

Mechanical properties of laminate materials based on polylactic acid and polyvinyl chloride meshes as reinforcement

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Abstract

The 3D printing parameters are known to have a significant impact on manufactured parts, and the layered morphology of these parts makes mechanical design analysis for engineering applications difficult. In this work, the tensile strengths and microhardness of 3D printed polylactic acid (PLA) specimens with different orientations and numbers of individual layers of mesh material (polyvinyl chloride – PVC) were investigated as a laminate composite. Composite specimens were obtained using 3D printing via fused deposition modelling (FDM). Moreover, the influence of printing parameters (i.e. infill density and layer height) and the number and orientation of reinforced meshes on the mechanical response was investigated. Fracture strength of PLA/PVC laminate composites ranges from 31.30 MPa (3 PVC mesh layers; mesh height position: 25 % | 50 % | 75 %; infill density: 60 %; PVC mesh orientation: 90° | 45° | 90°; layer height: 0.2 mm) to 18.62 MPa (without PVC mesh; infill density: 30 %; layer height: 0.1 mm) demonstrating a significant impact of the number of the PVC mesh layers, infill density of PLA and layer height on the final mechanical parameters of printing PLA/PVC elements. The surface hardness at the micro load level showed that the number of reinforcement layers affects the microhardness value, as well as material filling and mesh orientation. The specimen with the following parameters gave the best results: layer height: 0.2 mm; 3 PVC mesh layers; infill density: 60 %; PVC mesh orientation: 90° | 45° | 90°. The average hardness values for one layer and three layers of mesh were in accordance with tensile test results.

1. Introduction

The additive manufacturing (AM) process, also referred to as 3D printing, is being used these days extensively not just for prototyping of industrial parts but also to create functioning components for numerous purposes in everyday life using a variety of materials, including metals, composite materials, polymers and ceramics [1,2]. Fused deposition modelling (FDM) is extensively employed in industrial applications, including automotive and aerospace with the application of sandwich structures with excellent mechanical properties [3].

In biomedicine, PLA was used as scaffold architecture for tissue engineering applications [4]. PLA is commonly blended with ceramics (hydroxyapatite and beta-tricalcium phosphate) to achieve excellent properties of a 3D printed scaffold [5]. Low-cost compostable polyesters, bio-absorbable and bio-degradable thermoplastic polymers such as PLA have numerous benefits for biomedical applications for fabricating implant devices [6], as bone repair materials [7] or to influence effectiveness activity under changing alkaline phosphatase in cells in combination with hydroxyapatite [8].

Composite materials manufactured by FDM technology require a certain quality for use in biomedicine. Those qualities are monitored based



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on the following parameters of the composite material: composite type, i.e. the ratio of primary/secondary material in composite, infill, top-bottom pattern, layer thickness, gap between molten lines, pore size, porosity, etc. [9].

Excellent nanofiltration and separation with excellent physical performances in membrane applications are done using PLA material. The TF-629C dip-coated PLA as a synthetic porous membrane is an example of a combination material for this purpose [10].

PLA filaments with particle reinforcement are a new trend in the development of these composites. The physical, mechanical and degradation properties of the PLA matrix with magnesium (Mg) particles were investigated recently [11,12]. The study showed that the addition of Mg particles in PLA promotes the entanglement of polymer chains, lowers the mobility of PLA molecular chains and raises the peak temperature (glass transition temperature) [11]. The presence of Mg particles in PLA improved the mechanical properties (tensile and compressive strength) and increased the degradation rate of 3D printed specimens. Due to increased surface roughness and the inclusion of Mg particles, the PLA/Mg composites demonstrated higher hydrophilicity [11]. Alumina, silica and other fillers are examples of several ceramic powders that are frequently utilised as reinforcement in polymeric matrices to enhance their mechanical, morphological and thermal properties.

Studying 3D printing parameters shows that when the filling orientation or nozzle motion pattern direction changes (from 0 to 90 degrees) from the tensile direction, the tensile strength decreases in the PLA specimen and all composite systems [12].

By adding high-elongation natural fibres for enhanced impact or fibre for increased stiffness, we may create a composite with better mechanical qualities by learning from nature about the functions of different types of fibre in plants, such as microfibrillated cellulose [13], jute, cellulose pulp from wood and abaca [14]. Additive technologies have great potential applications in the textile industry, especially in self-cleaning, 3D printing on textile substrates, 3D printing of textiles or combination techniques [15]. Insertion of different fabrics, scaffolds, and composites reduces cracks in the infill; that is, the complete structure is strengthened, thereby reducing the possibility of moisture absorption [16].

The research aims to use FDM technology with the additional mesh insertion made of

another type of material. This experiment should show how significant mesh insertion (with one or more layers) is for improving the specimen's specific mechanical characteristics. The procedure is increasingly applied in the textile industry (setting decorations, stickers, buttons, etc) [17]. In the textile industry fashion items can be made on demand while clothing becomes readily available. This way, only the necessary material is used and no waste is created [18]. Different combinations of textile mesh with PLA-printed matrices were investigated and their relationship in terms of mechanical properties was also investigated [19].

A review of the literature revealed that while it has been possible to successfully print clear PLA and their composite in three dimensions, relatively little research has been done about choosing polyvinylchloride (PVC) reinforcement in PLA base matrix for potential applications. The idea of this investigation is based on a similar combination of materials, i.e. polylactic acid (PLA) reinforced with polyvinylchloride (PVC) [20,21], but without wood dust and magnetite. Differences are that the composite is in the form of a laminate (printed filaments of PLA/commercial PVC meshes), and not in the form of a blend of PLA/PVC material that was then printed [20,21].

In this way, the original properties of both materials are preserved and the mechanics of the composite is investigated. On the other hand, the poor dispersion that is characteristic of polymer blends is avoided, but the problem of inserting commercial PVC can lead to debonding and poor interfacial adhesion strength between two polymers in a laminate composite.

2. Experimental

2.1 FDM process and materials

The fused deposition modelling (FDM) process was used to produce specimens. Unfortunately, compared to other additive processes, the FDM process produces prototypes of lower quality in terms of supplementary structure, final dimensions and surface quality [22].

The specimens manufacturing process begins with a 3D CAD model, which is realised using one of the CAD software. The model is converted to an *.stl file in a specialised open-source software Creality Slicer V.4.8.2, in which print parameters are adjusted. After that, a G-code with the

extension *gcod is generated, which is essential for starting the printer [23,24]. The pattern is formed by applying thermoplastic filament layer by layer. A Creality Ender-6 3D printer was used to create the specimens, with a working volume of 250 × 250 × 400 mm, a printing accuracy of 0.1 mm and a printing speed of 150 mm/s.

All specimens are made of polylactic acid white filament (PLA), with a diameter of 1.75 mm (Gembird, Netherlands). According to the manufacturer's specification, some of the filament's essential properties are shown in Table 1.

Table 1. Properties of the commercially available PLA filament [25]

Property	Value
Diameter, mm	1.75
Density, g/cm ³	1.25
Melt temperature, °C	190 – 220
Heat deflection temperature, °C	50 – 60
Yield strength, MPa	45
Impact strength, kJ/m ²	5

PLA is a thermoplastic polymer that has good strength [22]. It is widely used in the development of various prototypes. PLA is a material that has better mechanical properties than other plastic materials [26]. The filament melts through the heat nozzle and forms layers on the heated surface through the extruder's nozzle. To achieve high-quality specimen manufacturing, it is necessary to adjust the parameters of the 3D printer [27]. The

selected parameters for manufacturing specimens are given in Table 2.

Table 2. Parameters for manufacturing specimens

Parameter	Value
Layer height, mm	0.1 or 0.2
Wall line contour	2
Top/bottom layers	2
Infill density, %	30 or 60
Infill pattern	lines
Infill line directions, °	– 45/45
Printing temperature, °C	200
Build plate temperature, °C	60
Print speed, mm/s	60
Build plate adhesion type	skirt

2.2 Specimens preparation

A total of 24 specimens were made with dimensions according to the EN ISO 527-2 standard [28]. The specimens were made in a standard form with a thickness of 4 mm and an initial distance between the grippers of 80 mm. This specimen type was chosen due to savings in material and a shorter 3D printing time [29].

The specimens were divided into three series with 8 specimens each. The first specimen series was not reinforced and was used as a referent. The other groups (reinforced) had a sandwich structure. This new structure represents a laminated composite formed by two stiff facings at while the lightweight cores were commercial PVC

Table 3. Specimen types and parameters for their realisation

Series	PLA specimen					PVC mesh		
	type	layer height, mm	infill density, %	designation	amount	height position, %	orientation, °	number of layers
1	unreinforced	0.1	30	NO – 1.1/1.2	2	–	–	–
			60	NO – 2.1/2.2	2			
		0.2	30	NO – 3.1/3.2	2			
			60	NO – 4.1/4.2	2			
2	reinforced 1	0.1	30	1L – 1.1/1.2	2	50	90	1
			60	1L – 2.1/2.2	2			
		0.2	30	1L – 3.1/3.2	2			
			60	1L – 4.1/4.2	2			
3	reinforced 2	0.1	30	3L – 1.1/1.2	2	25 50 75	90 45 90	3
			60	3L – 2.1/2.2	2			
		0.2	30	3L – 3.1/3.2	2			
			60	3L – 4.1/4.2	2			

mesh. The lightweight PVC core can be seen as a reinforcement layer. The number of reinforced layers within the sandwich structure was 1 and 3. The specimen's designation, corresponding to the series type was reported in Table 3.

The printing parameters and the properties of the mentioned polymers greatly influence the formation of a solid bond, i.e. composite. Figure 1 shows the specimens immediately after printing.

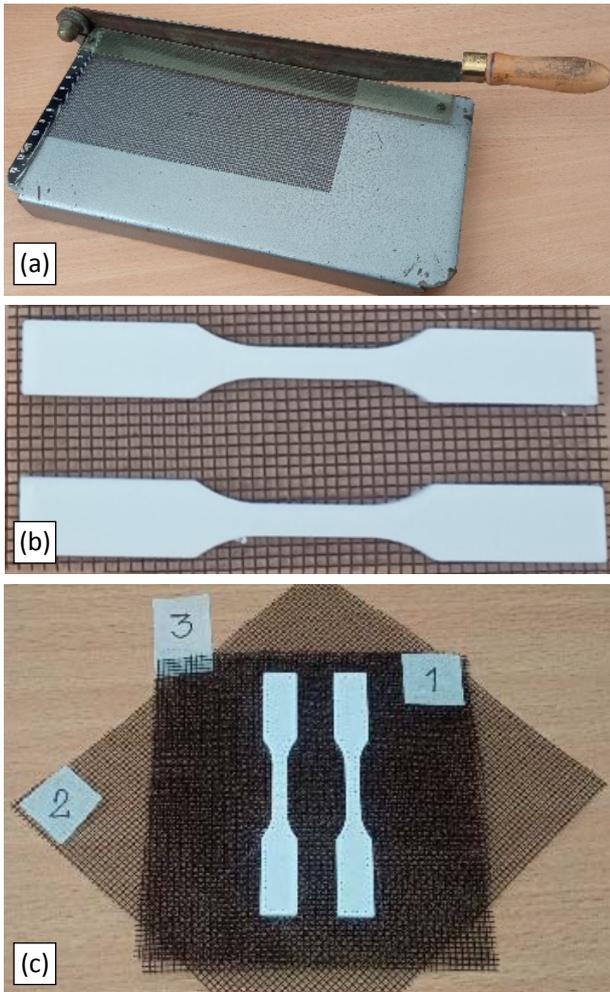


Figure 1. Specimens preparation: (a) mesh, (b) mesh in one layer and (c) mesh in three layers

For one-layer composite, at 50 % of the realised specimen height, the 3D printer is paused. The prepared PVC mesh is placed over the specimens and strengthened with tape. Also, adhesive coating for 3D printing (Devil Design, Poland) is applied to continue printing and assembling a joint between the printed layer and the placed mesh. The same procedure was repeated for the specimens with three layers of mesh (at 25, 50 and 75 %). Figure 2 shows the shape of a solid bond instantly after the 3D printer starts (at around 76 % of the specimen height). The meshes are installed very quickly without interfering with the printing process.

Therefore, it can be concluded that PLA bonds well to the PVC mesh, which achieves a quality print.



Figure 2. Bonding process

After 3D printing, the surplus PLA material and PVC mesh are removed (Fig. 3).

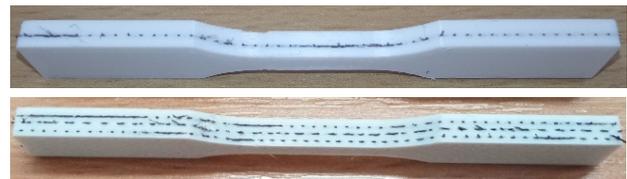


Figure 3. Specimens with PVC mesh (one and three layers) ready for testing

2.3 Characterisation of PLA/PVC laminate composite

Tensile tests were performed on the prepared 24 specimens at room temperature of approximately 20 °C, according to the EN ISO 527-2 standard [28]. The test was carried out on the Instron 1122 universal testing device, which has a maximum load capacity of up to 5 kN. The specimens were clamped with wedge grips. The length of the specimens between the clamps was 33 mm. The test speed is set to 50 mm/min. Load, displacement and time were registered during the experiment via the TRC PRO acquisition system connected to the tensile machine. Subsequently, maximum tensile strength, strain at break and modulus of elasticity were calculated based on the obtained (recorded) data.

To determine the microhardness of the PLA/PVC laminate composite materials, the Vickers method [30] was used on the TIME TH710 micro Vickers hardness tester with an applied normal load of 0.49 N and a dwell time of 15 s. Five indents were made on each specimen on the top

surface of the composite and the diagonals were recorded. The mean value of the Vickers hardness number (HV) is usually used to estimate the microhardness of the composite according to standard ASTM E384 [31]. In order to compare with previous microhardness results, the results are expressed in MPa [30].

3. Results and discussion

The appearance of the specimens after the tensile test is shown in Figure 4 and the tensile test results are summarised in Table 4, giving the average values and standard deviations (*SD*) of maximum force (F_{max}), fracture strength (σ), strain at break (ϵ) and modulus of elasticity (E).

It is interesting to observe and compare stress-strain dependencies for the unreinforced and reinforced samples produced in different directions of 3D printing as shown in Figures 5 to 8 (representative curves).

As it may be observed, the maximum fracture strength is increased for the reinforced samples, as expected, and the lowest values were noted for unreinforced PLA specimens designated as NO – 1. For the specimens with 1 and 3 layers, in most cases, higher fracture strength was noted for the samples with one layer of the reinforcing PVC mesh, except for the specimens designated as 1L – 4. The reason for this may be in the overall weaker structure of this mesh orientation.

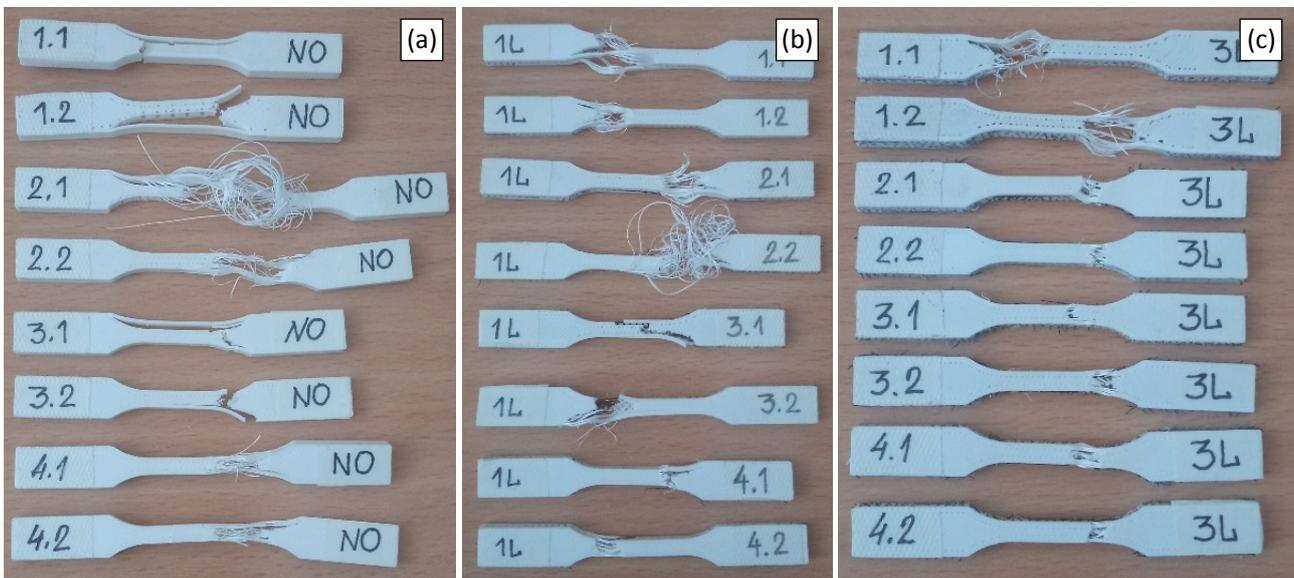


Figure 4. Specimens after tensile testing: (a) unreinforced (without mesh), (b) reinforced with one layer and (c) reinforced with three layers

Table 4. Tensile test results

Group	Force F_{max} , N	$SD F_{max}$, N	Strength σ , MPa	$SD \sigma$, MPa	Strain ϵ , %	$SD \epsilon$, %	Modulus E , MPa	$SD E$, MPa
NO – 1	323.25	6.01	18.62	0.29	3.75	0.33	699	73
1L – 1	484.00	2.12	28.11	0.12	3.67	0.06	984	33
3L – 1	382.50	45.96	22.11	2.77	4.36	1.12	747	49
NO – 2	459.25	22.27	26.61	1.26	3.99	0.06	930	7
1L – 2	493.17	16.10	29.07	0.73	3.82	0.17	1102	14
3L – 2	498.50	19.80	28.94	1.17	3.94	0.04	1077	184
NO – 3	406.50	6.36	23.55	0.33	4.48	0.23	832	54
1L – 3	445.00	10.69	25.81	0.60	4.24	0.04	758	59
3L – 3	420.25	62.58	24.38	3.63	3.99	0.47	791	66
NO – 4	492.50	3.54	28.55	0.16	4.24	0.02	945	23
1L – 4	531.83	12.47	30.84	0.69	3.82	0.17	1059	77
3L – 4	539.25	0.35	31.30	0.00	4.37	0.35	897	25

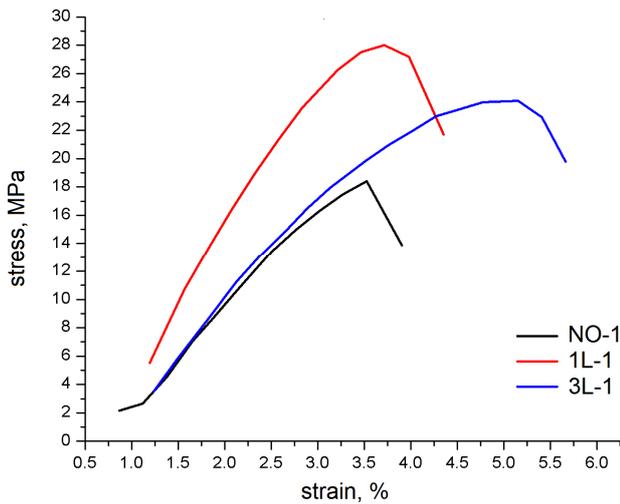


Figure 5. Stress-strain curves for PLA – group 1

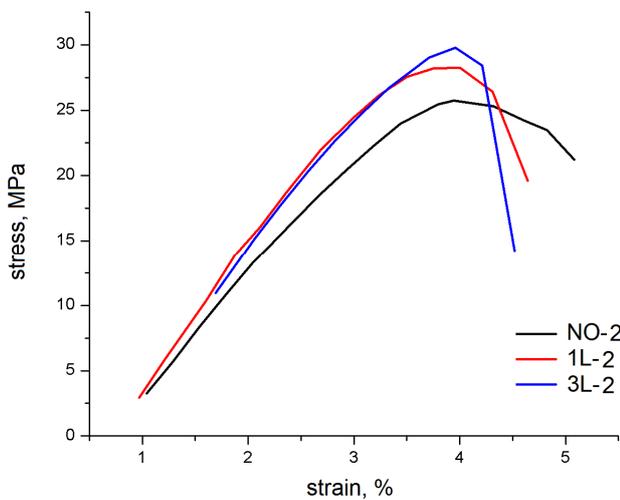


Figure 6. Stress-strain curves for PLA – group 2

As it may be observed, the maximum fracture strength is increased for the reinforced samples, as expected, and the lowest values were noted for unreinforced PLA specimens designated as NO – 1. For the specimens with 1 and 3 layers, in most cases, higher fracture strength was noted for the samples with one layer of the reinforcing PVC mesh, except for the specimens designated as 1L – 4. The reason for this may be in the overall weaker structure of this mesh orientation.

The lowest fracture strength value of 18.62 MPa was observed for the unreinforced specimen designated as NO – 1 and the highest value of 31.30 MPa for the specimens with 3 layers of PVC mesh, labelled as 3L – 4, which is approximately 68 % increase of fracture strength. Maximum strains at break were observed for groups 3 and 4 of the specimens. When we compare only the effect of printing parameters, it has led to an improvement from 18.62 to 28.55 MPa for unreinforced specimens designated as NO – 4. Further, the failure mechanism is different for the different

specimens. As it can be noticed in Figure 5, for the group 2 specimens, there is a higher extent of PLA filament stretched and pulled out of the specimen structure. Other specimens seem to have more brittle fractures. Also, the presence of the PVC meshes has significantly contributed to the gradualness of the failure and the fracture strength.

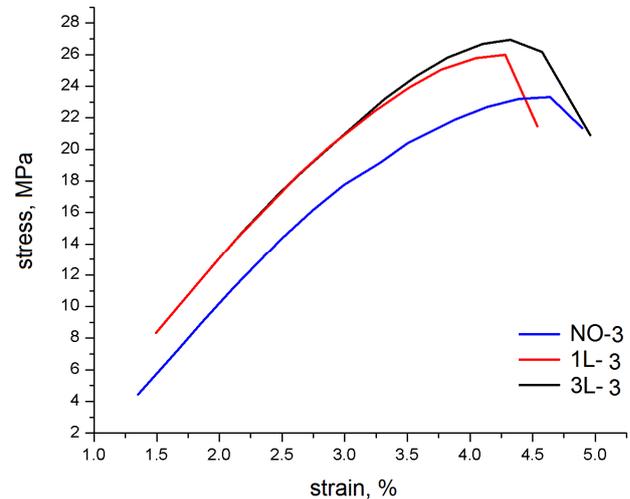


Figure 7. Stress-strain curves for PLA – group 3

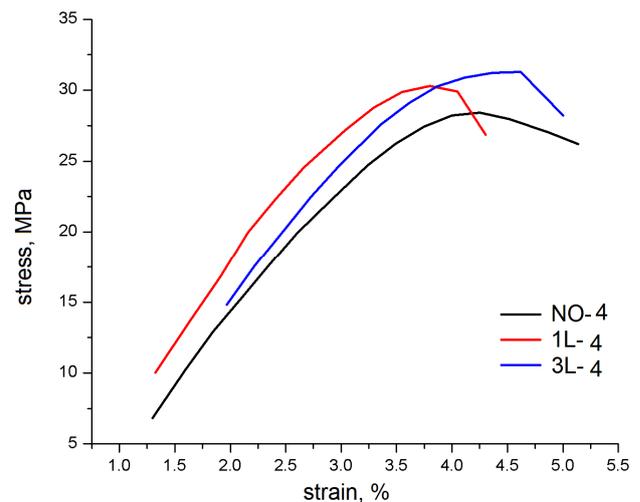


Figure 8. Stress-strain curves for PLA – group 4

A comparison of the results of the tensile test in previous research also indicates lower values [20,21,32]. Compared to previous research [32], this is expected due to the dimensions of the standard sample. Also, metal nanoparticles and the incorporation of wood particles have a greater influence on the mechanics of the composite than the combination of two polymers [20,21]. The flexural strength of the PLA samples, which were printed with 100 % infill, was 214 MPa [33], which indicates that the infill density during 3D printing significantly influences the mechanical properties and satisfactory results on the three-point bending test.

Figure 9 shows microhardness values in MPa for all three series. An increase in hardness in the series with one layer can be noticed. The lowest average hardness of 87.74 MPa was noticed, as expected, for unreinforced specimens designated as NO – 1. The highest value of microhardness of 104.11 has a sample with one PVC mesh layer designated as 1L – 4 (0.2 mm layer height and infill density of 60 %). In this case, the increase in hardness is approximately 15.7 %. By comparing the microhardness results, the high hardness of the specimen indicated and reflected the low values of porosity [21].

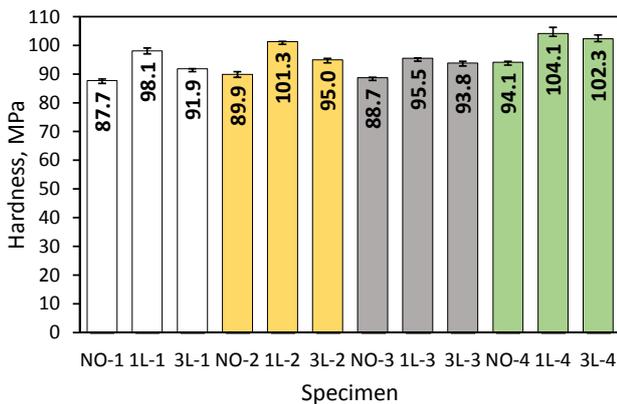


Figure 9. Average microhardness values for three series of specimens

The microhardness of PLA printed samples in our previous research ranges from 108 to 125 MPa depending on the printing orientation and thickness of the layer of the PLA [32]. A slightly lower hardness value in this research indicates an increase in porosity and possibly cooling of the 3D printing filament when inserting the PVC mesh during the printing process. This behaviour is indicated by the decrease in the microhardness of the group of samples with three layers. It should also be taken into account that the penetration of the indenter is 1/7 of the size of the diagonal [32], which indicates that the influence of PVC mesh located in the volume of the PLA matrix is insignificant in relation to the print line width and orientation.

4. Conclusion

A new kind of additively manufactured PLA/PVC laminate composite has been developed and characterised. It was shown that by introducing only 3 layers of PVC mesh and selecting the optimal 3D printing orientation, it is possible to increase the fracture strength from 18.62 to 31.30 MPa, i.e. by 68 %. Also, the hardness test proved that the composite structures with one and three

mesh layers have better mechanical characteristics than unreinforced specimens. Specimens without PVC mesh showed the lowest elasticity while the highest elasticity was at specimens with one layer of PVC.

The highest increase in fracture strength was obtained in the specimens of group 4, with mesh orientation 90° | 45° | 90° and layer thickness of 0.2 mm, infill density of 60 % and maximal number of PVC mesh layers (3 layers) inserted in PLA matrix with height position 25 % | 50 % | 75 %; while the highest increase in microhardness was obtained in the specimens of series 2 (designated as 1L), with 1 PVC mesh layer of height position 50 % and mesh orientation 90°. Increasing the infill density (from 30 to 60 %) also contributes to the increase of microhardness due to better compactness and reduced porosity in specimens.

The hypothesis was that strength should increase with the number of inserted layers. However, stopping the printing (especially for three layers) caused the model to cool rapidly, which meant weaker bounding and delamination. Future research will focus on the type of fracture of the laminate material, observing the intersection of the two polymers, and optimising the printing parameters and reinforcement in the matrix of the printed material for potential medical applications.

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