Innovations

Determination of the Mechanical Properties of Composited High Density Polyethylene, Medium Density Polyethylene and Leather Wastes

[1]Miriam Oguji & [1]Chinedu D. Ani (PhD)
[1]Centre for Environmental Management and Control (CEMAC), University of Nigeria (UNN), Enugu, Nigeria

Abstract: *Plastics do not decompose easily and some of them are not recycled or reused for the initial product. Up cycling presents another alternative to increase their lifecycle. The aim of this work was to determine the alternative uses of mechanical properties of identified and isolated composites from Medium Density Polyethylene (MDPE), High Density Polyethylene (HDPE) and leather wastes. Compression mould was used to prepare the composite samples, while Universal Testing Machine determined the tensile strengths and elongations at break of the samples. The variation containing the three material, but with more of HDPE, had tensile strengths above 25N/mm^2, which is about the standard required for belt (leather) by United Nations Industrial Development Organisation (UNIDO) and Standard Organisation of Nigeria (SON). Artificial Neural Network (ANN), a type of machine learning, was used to predict ratios for certain mechanical properties. These properties were based on requirements by UNIDO's and SON's requirements for insole non-leather and leather bags, respectively. One of the predictions made using neural network had 78.83% and 83.10% accuracies for tensile strength and elongation at break, respectively. More works were recommended to improve on the elongation at break, and to determine other applications for the composites. In addition, it is necessary to study other properties.*

Keywords: *plastics, composites, neural network, mechanical properties, recycle*

Introduction

With the generation of tons of plastic wastes annually and few of them recycled, it is necessary to find alternative applications for them in order to reduce their number as pollutants and possibly, create wealth (Sowunmi, 2019). Plastics (synthetic polymers) are on high demand due to their durability, flexibility and light weight. However, one of these qualities,

durability precisely, is the main reason for their being threat to the environment.
As a natural polymer in mammals, hides contain collagen. Though different animal skins or hides can be used to make leather, the most common is from cattle (Rafferty, 2019). While some out rightly denounce the use of animal skin in the textile industry, others advocate recycling or re-purposing of leather products to reduce the number of animals required to meet the demand of the Industry (Romanova, 2020).
Polyurethane and some other polymers find application as synthetic leather. Most are used in upholstery works. Recycled plastics are not usually re-applied in the food and beverage companies as there are possibilities of them containing toxic substance due to exposure to heat and UV. Polyethylene (PE) products are not regarded as easily recyclable as some other plastics (Goodship, 2007). PE is of different types: the most popular being Low Density Polyethylene (LDPE), Linear Low Density Polyethylene (LLDPE) and High Density Polyethylene (HDPE) (Augustyn, 2019). In addition, there is Medium Density Polyethylene (MDPE).

Most of the work on plastics and leather composites did not relate to the use of MDPE, HDPE and leather combined as composites. This work will compare MDPE/leather composites, HDPE/leather composites and MDPE/HDPE/leather composites to determine their strengths and weaknesses in order to make recommendations for their application. There is no available information on this intent.

The aim of this research was to determine the alternative uses of mechanical properties of identified and isolated composites from plastics and leather wastes. This was achieved using the following objectives:

- Determining the tensile strengths of varied compositions by weight of MDPE/leather, HDPE/leather and MDPE/HDPE/leather composites using universal testing machine.
- Determining the elongations at break of varied compositions by weight of MDPE/leather, HDPE/leather and MDPE/HDPE/leather composites using universal testing machine.
- Producing prototype designs for the mechanical properties using Artificial Neural Network Software for the application of these composites in the leather and other related industries.

The following hypotheses were proposed:

- There is no significant difference between the tensile strengths of the prepared composites (MDPE/leather, HDPE/leather and MDPE/HDPE/leather).

- There is no significant difference between the elongations at break of the prepared composites (MDPE/leather, HDPE/leather and MDPE/HDPE/leather).

Musa et al. (2017) determined the tensile strengths of HDPE and chrome tanned waste. The highest tensile strengths were between 10 to 20%wt of fibre. Awad (2020) in order to improve the mechanical properties of LDPE using nanoclay, observed that 8%wt of nanoclay composite had the highest tensile strength.
Choudhury et al. (2021) prepared composites of HDPE and carbon black. They observed that 2%wt of carbon black had the highest elongation at break. Yousef (2022) applied Analysis of variance (ANOVA test) to show that there were significant differences between the effect of different weights of LLDPE/LDPE mixture (additive) and calcium carbonate filler on mechanical properties of polypropylene/.
Turco et al. (2021) made predictions using chemical composition, temper and Brinell Hardness as input. Yield stress and UTS were the output. Khalefa (2019) attempted to predict tensile strength, hardness and wear loss of Aluminum-Silicon alloys. He obtained output close to the experimental results using % Silicon as input.
With several research works carried out on HDPE and leather composites, there seems to be none that has used the three materials (MDPE, HDPE and Leather) to make a composite. This work determined and compared some of the mechanical properties of the prepared MDPE/HDPE/leather wastes' composites and made recommendation for their application according to standard. There is no available information on this intent.

Materials and Methods

Experimental method was used. Materials included sachets from packaged drinking water (MDPE), broken or damaged plastic jerry can (HDPE) and scraps of leather from Nigerian Institute of Leather and Science Technology, Zaria, Kaduna State. They were dried and characterized. The HDPE and MDPE were cut using scissors. Locally fabricated dry mill ground the leather. Thermogravimetric and differential scanning calorimetric analyses determined their thermal stability and melting points using TA 2950 and TA 2920 equipment, respectively. From these, the matrices were determined. In addition, densities of the materials were determined using methods explained by Smith and Wood (2023).

Weighing and Mixing

The materials were weighed using digital weighing balance (Mettler Instruments Ltd Model no: AE200). According to the formulation tables in the Sampling Technique Subsection, the composites samples were produced by a mixing process involving the introduction of the polymer, while the rolls of the two rolls mill machine (North Bergen, U.S.A Model: 5183) were in counter clockwise motion, to soften for a period of 5 minutes at a temperature of 170°C. Upon achieving a band and bank formation of the polymer on the front roll, the prepared filler was introduced gradually to the bank, cross mixed and allowed to mix for 3 minutes to achieve homogeneity. The composite was sheeted out and labelled accordingly.

Hot Pressing

The composite obtained from the mixing process was placed into a metal mould of dimensions of dumbbell shape of total length 110 mm, gauge length 40 mm, thickness 5mm and grip length of 30 mm at both ends of the sample. It was placed on the hydraulic hot press (Wenzhou Zhiguang Machine Ltd, China Model: 0557)) for shaping at temperature of 160°C and pressure of 2.5 Nmm^{-2} for 5mins. It was then cooled under same pressure 2.5 Nmm^{-2} on a cool press platen for 5 minutes, removed and labelled accordingly.

The same procedure was used for all the samples.

Tensile Strength and Elongation at Break

The tensile strength was carried out in accordance with ASTM D-638 using Universal Testing Machine (D-100KN SN: 190536). Dumbbell shaped samples (Figure 1) were subjected to a tensile force, and tensile strength (Eq. 1), tensile modulus and percentage elongation at break (Eq. 2) for each sample, which had three specimens each, were calculated (average of three) and recorded automatically by the machine.

Tensile Strength (N/mm^2) = $\frac{Force}{Crosssectionalarea} = {F}/{A}$ (Eq. 1) (Callister and Rethwisch, 2020)

Elongation at Break (%) = $\frac{(Instantaneous\ length - Original\ length)}{Original\ length} = {\Delta l}/{li}$ (Eq. 2)

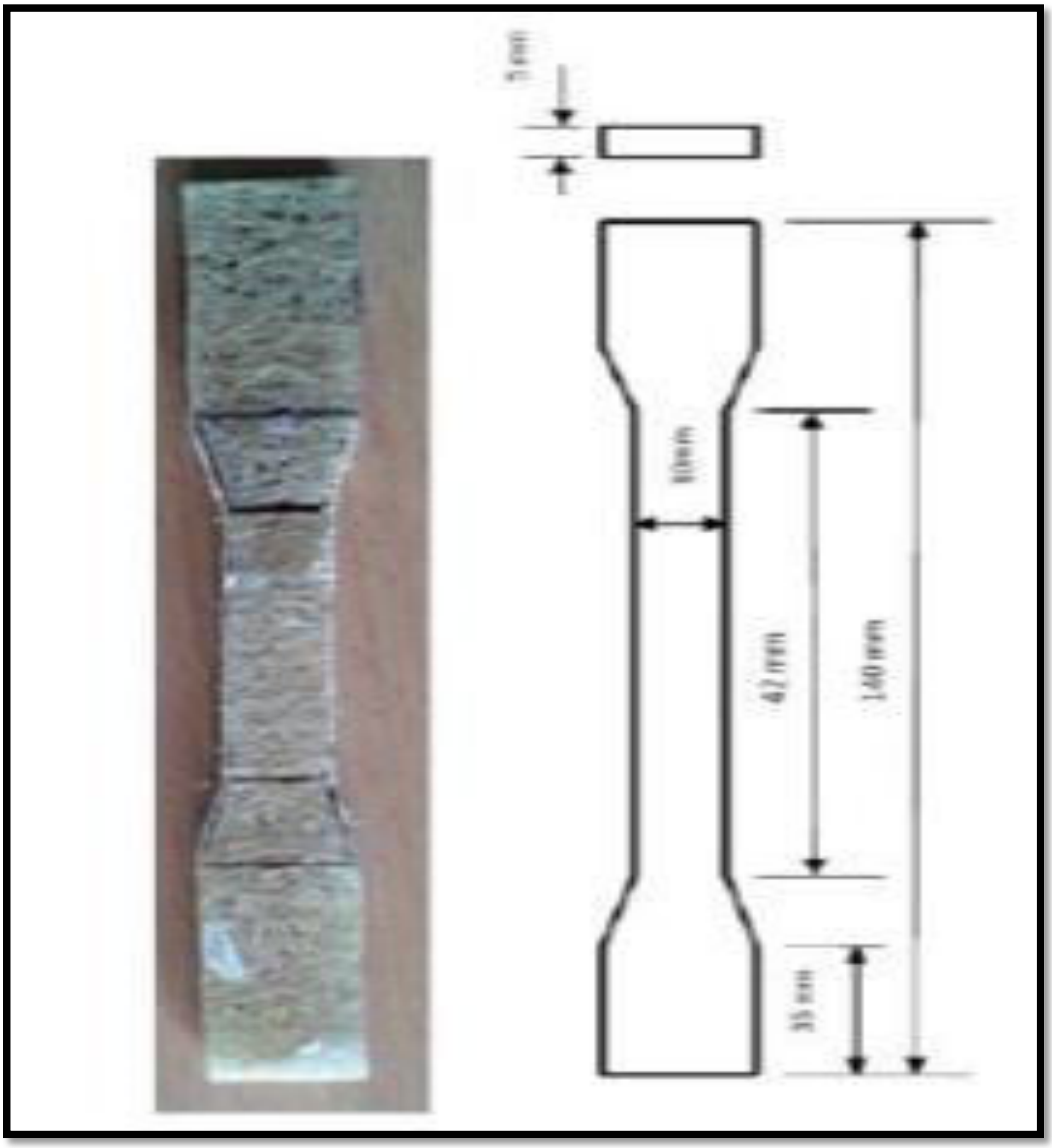

Figure 1: Dumbbell Shaped Sample

Sampling Technique

This was a laboratory work. Based on literature, increase in fillers can lead to brittleness hence, the filler (leather) did not exceed 50% (wt).The ratios or percentages of the compositions were as follows in Tables 1, 2, 3 and 4:

Table 1: MDPE/Leather Composites (1st variation)

S/No	MDPE%wt	LEATHER %wt
MPL 1	50.00	50.00
MPL 2	60.00	40.00
MPL 3	70.00	30.00
MPL 4	80.00	20.00
MPL 5	90.00	10.00

Table 2: HDPE/Leather Composites (2nd variation)

S/No	HDPE%wt	LEATHER %wt
HPL1	50.00	50.00
HPL2	60.00	40.00
HPL3	70.00	30.00
HPL4	80.00	20.00
HPL5	90.00	10.00

Table 3: MDPE/HDPE/Leather Composites (3rd variation)

S/No	MDPE%wt	HDPE%wt	LEATHER %wt
MHL 1	25.00	25.00	50.00
MHL 2	24.00	36.00	40.00
MHL 3	21.00	49.00	30.00
MHL 4	16.00	64.00	20.00
MHL 5	9.00	81.00	10.00

Table 4: HDPE/MDPE/Leather Composites (4th variation)

S/No	HDPE%wt	MDPE%wt	LEATHER %wt
HML 1	25.00	25.00	50.00
HML 2	24.00	36.00	40.00
HML 3	21.00	49.00	30.00
HML 4	16.00	64.00	20.00
HML 5	9.00	81.00	10.00

However, 50g was the total mass used to prepare three specimens for each sample during compounding.

Analysis of Data

One Way Analysis of Variance (ANOVA) was used to determine if there is significant difference between the values of each mechanical property of the four variations. Microsoft Excel served as the package for determining the statistical values.

MathWork MatLab Simulink (2018) artificial neural network package simulated the results of the mechanical properties in order to propose the required compositions of the composites for United Nations Industrial Development Organisation (UNIDO, 1996) and Standard Organisation of Nigeria (SON, 2006) standards for leather products. Tensile strength, elongation at break and hardness were the input while, compositions of materials in grams were the output. The neural network was trained using 17 data sets, one for validation and two for testing. Default 10 hidden neurons were used, while due to the size of the data, Bayesian Regularization was used for the training as recommended by the Package. Re-training of the Network occurred several times until training's and testing's regression values had close values of 0.46 and 0.47, respectively. The predicted compositions were produced and analyzed for their mechanical properties. Percentage accuracies for the predictions were determined using the formula below (Deziel, 2018).

$$\text{Percentage Accuracy} = 100 - \frac{|\text{expected result} - \text{observed result}| \times 100}{\text{expected result}} \qquad \text{(Eq 3)}$$

Results

Figure 2 shows some of the results of thermogravimetric and differential scanning calorimetric of the waste materials (refer to Appendix for the remaining results). It was observed that the matrices (MDPE and HDPE) melting points had a difference less than 7˚C (127.21˚C for MDPE and 132.18˚C for HDPE) and maintained stability up to 170˚C. Major decomposition of the three materials did not occur at that temperature (Figure 2 and Appendix). The densities of the plastics evaluated were in line with that of literature with MDPE 0.941gcm^{-3}, HDPE 0.962gcm^{-3} and leather 0.965gcm^{-3} (Net, 2023). Average tensile strengths and elongations at break were 9.6 Nmm^{-2} and 84.51% for MDPE, and 16.40 Nmm^{-2} and 32.84% for HDPE, respectively.

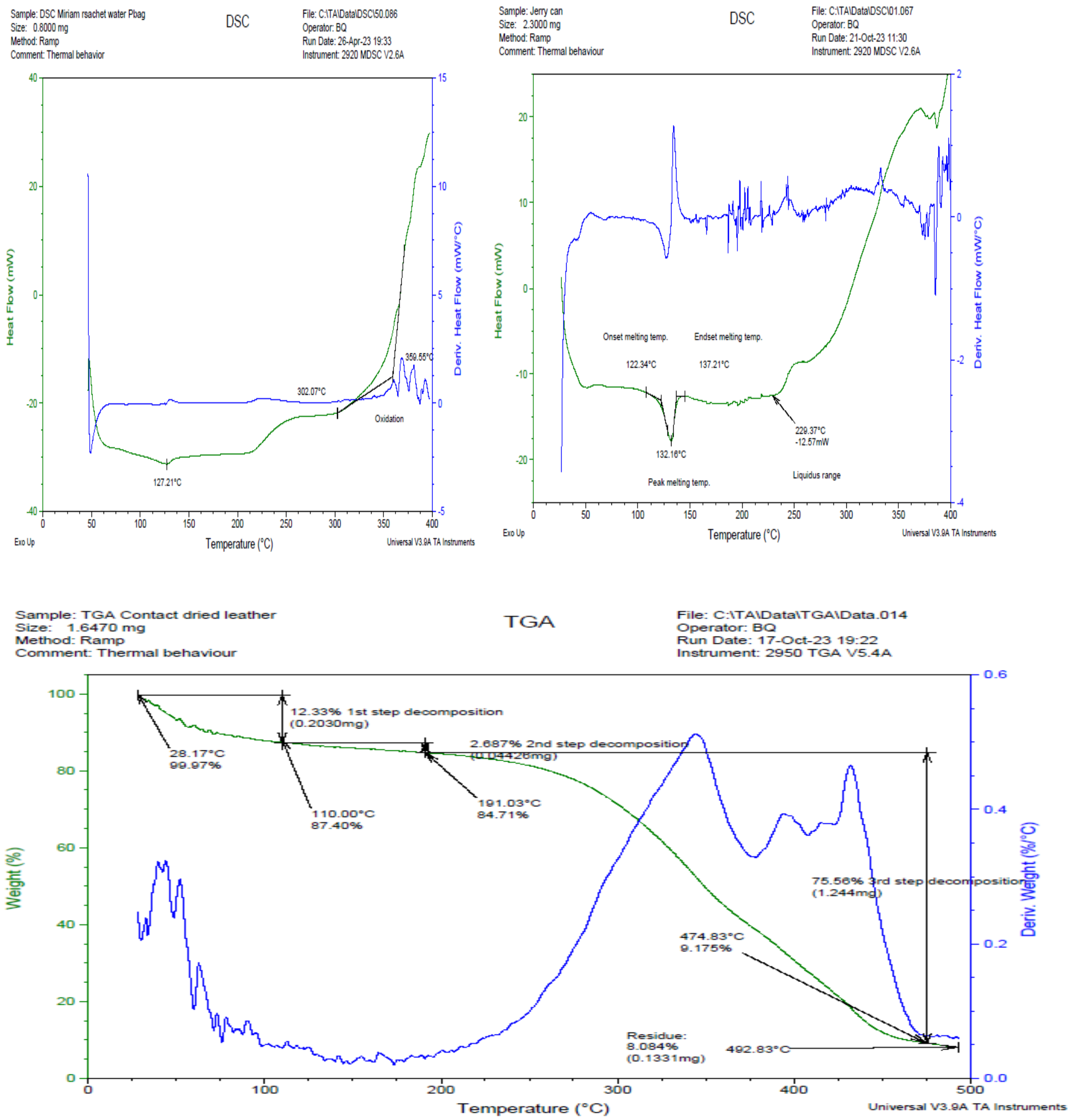

Figure 2: DSC of MDPE (rsachet water bag) and HDPE (jerry can), and TGA of Leather

Tables 5 and 6 present the values of the mechanical properties of the samples of composites analyzed. Table 5 listed the values for tensile strength while, 6 was for elongation at break, respectively.

Table 5: Tensile Strengths of Composites

Composite	Tensile Strength (Nmm^{-2})	Composite	Tensile Strength (Nmm^{-2})	Composite	Tensile Strength (Nmm^{-2})	Composite	Tensile Strength (Nmm^{-2})
MPL 1	17.33	HPL 1	23.33	MHL 1	26.43	HML 1	26.43
MPL 2	27.27	HPL 2	25.20	MHL 2	28.67	HML 2	20.24
MPL 3	22.87	HPL 3	23.41	MHL 3	30.93	HML 3	17.84
MPL 4	24.33	HPL 4	21.20	MHL 4	29.71	HML 4	18.69
MPL 5	18.48	HPL 5	19.47	MHL 5	28.40	HML 5	18.48

Table 6: Elongations at Break of Composites

Composite	Elongation at Break (%)	Composite	Elongation at Break (%)	Composite	Elongation at Break (%)	Composite	Elong at B (%)
MPL 1	9.72	HPL 1	12.10	MHL 1	16.05	HML 1	16.05
MPL 2	10.88	HPL 2	10.82	MHL 2	16.57	HML 2	8.85
MPL 3	9.59	HPL 3	15.93	MHL 3	15.14	HML 3	16.26
MPL 4	11.00	HPL 4	12.20	MHL 4	12.95	HML 4	9.78
MPL 5	15.75	HPL 5	9.72	MHL 5	13.70	HML 5	15.73

For the predictions made by neural network, Appendix displays the ANN predictions or the "ans". The first "ans", was named Sample A, which predicted MDPE, HDPE and leather to be 20.44g, 17.96g and 12.06g, respectively, for composite with tensile strength 20 Nmm^{-2} and elongation at break 15%. While the second "ans", in the same order, showed 5.98g, 25.48g and 17.65g for 25 Nmm^{-2} and 15%. The predicted compositions were produced and their mechanical properties were determined.

Using One Way Analysis of Variance, the following p-values in Table 7 were obtained for Tables 5 and 6

Table 7: P-values of Tensile Strength and Elongation at Break of the Composites

Mechanical Property	P-value
Tensile Strength	0.002447
Elongation at Break	0.214027

Table 8 shows the predictions made using neural network. Percentage accuracies were determined by using equation 3.

Table 8: Analysis of Predictions Made by ANN

	Observed Tensile Strength (N/mm2)	Expected Result (N/mm2)	Accuracy (%)	Observed Elongation at Break (%)	Expected Result (%)	Accuracy (%)
Sample A	24.235	20.00	78.825	12.465	15.00	83.100
Sample B	24.465	25.00	97.860	10.500	15.00	70.000

Test of Hypothesis

- There is no significant difference between the tensile strengths of the prepared composites (MDPE/leather, HDPE/leather and MDPE/HDPE/leather).

Since p-value is less than 0.05, the null hypothesis is rejected. Hence there is significant difference between the tensile strengths of the prepared composites

- There is no significant difference between the elongations at break of the prepared composites (MDPE/leather, HDPE/leather and MDPE/HDPE/leather).

Since p-value is greater than 0.05, the null hypothesis is accepted. Hence, there is no significant difference between the elongations at break of the prepared composites

Discussion

The tensile strengths of MDPE and HDPE increased with the addition of leather for all the ratios used. However, MHL3 had the highest value with 30.93Nmm^{-2}. 50% MDPE and 50% leather yielded the lowest tensile strength.

60% MDPE and 40% leather composition resulted in the highest for the MPL composites. HPL composites exhibited optimum tensile strength of

25.20Nmm^{-2} with 60% HDPE and 40% leather. Their lowest was HPL4 at 21.20Nmm^{-2}. MHL had their lowest at 26.43Nmm^{-2}, which was the highest for HML. The lowest for HML was 17.84Nmm^{-2}.

It was observed that though MDPE's tensile strength was low, its composite with leather yielded higher tensile strength than HDPE's. With the trio in samples, more of HDPE seemed to result in the highest tensile strength.

Elongation at break of the composite samples plummeted, when compared with the initial values of the matrices. The lowest value was 8.85% obtained from HML2 while, the most elongated was 16.57% from MHL2. HML had 16.05% as its highest, but the least for MHL was 12.95%.

HPL and MPL variations peaked at 15.93% and 15.75%, respectively. They both plunged to 9.72% with HPL5 and MPL1.

Predictions made with ANN had percentage accuracies of 78.83% and 97.86% for tensile strength, and for elongation at break, 83.10% and 70.00%. The expected results of the mechanical properties were derived from UNIDO's standard for insole non-leather materials (Appendix), and SON's standard for leather bag. SON's standard for leather bag varies from 20 to 25N/mm^2 (tensile strength) with a maximum of 50% for elongation at break. It is pertinent to note that the ratios or compositions predicted did not sum up to exactly 50g like the input. Sample A was 20.4440g of MDPE, 17.9612g of HDPE and 12.0615g of leather, making a total of 50.4667g. With a total of 49.1111g, Sample B contained 5.9832g of MDPE, 25.4779g of HDPE and 17.6500g of leather (ANN Predictions, Appendix). The percentages are presented in Table 9 below:

Table 9: Percentage Compositions of Samples A and B

Sample	MDPE %	HDPE%	Leather%	Total%
A	40.51	35.59	23.90	100.00
B	12.18	51.88	35.94	100.00

The intention for varying the ratios of the three contents was to capture different compositions and their characteristics as much as possible. This, expectedly, enabled the neural network make more reliable and wider decisions. It appeared a bit odd that an MHL composition, which has higher quantity of HDPE than MDPE, would have tensile strength that is not within the range of 26Nmm^{-2} and 30Nmm^{-2} as seen in Sample B. Hence, the system's indication that Sample B would yield about 25Nmm^{-2} proved that desired outcomes can be predicted or obtained from less likely combinations.

ANN training using Bayesian Regularization showed consistent "ans" (Appendix). The software used, MathWork MatLab, advised that Bayesian Regularization works better with small data than other processes available on it.

Based on UNIDO's standard, belt and football leather require tensile strength of 25N/mm^2. SON's range for belt (leather) is from 20 to 27N/mm^2, depending on the method of tanning (Appendix), though, the elongation at break should be up to or above 50%. For soles (leather), both organisations expect 20N/mm^2, but elongation at break should be about 30% or 35% for SON. These should be about the same requirement for insole leather. The MHL variation along with MPL2 had tensile strength above 25N/mm^2. HPL variation with the exception of HPL5 had values between 20 and 24.33N/mm^2 like HML2, MPL3 and 4. Elongation at break did not exceed 16.57%.

Conclusion

In terms of tensile strength, some of the composites meet the necessary standards. However, their elongations at break seem suitable for leather bag and insole non-leather materials especially as it pertains to SON's standards. This work did not use any additive to obtain any desired outcome but, in order to improve the elongation at break, including plasticizer in the composites is one of the ways mentioned in literature, for achieving this (El-hadi, 2017). Particle sizes were not considered in this work. The use of two-roll mill assisted in mixing the materials.
HML1 and MHL1 shared the same result through all the analyses. The HML variation had more of MDPE in each sample.

During the collection and characterization of the plastic wastes, it was discovered that some of them had the wrong recycling code: like a polypropylene bucket had number 2, which is for HDPE. It was the melting points and densities that enabled the determination of the types of plastics that they were. In addition, due to the small difference (less than 7°C) between the two matrices, their mixtures did not seem to be affected (segregate) while the composites were cooling.

Most plastics used for food and beverage are not recycled to be applied in the same industry. Rather, they are up cycled that is they find application in other areas. This is due to health implications. Other mechanical properties should be studied to determine the suitability of the composites in the leather industry or other applications like tiles.

The results of the mechanical properties of the composites indicate that leather has varying effects on their tensile strength.. Tensile strength, which determines a materials reaction to tension, increased considerably when compared to the binders'. This means some of the composites conform to the tensile strength requirements of leather bag and football. On the other hand, Elongation at break that shows the plasticity tended to decrease. Due to this, it could not meet certain SON's standards for products like leather sole and insole. Further work is encouraged to improve that quality.

ANN percentage accuracies of predicted ratios were not under 70%. Bayesian regularization was used to train the system as it could work with data below 1000 in number. Specific predictions were requested from the neural network, more than once, and it provided the same "ans" for each request.

It is recommended that further studies are carried out to determine the other physical properties of these composites in order to identify their suitability for some products. The purpose of finding applications for these composites is to up cycle them, thereby, increasing their lifecycle.

If they add or have values in other industries, waste collectors and recycling companies will be encouraged to properly remove them from cluttering the environment as they will receive financial gain in exchange for their effort. Most plastics are not easily degraded by nature. This necessitates the need to find alternative use for them.

Using them as composites to make other useful products could be another way of removing them from landfills. It may be a way of implementing one of the five "R"s of waste management.

Recommendations

- Effects of additives on MDPE, HDPE and leather composites should be studied.
- Studies to determine the end-of-life or shelf life of the composites need to be carried out.
- Other physical properties should be studied to determine possible applications of MDPE, HDPE and leather composites.
- Plastic product manufacturers should label their products appropriately to enable recyclers sort them.
- Use of ANN or artificial intelligence could lead to achieving desired outcomes in recycling.

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Appendix

Table 10: Acceptable Quality Standards in the Leather and Footwear Industry (UNIDO)

UNIDO Guidelines	Belt Leather (Vegetable)	Belt Leather	Football Leather	Safety Gloves
Tensile Strength N/mm^2	25	25	30	15
UNIDO Guidelines	Sole Leather Fexible	Sole Leather	Insole Leather	Insole Material (non Leather)
Tensile Strength N/mm^2	20	20	20	3-12
Elongation at Break (%)				15-40

Table 11: SON's Standards for Leather Products

Product	Tensile Strength (N/mm^2)	Elongation at Break (%)
Bag Leather	20.0-27.5 (min)	50 to 75 (max)
Belt Leather	20.0 (min)	50 to 75 (min)
Sole Leather	20 (max)	30-35
Insole Leather	20 (min)	35 (min)

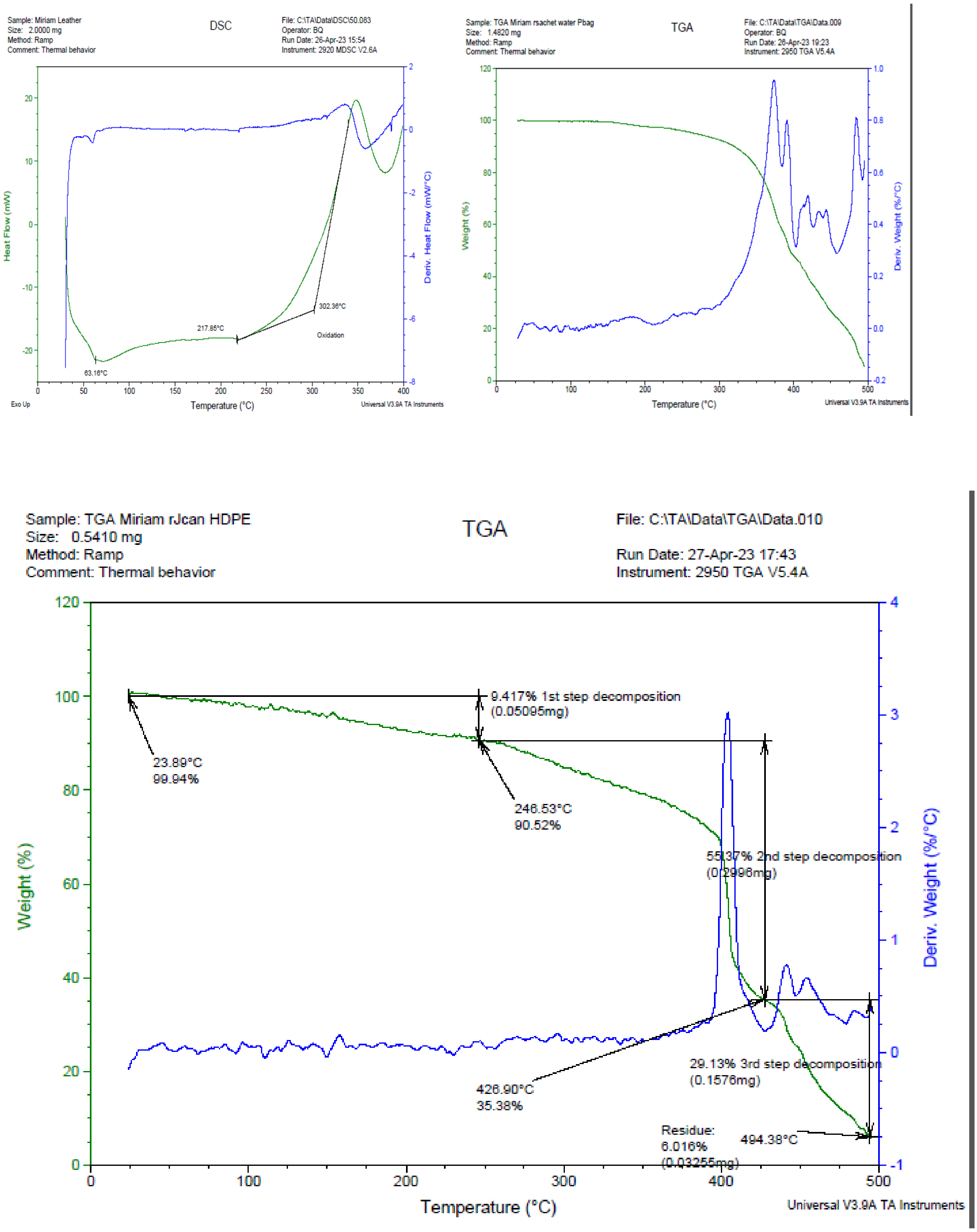

Figure 3: DSC of leather, TGA of MDPE (rsachet) and HDPE (rjcan.)

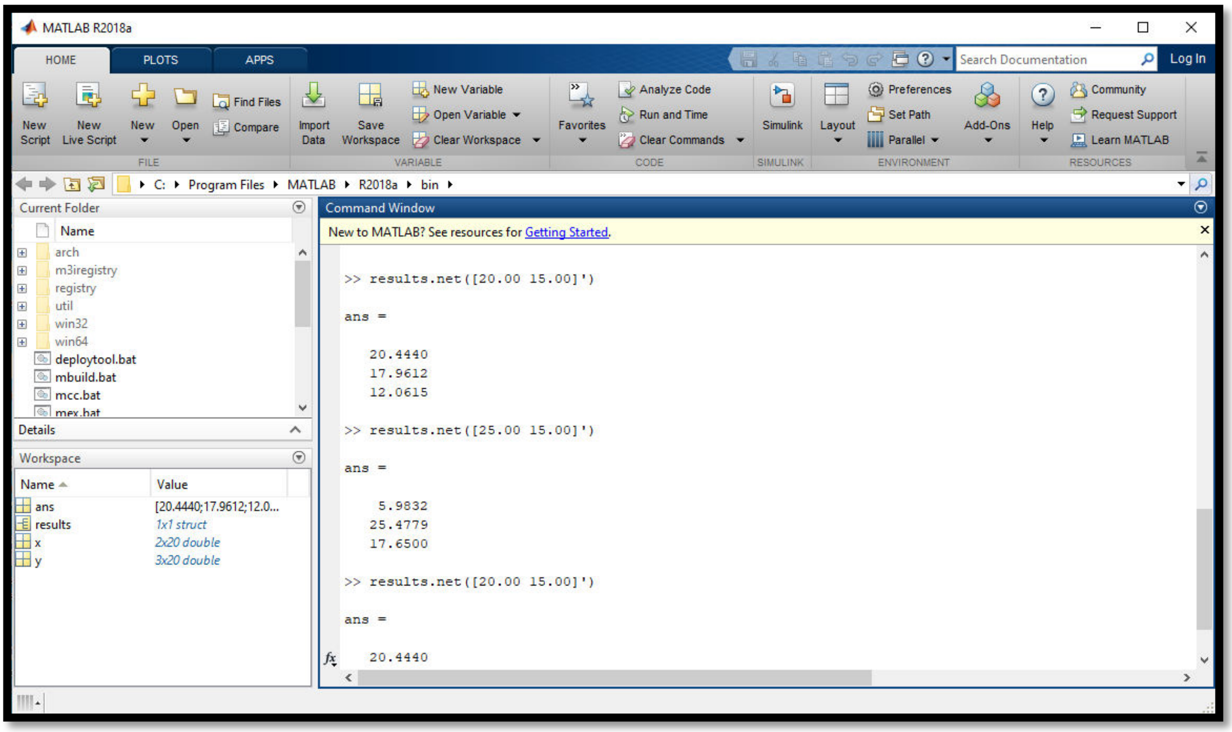

Figure 4: ANN Prediction