monomers:

CH_2= CR-COOH + R' OH↔CH_2=CR-COOR' + H_2O
Acrylic acid alcohol alkyl acrylate

Measurement of Viscosity of the Resin Binder

The solution viscosity measurement of the Binder was carried out at 25 ^{o}C using toluene as the solvent. 1 g of the solid polymer was dissolved in 50 ml of the solvent to give a stock solution of 0.02 g/dl. The stock solution was divided into four portions; one was left that way, while the other three were further diluted by adding the solvent in the order of 5 ml, 10 ml, and 15 ml, respectively. 10 ml of the pure solvent was introduced into a viscometer (state the type of viscometer in brackets) and the elution time, t_0 was obtained. This was repeated for each solution, and the corresponding elution time was obtained and recorded as t_1, t_2, t_3, and t_4, respectively (Keyong *et al.,* 2023).

Melting point determination of the resin binder: A melting point capillary was used. A tiny sample of the resin binder was placed on a piece of weighing paper. The samples were placed into three melting point capillaries which have been previously sealed at one end. The capillaries containing the samples were then inserted into the sample compartment of the melting point apparatus (Barnstead Electrothermal A9100, UK), and the instrument was switched on with a set temperature of 0-400 ^{o}C. The capillaries containing the samples were heated slowly, and the temperatures at which melting occurred were observed and read out from the instrument's scale (Keyong *et al.,* 2023).

Preparation of leather substrate: Three (3) pieces of sheep skins in the blue state were weighed (4.5 kg) and

then soaked in 9 kg of water (i.e., 200% of the sheep skins) at 50 °C for 15 minutes. The leather samples were then neutralized under these conditions with 1% $aHCO_3$ for 45 minutes, and the resulting pH of the bath was determined at the end of the operation. The samples were then rinsed with 200% water at 50 °C for 15 minutes. The leather samples were then retanned using the following retanning agents: liquid syntan (4 %) for 20 minutes, powdered syntan (6%) for 10 minutes, and *bagaruwa* (6%) for 60 minutes sequentially in accordance with established procedures (Keyong *et al.,* 2023) in 200% water at 60 °C. Finally, 6% of fatliquor was added in 80 % water at 60 °C for 30 minutes. The leathers were then horsed up overnight, hanged to dry at room temperature for 45 minutes. The samples were then conditioned and hand-stacked before finishing. The leathers were cut into two groups of three leathers each in order to finish them according to one with the finish labeled A1, A2, and A3, the other without a finish labeled B1, B2, and B3, respectively, with established procedures in 200% water at 60 °C. Finally, 6% of fatliquor was added in 80% water at 60 °C for 30 minutes. The leathers were horsed up overnight, hanged to dry at room temperature for 45 minutes, and the samples were conditioned and hand-stacked before finishing. The samples were cut into two groups of three leathers each in order to finish them according to one with the finish labeled A1, A2, and A3, the other without a finish labeled B1, B2, and B3, respectively.

Figure 1 A: Samples

Figure 1B: Samples

Preparation of the finish formulations

The ingredients for the preparation of the finish formulations were weighed in grams (g) using an electronic weighing balance and were introduced into five containers and was stirred vigorously for proper mixture. Three different quantities of the resin binder were incorporated as shown in Table 2 below

Table 2: Finish Formulations

Additives/Ingredients	Formulations (g)		
	A1	A2	A3
Acrylic Resin	125	150	175
Pigment	6.25	6.25	6.25
Water	50	50	50
Penetrator	6.25	6.25	6.25
Wax dispersions	8.75	8.75	8.75

Figure 3a: Pigments

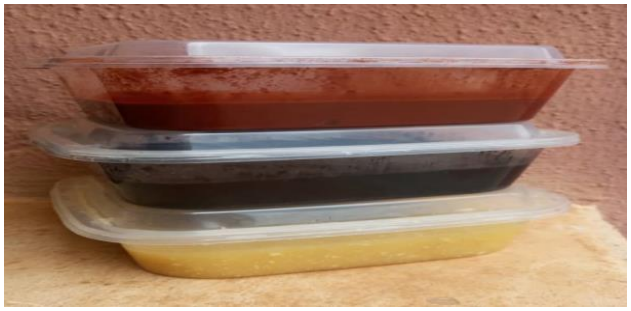

Figure 3b: Acyclic formulations

Figure 3c: Cream Pigment

Figure 3d: Black Pigment

Finish formulations and its Application on the prepared leather substrate

A spray gun was used to apply the finish on the prepared leathers at room temperature (specify the temperature) under a fume hood. The finish formulations were applied on the retanned leathers three times in order to obtain adequate cover and were allowed to dry at normal temperature, and after which it was plated using a hot electric pressing iron

Figure 4a: Sample A1

Figure 4b: Sample A2

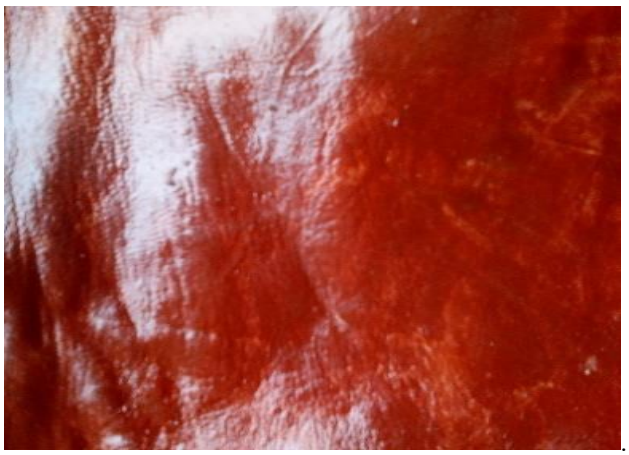

Figure 4c: Sample A3

Water Vapour Permeability Test

This test is used to determine the surface coat's effect on the leathers' porosity. Weighed coated leather samples were placed inside thermo-stated sample holders containing 20 cm^3 of water in a water vapour permeabilimeter (Muver Model 5011) for 1 hr. The test sample capsules and the leathers were weighed again to take the difference per IUP 15. Water vapour permeability (Pwv) is calculated as: Pwv ($mg/cm^2/hr$) = 7640 M/d^2xt, where, M= mass gain between weighings in milligrams; d^2 = area of diameter of sample in cm^2, t = time in minutes between first and second weighing(Keyong *et al.,* 2023).

Figure 5: Watber vapour Permeability test machine

Lastometer Test

Circular samples of the coated and uncoated leathers were cut and placed on an electronic lastometer (Muver, Model 5077-ET), respectively, and tested for distension and grain burst strength per official methods (SLTC, 1996). The forces (kg) and displacement (mm) at burst were obtained from the corresponding digital print-out. Distension as a good index of film strength is a measure of the extent to which the film will extend before film breaks. The film strength in kg/mm is established as the product of the force at break per unit area of the net distensions, i.e., grain film strength (Kg/mm) = Force k/Distension (mm) (Keyong *et al.,* 2023).

Figure 6a: Lastometer Machine

Figure 6b: Tested Samples

Shore A (°) hardness test

The grain side of the leather was placed on the Shore A Hardness Tester/Durometer (Muver Model: 5019/5023-1/5023-A) to measure the degree of hardness.

Wet Rub Fastness Test

A rectangular piece of the leather samples was cut, and for each track 20 mm wide. The grain side of the leather to be tested was rubbed with pieces of standard wool felt under pressure with a given number of forward and backward motions. The SATRA machine was used, the samples were examined after 32, 64, 128, 256, 512, and 1024 revolutions of the dry and wetted pad and given scores of 0, 1, ½, 2,2/3, 3, ¾, 4, 4/5, and 5 when compared with a standard grey scale. A score of 1 is given to a sample with very poor resistance to rubbing effect, while a score of 5 is ascribed to a sample with excellent resistance to rubbing effect. 0 is applicable to samples that were damaged due to the rubbing effect.

Figure 7a: Tested Samples

Figure.7b: Rub Fastness Machine

Figure7c: Grey Scale

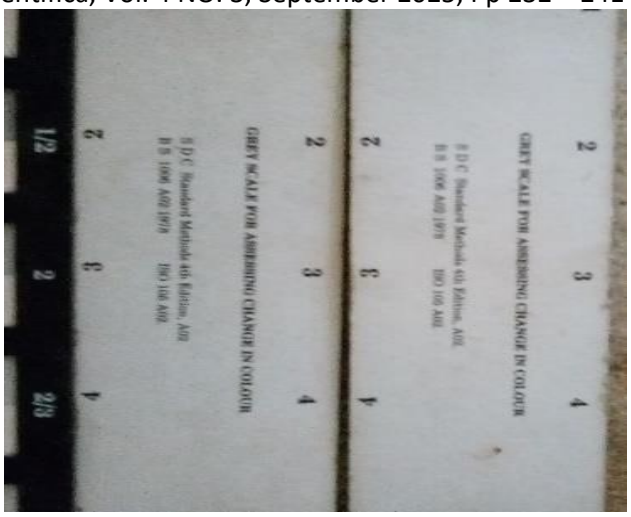

Figure7d: Grey Scale

Figure7e: Felt

RESULTS AND DISCUSSION

3.1 Measurement of Viscosity of the Resin Binder

Viscosity is an integral property of a fluid that offers resistance to flow. It is due to the internal friction of molecules and mainly depends on the nature and temperature of the liquid/solution. Results of solution viscometry of the polymer are presented in Table 8 below. The elution time measured in seconds of the varied concentrations was transformed into relative viscosity, specific viscosity, and reduced viscosity. The plot (Fig. 8.1) of reduced viscosity against concentration was extrapolated to the intercept, which correlates with the intrinsic viscosity of the polymer. The intrinsic viscosity was found to be 227 dL/g, and the calculated average molecular weight of the polymer was 4.03×10^5.

From the result of measurement of the viscosity of the resin binder obtained and presented in Table 8, in this study, the elusion time measured in seconds of varied concentration was transformed into relative (jrel) specific viscosity (jsp) and reduced viscosity (Jred (dL/g)). The plot of the reduced viscosity (Jred dl/g) against the concentration is (227 dl/g). The intrinsic viscosity is used to determine the molecular weight of the acrylic resin binder using the famous Huggins equation (J)= km^a. The calculated viscosity average molecular weight is 403,000;

the calculation details are shown in Figure 8.1. This result shows that the resin binder is a high molecular weight polymer suitable for use in leather finishing. It has been reported that for material to qualify as a binder in leather finishing, the material most have an average molecular weight of 300,000 to 1,000,000. Permeability to water vapour is paramount in garment leather, upper leather, and general clothing articles. It is these properties that permit the escape of perspiration from the body and thus contribute to the comfort of the wearer

Table 8: Measurement of Viscosity of the Resin Binder

Dilution factor Conc. (cm^3)	(g/dl)	Time (sec)	Relative viscosity (Hrel)	Specific viscosity ηsp	Reduced viscosity ηred (dl/g)
+5	0.040	155	131.6	5.62	140.5
+10	0.020	92	68.6	2.93	146.5
+15	0.013	74	50.6	2.16	166.2
+20	0.010	60	57.6	2.46	246.0

t_0 (secs) = 23.42

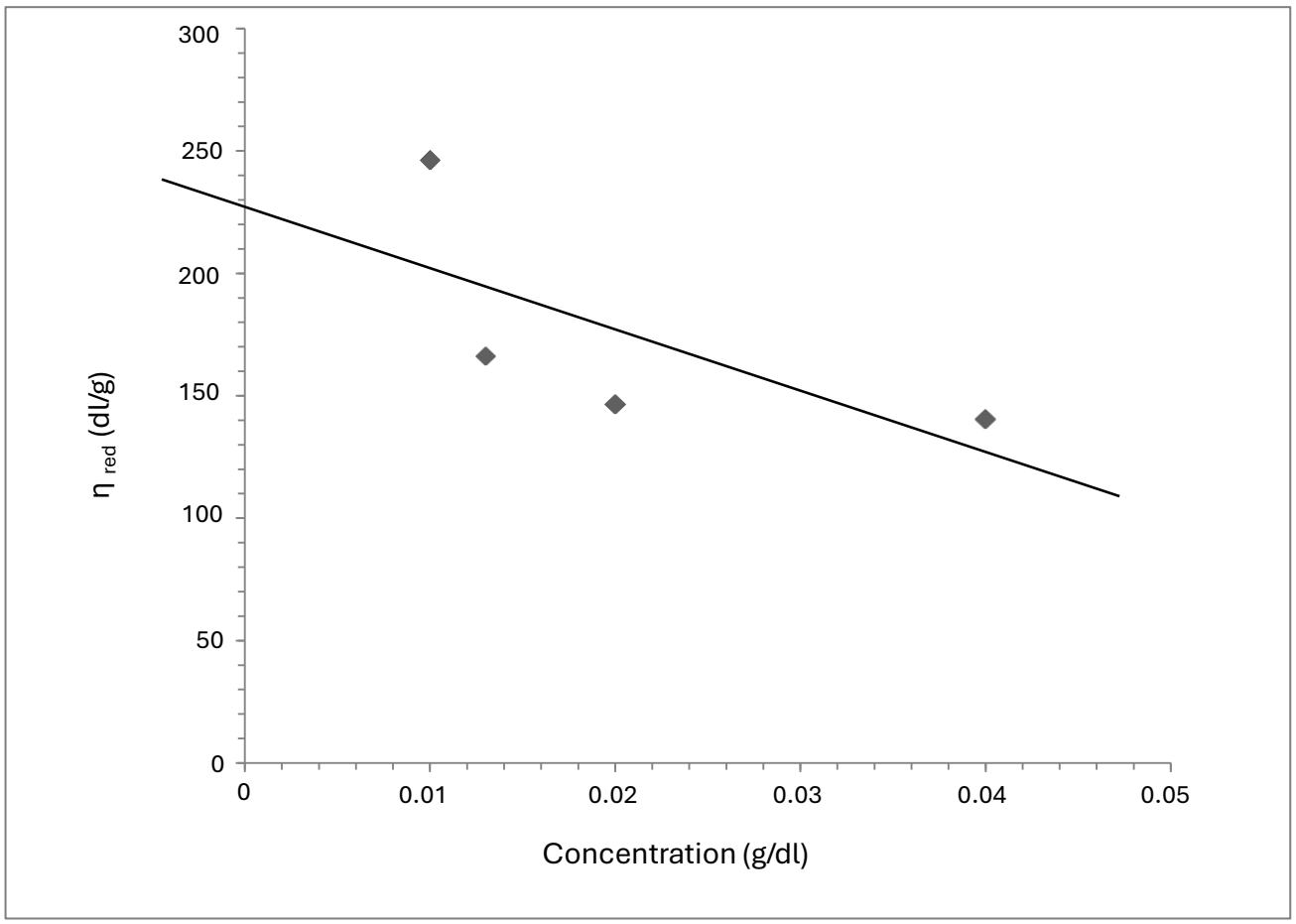

Figure 8.1: Plot of Reduced viscosity versus Concentration

3.2 Physical characteristics testing of finished leathers

Water vapour permeability

Water vapour permeability tests on the coated and uncoated acrylic resin finished leathers were conducted, and the results are shown in Table 9

Permeability to water vapour: The permeability to water vapour is a major importance property in garment leather, upper leather and articles of clothing in general, it's also a property which permits escape of perspiration from the body and thus contributes to the comfort of wearer as presented in Table 9 which shows the effect of acrylic finish on the water vapour permeability of the original retanned leathers. The results also tried to demonstrate the effect of variations in the quality of the acrylic offer in the formulations and their impact on the water vapour permeability of the finished leathers. First, it is worth pointing out that the water vapour permeability of leathers, especially for upper shoes, determines the level of foot conformity obtainable when shod. For this particular test, the general average of this result is surprisingly high. This is partly because the quantity of the finish applied is not large enough to obtain adequate cover, or the pores were probably not properly closed (contribution from retannage) (Mamza and Folaranmi,2023). However, the permeability of uncoded samples was significantly reduced by the application of the finished formulation when compared of that of the coated leather (Figure 10.1.) sample A1, which has the lowest quantity of the resin binder in the formulation, expected

the least permeable with the water vapour permeability of 137.4288 ($gcm^{-3}h^{-1}$) and sample A3 with the 138.1133 ($gcm^{-3}h^{-1}$) was the most permeable. For coated samples, increasing the amount of acrylic offer in the formulations produced a corresponding increase in water vapour permeability of the finish leathers. The uncoated samples are generally expected to produce much larger water vapour permeability of the originally returned when compared with their finished counterpart

Table 9: Effect of acrylic dispersion on water vapour permeability of leather

Sample	WEIGTH			WATER VAPOUR PERMEABILITY ($gcm^{-3}h^{-1}$)					
Coated	0	1	2	3	1	2	3	**Average**	**S.DEV**
A1	137.4515	137.4288	137.4035	137.3960	42.501	47.369	14.042	34.638	14.6981
A2	138.1358	138.1133	138.0896	138.0833	42.127	44.561	11.608	32.765	14.9932
A3	138.4492	138.3918	138.2583	138.2014	107.470	249.953	106.534	154.652	67.386
A	136.2006	135.9133	135.3578	134.6897	537.913	1040.065	1254.256	944.078	300.2189
Uncoated									
B1	138.6242	137.8000	136.5674	136.0698	1543.153	2307.802	931.568	1594.304	562.9668
B2	139.8710	139.6080	139.0865	138.5405	491.105	977.717	1022.278	830.367	240.5829
B3	137.0437	136.2193	135.0840	134.6876	1543.527	2125.627	742.181	1470.445	567.1484
B	135.3739	135.0696	134.5470	134.0622	569.742	978.466	907.693	817.634	178.3489

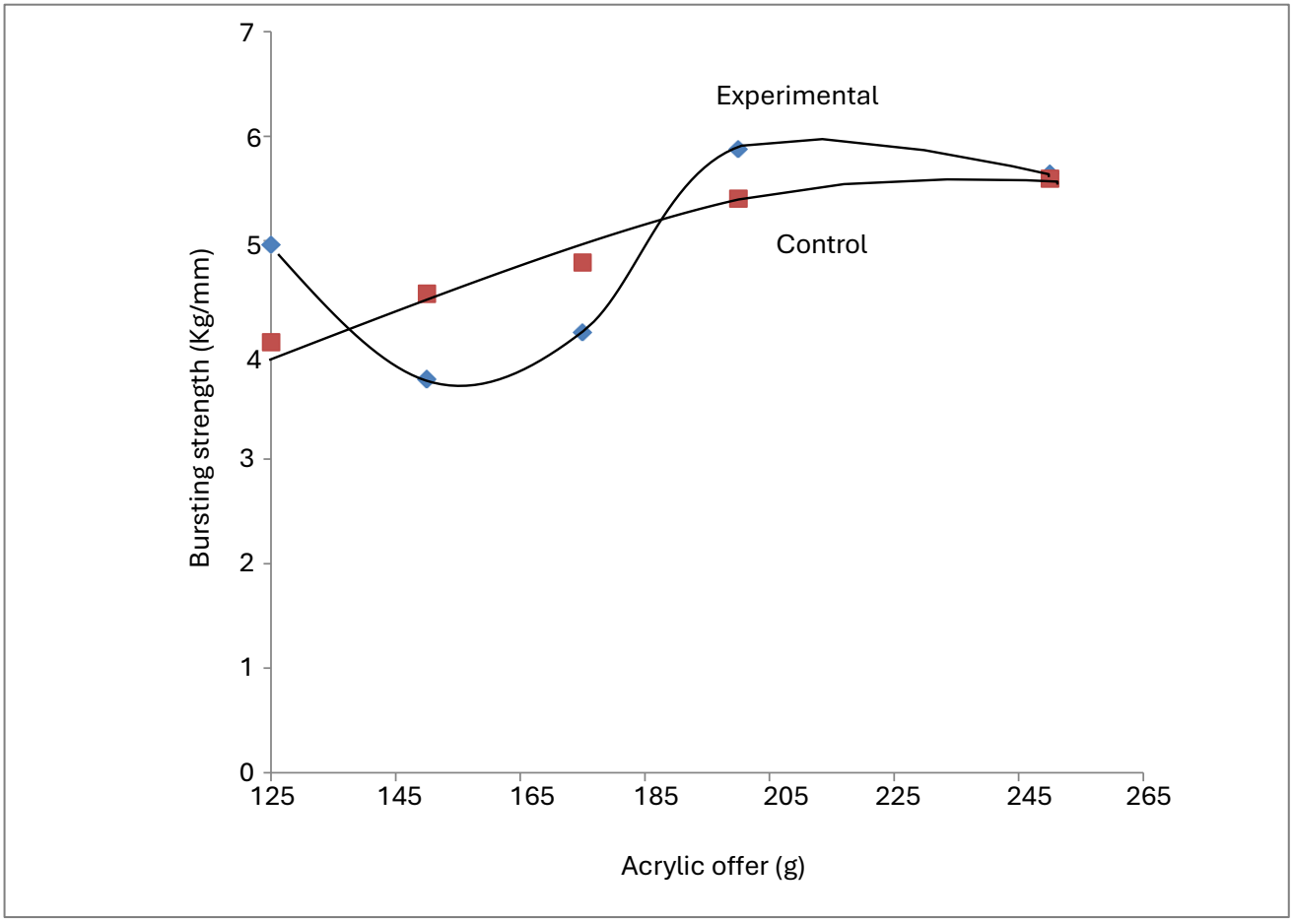

Figure 10.1: Bursting Strength of Finished (Experimental) and Unfinished (Control) Leathers

Lastometer Tests on Leather Samples

Lastometer Tests on the coated and uncoated acrylic resin finished leathers were conducted and the results are shown in Table 10.

Lactometer: the result of the force by displacement of both coated and uncoated samples is shown in Table 10 with the displacement (MM) coated acrylic binders are higher than that of uncoated acrylic binders i.e., the coated samples A1, A2, and A3 have higher displacement than uncoated samples B1, B2 and B3. And also there is an increase in the displacement of coated samples from A1, A2, and A3 (11.02 mm, 11.28 mm, and 12.77 mm) and there is a decrease in the uncoated samples from B1 to B2 (10.06 mm and 10.04 mm) but B3 s higher than B1 and B2.

Shore A^{o} hardness of finished leather and melting point of binder

Film hardness of the acrylic resin-coated and uncoated leathers and the melting point of the binder were carried out on the acrylic resin-coated leathers, and the results are presented in Table 11 below:

Table 10: Lastometer Tests on Leather Samples

Sample	Force (kg) (Maximum)	Displacement (mm) (Maximum Value)
Coated		
A1	36.67	11.02
A2	37.35	11.28
A3	36.67	12.77
A	38.53	11.69
Uncoated		
B1	31.11	10.06
B2	29.10	10.04
B3	30.74	11.25
B	33.76	11.06

Shore hardness (^{0})

The shore harness of the acrylic coated leathers was determined and the melting points of the resin binder were carried out and their results as presented in Table 11. The shore A value range between 60 ^{0} to 77 ^{0} and shore B value range between 70 ^{0} to 80 ^{0}, with the uncoated samples i.e. B1, B2 and B3 (control samples) are harder than the coated sample A1, A2 and A3 (finished samples). The hardness of surface coating film is determined by the glass transition temperature (Tg) which occurs at temperature where polymer changes from hard, brittile, glassy substance to soft and flexible one.

Table 11: Determination of Shore A^o of finish film and melting point of resin binder

SAMPLE ID	DUROMETER READING						
	1	2	4	5	5	MEAN	SDEV
A1	77.0	76.0	76.0	74.0	74.0	75.0	1.1
A2	72.0	75.0	72.0	76.0	75.0	74.0	1.6
A3	66.0	71.0	71.0	69.0	70.0	69.4	1.5
D	74.0	75.0	74.0	77.0	75.0	75.0	0.8
B1	79.0	79.0	80.0	79.0	78.0	79.0	0.4
B2	78.0	80.0	79.0	75.0	75.0	77.4	1.9
B3	80.0	78.0	75.0	78.0	77.0	77.6	1.3
D	75.0	77.0	73.0	72.0	78.0	75.0	2.0

Melting temperature (^{0}c) 361.72-370

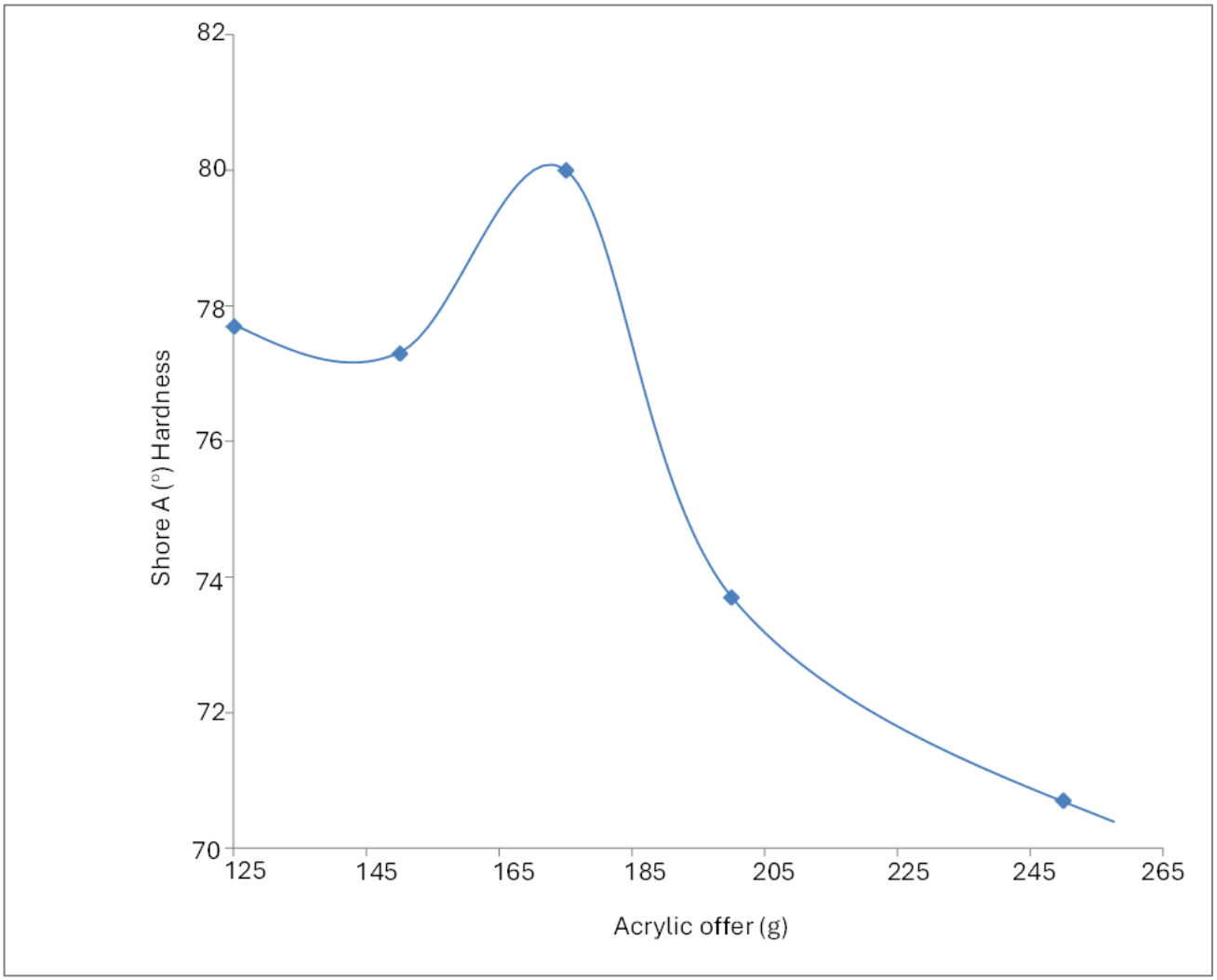

Figure 11.1: Shore A (^{o}) Hardness of Finished Leathers

Wet rub fastness

Wet rub fastness tests were carried out on the acrylic resin-coated leathers, and the results of counts in seconds and the corresponding Grey Scale ratings are presented in Table 12.

Table 12: The effects of finish formulations on wet rub fastness of the coated leather samples

Sample	Acrylic Offer (g)	score	count
A1	125	3	512
A2	150	4	512
A3	175	5	512
A	200	1	512

Acrylic binder: The Acrylic binder is known to have a higher Tg than other binders e.g., polyurethane and polybutadiene (Weijun, 2019), and the harder film should have higher Tg and versa-versal. In leather finishing where the binder is of the soft type with low Tg, during hot plating these film are very tacky and the finish formulation will require a plate release agent, the melting point (™) of a polymer is a temperature at which the crystalline melt and polymer becomes a viscos liquid (Hirose, *et, al.* 2019). The melting point of acrylic was in the range of 346 ^{0} - 371^{0}. Tables 12: Dry rub fastness: sample A1 with the acrylic resin 125 g and score 3 have fire resistance to rubbing effect, sample A2 with acrylic resin 150 g has score 4 and has very good resistance to rubbing, while sample A3 with acrylic resin 175 g have score 5 with excellent resistance rubbing effect. Therefore, between A1, A2, and A3 samples, the A3 sample has excellent resistance to the rubbing effect due to good finish formulations and finishing.

CONCLUSION

The acrylic resin binder is a hard binder with a very high molecular weight, with a melting temperature of 361-370 ^{o}C. This shows that the binder is a high polymer and possesses the properties suitable for application in leather finishing. The effect of the resin formulations on some physical properties of the originally retanned leathers has been studied. It has been observed that the finish had a significant effect on such properties as water vapour permeability, lastometer, Shore A hardness, and wet rub fastness.

The results showed that the finished leathers were better than their unfinished counterpart. Increasing the quantity of the acrylic resin in the formulations also increased the water vapour permeability and wet rub fastness of the finished leathers, except for the lastometer tests and shore A hardness, where there was no specific trend in behaviour. All the formulations showed better and adequate response to all the properties tested compared to the unretanned leathers. However, sample A1 (125 g resin offer) would not be suitable for use in leather finish formulations where wet rub fastness is a priority. Aesthetic properties are very important, but finishes must be durable and standards for upper leathers must include an assessment of finish properties. This has been highlighted in the literature because of the increasing incidence of worn shoe complaints involving lack of finish fastness, and this report has shown that the quantity of components in finish formulations play an important part in determining wet rub resistance.

RECOMMENDATIONS

finishing plays a very important role in any end product; improving the water vapour permeability of leathers is also very important. Studies should be done on how to improve the water-absorbing capacity of some finishing agents, literatures shows that if the water absorbing capacity of finishing agent is too high the wet rubbing resistance may be decreased, so the work should be done to find a balance to improve the water absorbing capacity without decreasing the wet rubbing resistance of leathers.

CONFLICTING INTERESTS

I declared that there is no potential conflict of interest with respect to the research authorship and publication of this article.

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