

Studying the effect of silica (SiO₂) addition on the adhesive properties of polyvinyl alcohol

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Abstract

The work concerned with studying the effect of (SiO₂) addition as a filler on the adhesive properties of (PVA). Samples were prepared as sheets by using casting method. The mechanical properties showed that increase in tensile strength from (34MPa) to (68MPa) when (SiO₂) added to (PVA). The adhesive strength showed that joint properties depend upon specific adhesive characteristic of material (PVA) and (SiO₂\PVA)composites at different concentrations (1.5%, 2.5%, 3.5%, 4.5wt%), the cohesive strength of the adhesive material, the joint design, and adherent type (Sponge Rubber(SR), Natural leather (NL), Vulcanized Rubber(VR), and Cartoon). The results proved the tensile strength increased with (SiO₂) ratio, so it can be used as the adhesive material. Shear strength showed an increase with (SiO₂) ratio of sponge rubber, and cartoon adherent, whereas it was increased up to 2.5% for Natural Leather, and Vulcanized Rubber then decreased; That suggested it is most suitable for sponge rubber adhesive and cartoon than the other adherents.

Key words

Adhesive strength, PVA, SiO₂.

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دراسة تأثير اضافة السليكا (SiO₂) على خصائص اللصق للبولي فينيل الكحول (PVA)

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الخلاصة

تمت دراسة تأثير اضافة (SiO₂) على خصائص الالتصاق لل (PVA)، حضرت النماذج بشكل الواح وباستخدام طريقة الصب. أظهرت الخواص الميكانيكية زيادة في متانة الشد لمنطقة الالتصاق باضافة (SiO₂) الى (PVA) من (34MPa) الى (68MPa). بينت قوة اللصق بان خواص الربط تعتمد على خواص اللاصق (PVA) او (SiO₂\PVA) بمختلف التراكيز (1.5%, 2.5%, 3.5%, 4.5wt%), متانة اللصق لمادة اللصق، تصميم نقطة الربط، نوع المواد المراد لصقها (المطاط الاسفنجي، الجلد الطبيعي، المطاط المفلكن، الكارتون) أظهرت النتائج ان متانة الشد تزداد بزيادة نسبة (SiO₂)، لذا يمكن استخدامها كمادة لاصقة. بينت نتائج قوة القص زيادة في متانة القص بازدياد نسبة (SiO₂) بالنسبة للمطاط الاسفنجي (SR) والكارتون (Cartoon) في حين هناك زيادة في قوة القص بازدياد نسبة SiO₂ 2.5% بالنسبة للجلد الطبيعي والمطاط المفلكن (VR) ومن ثم تقل. من خلال نتائج البحث يمكن التوصية بامكانية استخدام SiO₂ كمادة لاصقة واعدة واكثر مناسبة للمطاط الاسفنجي والكارتون.

Introduction

Adhesive is a material that can bind solid substance together by surface attachment; it represents that the attraction between the surface of two material. Adherent is the solid substance to which the adhesive adheres. The

whole structure that is made by the adhesive system and the adherends together is called adhesive bond or joint. The physical strength of these adhesive bonds is named practical adhesion. The practical adhesion effects by the force of adhesion, but its magnitude calculated

by the physical properties of the adhesive and these adherends, besides the geometry of the adhesive bond [1].

Adhesives are providing the means by which substrates will be held together in addition to some other secondary functions. The most important characteristic of adhesive is their multi-functional nature, as well as performing a mechanical fastening operation. There are also another application, where adhesive can be useful, it can be used as a sealant, insulator, vibration damper, and gap fillers. Because of its viscoelastic materials nature, it can reduce the vibration and noise that occur in some assemblies. Adhesives can also be used as sealing functions because it works as a barrier, which inhibits the passage of gases and fluids. It can also be useful as thermal and electrical insulator in a joint. The insulation degree depends on the adhesive formulations and fillers [2].

Adhesive can be utilized as a silver filler and thermal conductors when it is used with boron nitride fillers. When dissimilar metals are bonded, adhesives will be used as galvanic corrosion resistance because it is a dielectric material [3].

Experimental part

1- Materials

PVA was used as a white powder, which was supplied by (Barcelona Espana) (DIDACTIC), Silicon dioxide (SiO_2) of particle size ($1.112\mu\text{m}$) as additive material, and distilled water was used as a solvent for PVA.

Natural Leather (NL) is a durable and flexible material, created by the tanning of skin and animal rawhide, often cattle hide. It can be processed through a manufacturing process ranging from cottage industry to heavy industry. Leather, is utilized for different purposes including clothing (hats, jackets, shoes, skirts, and trousers), bookbinding, Leather

wallpaper and as a furniture covering. It is produced in a wide variety of types and styles, and it is used for decoration in wide range of techniques.

Vulcanized Rubber (VR) is a synthetic rubber exposed to a vulcanization process. Vulcanization process treats rubber to give it certain qualities, strength, elasticity, and resistance to solvents, and to render it impervious to moderate heat and cold. Chemically, the process involves the formation of cross-linkage between the polymer chains of the rubber molecules.

Sponge Rubber (SR) It is a rubber, that can be synthesized in several different types with low, medium or high-density firmness. Benefits and advantages are controlled compression and recovery characteristic, shock absorption attributes highly effective sealing capabilities. Applications are door seals, sound and vibration isolation, cushion pads and gaskets.

2- (PVA) films preparation

The required weight of (7g) of purified (PVA) was dissolved in (100ml) of distilled water. The stirring process done by using heating, magnetic stirrer heated up to $80\text{ }^\circ\text{C}$ until we get a homogeneous solution. The solution then left to cool down until it reaches room temperature. The homogeneous solution was casted at "glass flat" glass plate, Then it was left to dry. Then the dried sample put in the oven with a temperature between ($60\text{--}70\text{ }^\circ\text{C}$) for six hours.

The casted film can be dismantled easily from the glass plate by using forceps.

3-Preparation of SiO_2 /PVA composites

Polymer systems of PVA and Silica micro particles with different concentrations (1.5%, 2.5%, 3.5%, and 4.5 wt %), were prepared by solution

casting method using distilled water as a solvent.

For SiO_2 addition (1.5%, 2.5%, 3.5%, and 4.5 wt %), the previous process of preparing (PVA) sample up to reach to the room temperature must be followed. When the solution was cooled down to room temperature SiO_2 added at different concentrations then mixed by using magnetic stirrer until it forms a homogeneous solution. The homogenous solution then casted on a flat glass, and left for 24 hours to dry out. The casted sample must be placed in an oven with (60-70°C) for six hours to dry.

These films were placed on the glass plate; all obtained films were semi-transparent, uniform in thickness, and free from air bubbles.

There are two types of tested samples used to measure adhesive strength (tensile strength, and shear tensile strength).

The optical microscope was used to be assurance from the prepared films quality; it must be free from bubbles. Fig. 1 shows optical microscope images of sample involved.

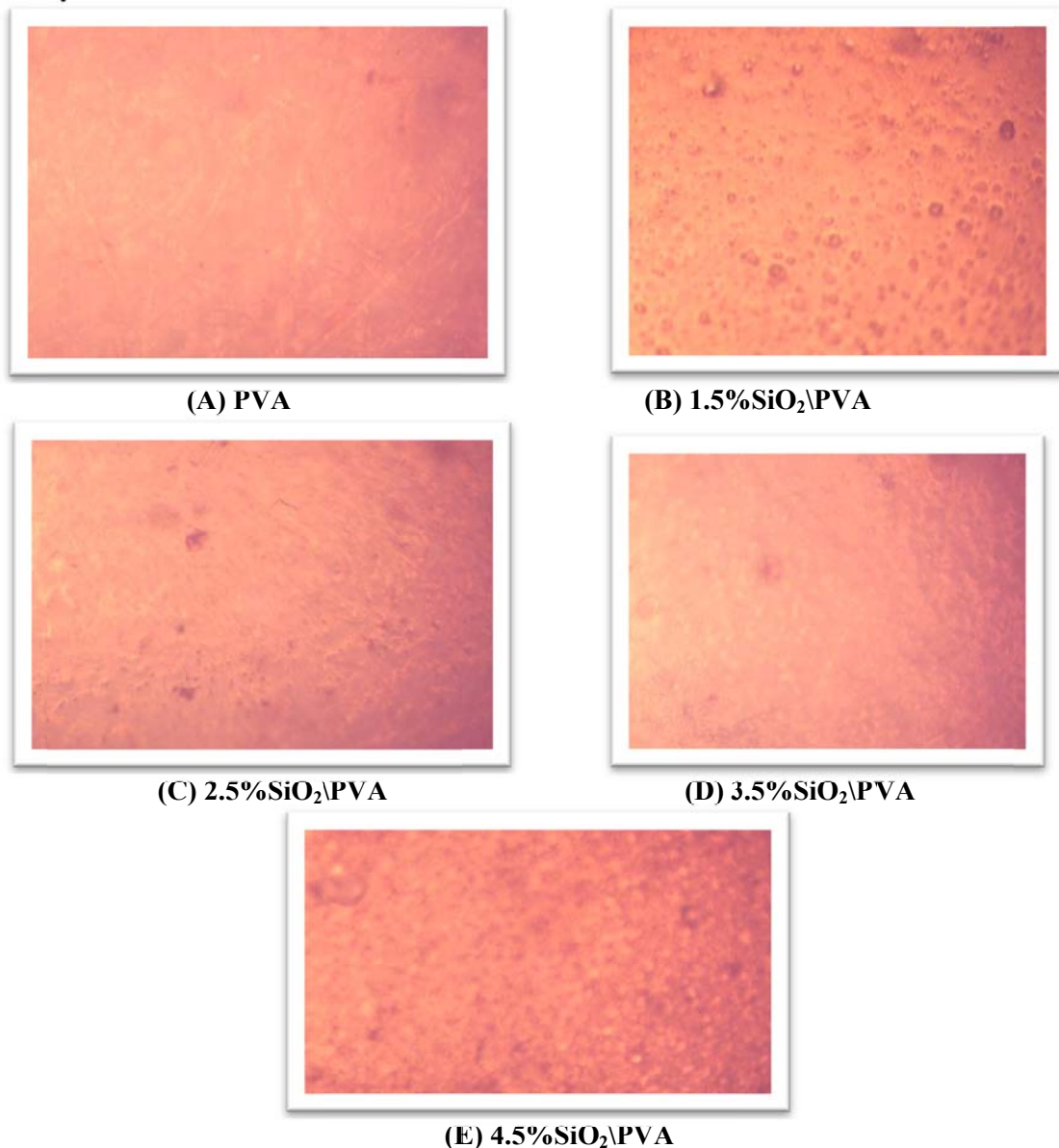


Fig.1: Optical microscope images of PVA before and after SiO_2 addition at different concentration.

4-Tensile test

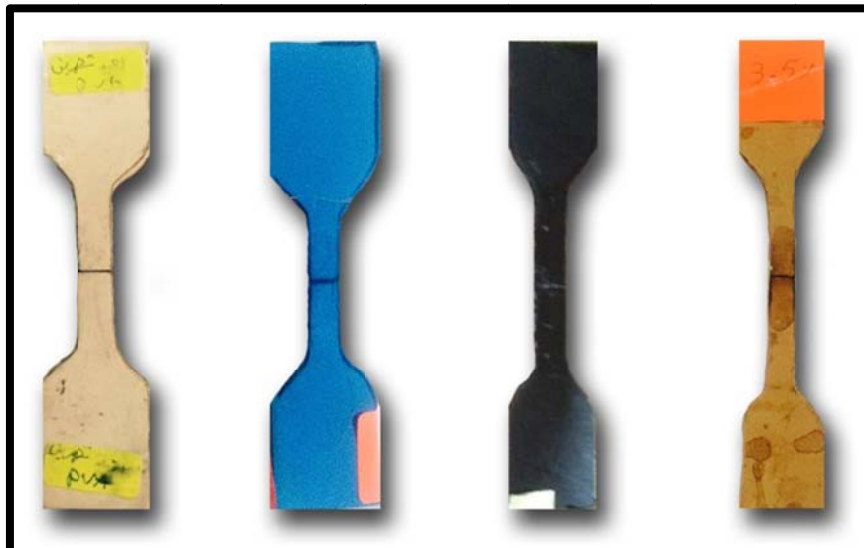
Four types of materials used as adherents (Sponge rubber (SR), Natural leather (NL), Vulcanized rubber (VR), and Cartoon) according to the (IQS 1330) standard [4].

The samples were tested as a standard before cutting. Then it was cut and adhesive from its middle region. The samples were adhesive by PVA and SiO_2/PVA were previously prepared at different concentration.

Then it has been left for 24hr to be ready for testing. Its stress-strain curves and load extension curves, the Ultimate and F_{max} were obtained irrespectively by using a tensile machine of (LARYEE-50 KN, China), with the crosshead speed of (5mm/min). Fig.2 shows the Vulcanized rubber, Natural Leather and Sponge Rubber, and Cartoon samples before and after adhesive by PVA and SiO_2/PVA .



(A)



(B)

Fig. 2: (A) images shows the vulcanized rubber, natural leather and sponge rubber, and cartoon samples before adhesive with PVA and SiO_2/PVA .

(B) images shows the vulcanized rubber, natural leather and sponge rubber, and cartoon samples after adhesive with PVA and SiO_2/PVA .

5- Shear test

As in section 2.4, It was used four types of materials as adherents (Sponge rubber, Natural leather, Vulcanized rubber, and Cartoon) according to (IQS 1330) standard [4]. In addition, the previous literatures, the shear strength was obtained by using a

tensile machine of (LARYEE-50 kN, China), with the crosshead speed of (5mm/min) for samples involved. Fig.3 shows the Vulcanized rubber, Natural Leather and Sponge Rubber, and Cartoon which were adhesive by PVA, SiO_2/PVA at different concentration.



Fig.3: The vulcanized rubber, natural leather and sponge rubber and cartoon samples, which were adhesive by PVA, SiO_2/PVA at different concentration.

Result and discussion

1. Adhesive strength

In modern technique adhesive bonding is a joint of two similar or non-similar (metal, plastic, compositions,..... etc) by using an adhesive material [4]. In this process a layer of an adhesive composition (PVA, SiO_2/PVA) chemically different from the adherent. The (Sponge Rubber (SR), Natural Leather(NL), Vulcanized rubber(VR) and cartoon) to be bonded is interposed between them. The resultant joint properties depend upon the specific adhesion characteristic of materials, the cohesive strength of adhesive, and the design of the joint [5,6].

The joint bonded with adhesive are generally stronger in tension and shear than other joints. It is much easier to break an adhesive joint by accessing an edge and peching it a way[7]. Figs.4 to 7 show the stress-

strain curves for (Sponge Rubber(SR), Natural Leather(NL), Vulcanied rubber(VR), and Cartoon); it was seen increase in ultimated strength with SiO_2 ratio; that was attributed to (PVA), which possess a unique characteristics as excellent film forming ability , good water solubility, appropriate compatibility with superior resistance to solvent; and excellent in properties such as remarkable abrasive resistance, So it is used as an adhesive material; It is evident that that the molecular structure of (PVA) plays an important role in adhesion to materials[8]. The increase in ultimate strength was attributed to the good compatibility of (SiO_2) with (PVA) (good distributions in matrix); there is no unifying theory of adhesion describing the relationship between partical adhesion and basic intermolecular and interatomic interactions, which take place between

the adhesive material and adherent either at the interface or within the interphase[5,8]. Fig. 7 cartoon; it was seen increase in ultimate strength with SiO₂ ratio up to 2.5% then decrease; it was attributed to processes, with less the intermolecular bonds. Figs. 8A-8B shows the Ultimate strength for adherents standard (SR, NL, VR, Cartoon, and PVA before and after

SiO₂ addition. Figs. 9A-9F show the relationship between the adhesive forces which was of the same behaviour of ultimate strength-for all samples. It was seen the adhesive force within the cohesive force of adherents. Fig. 13 shows the F_{max} of adherent and adhesive material for samples involved. It was followed the ultimate strength changes [5-10].

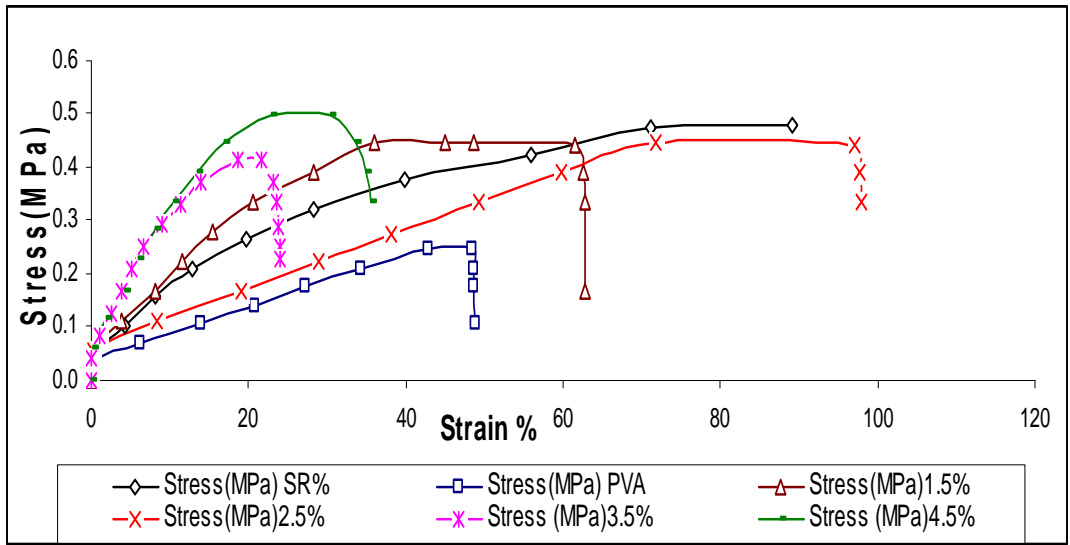


Fig. 4: Stress-strain curves of sponge rubber samples.

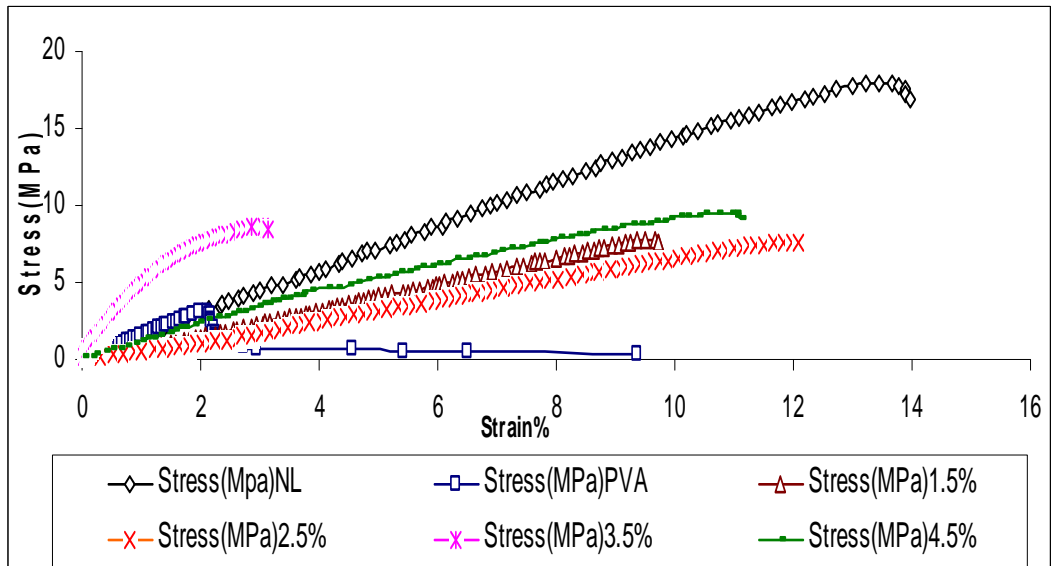


Fig. 5: Stress-strain curves of natural leather samples.

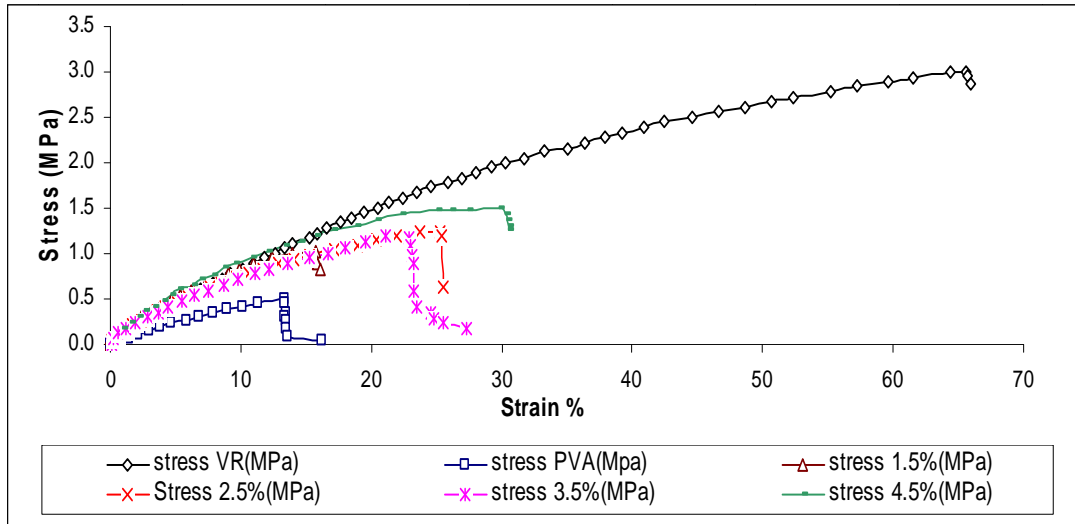


Fig. 6: The stress-strain curves of vulcanized rubber.

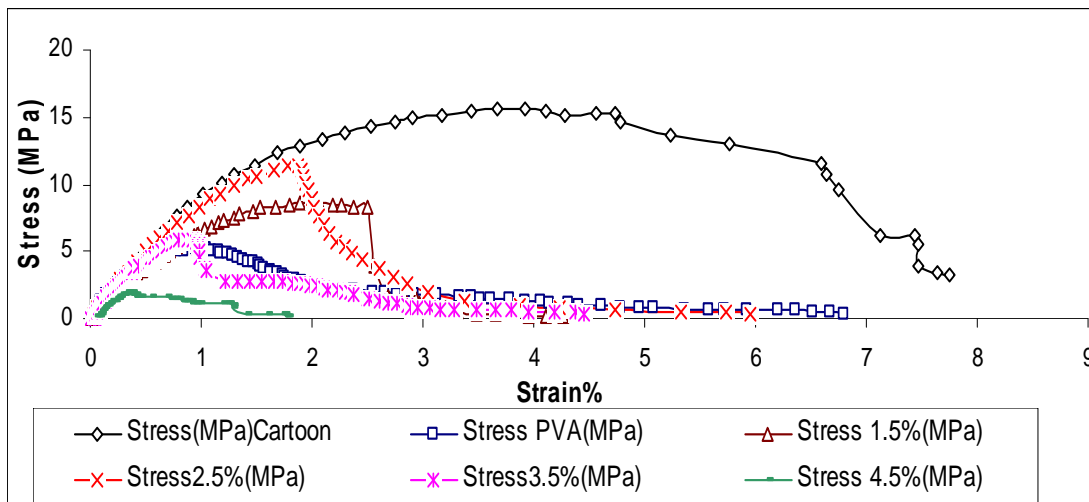
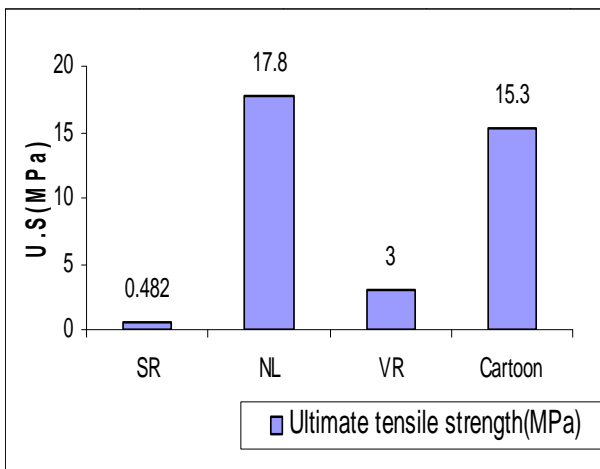
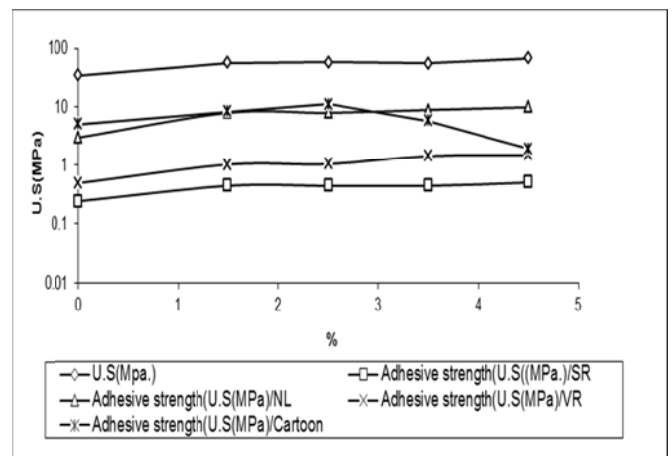


Fig. 7: Stress-strain curves of cartoon sample.

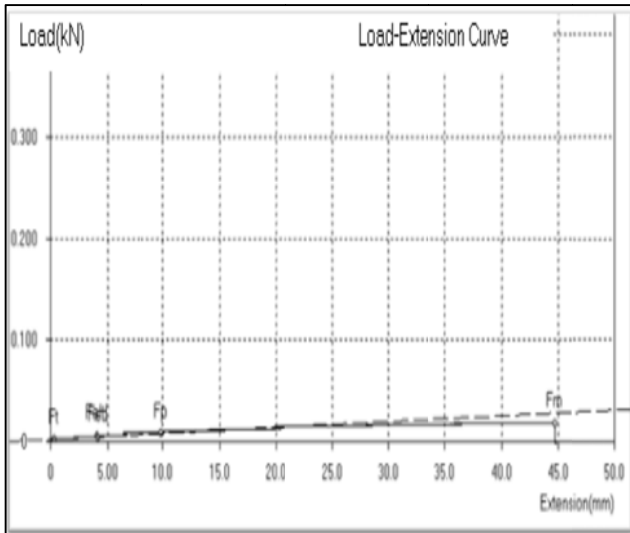


(A)

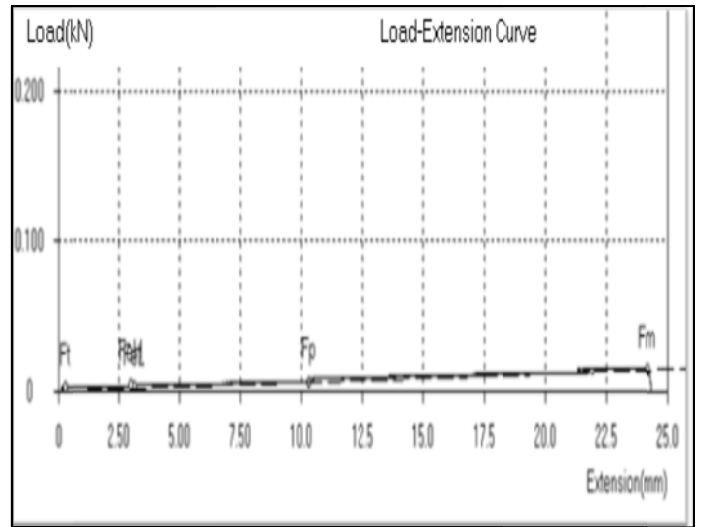


(B)

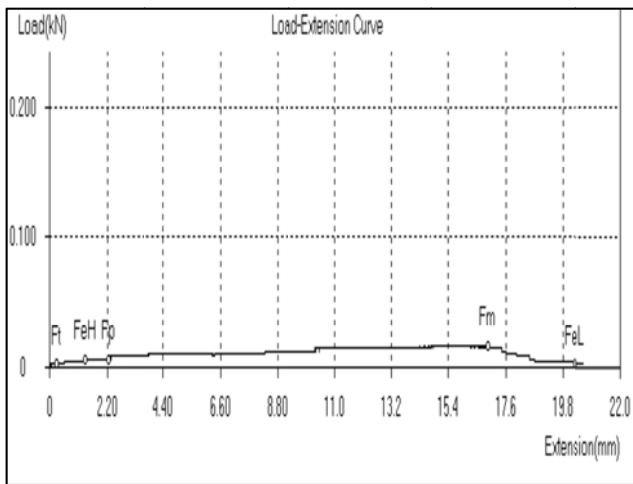
Fig. 8: (A) The Ultimate strength of adherent. (B) The ultimate strength of PVA and SiO₂ addition as adhesive material.



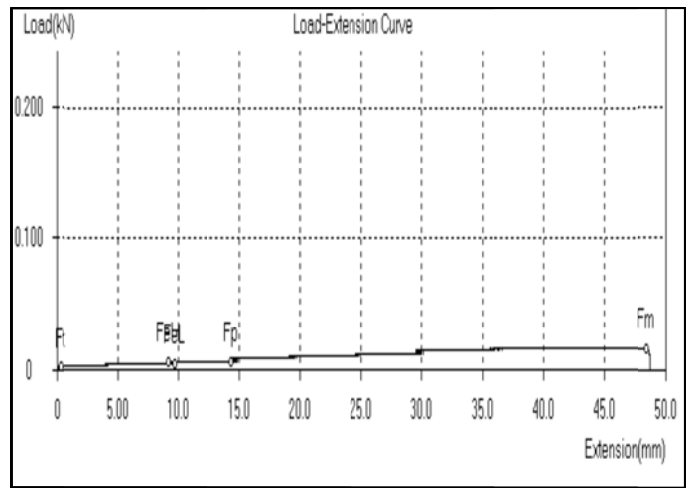
(A) Sponge rubber standard



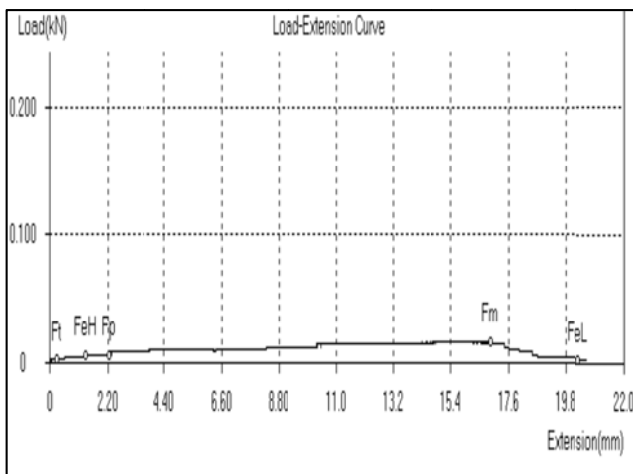
(B) PVA



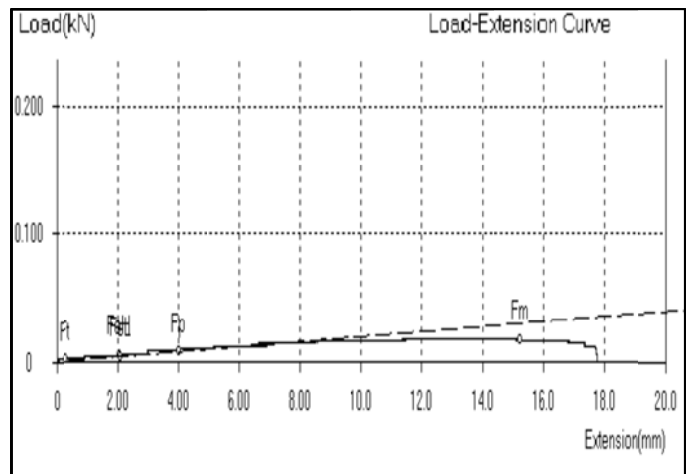
(B) 1.5%SiO₂/PVA



(D) 2.5%SiO₂/PVA

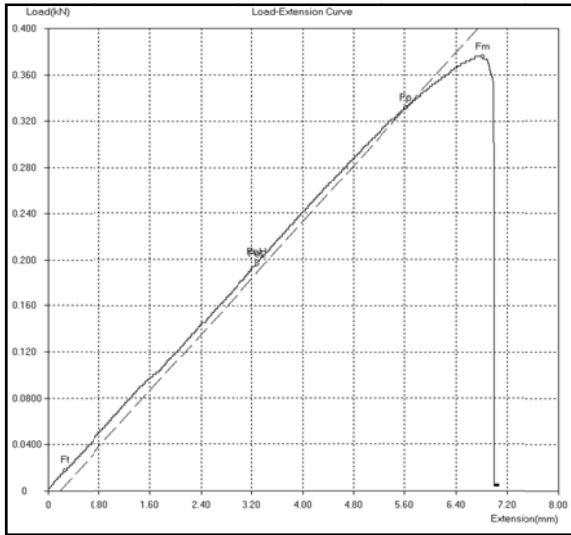


(E) 3.5%SiO₂/PVA

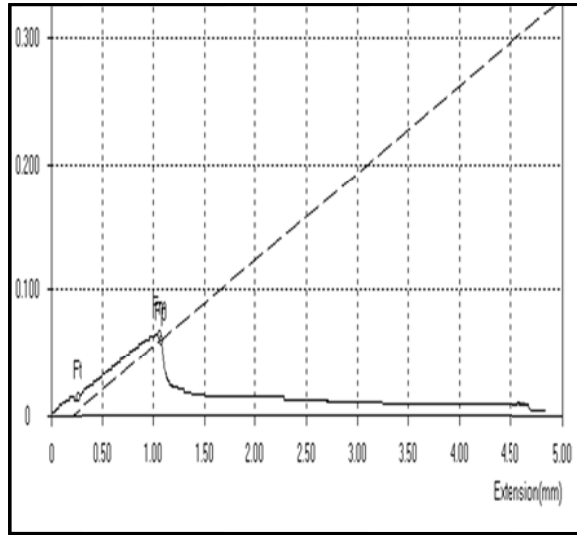


(F) 4.5%SiO₂/PVA

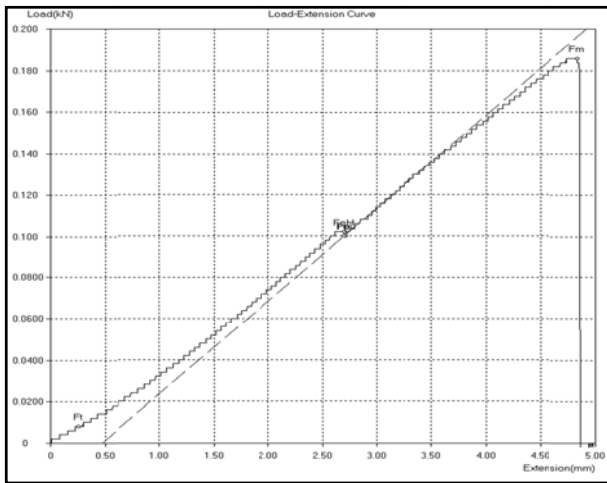
Fig. 9: The load-extension curves of sponge rubber.



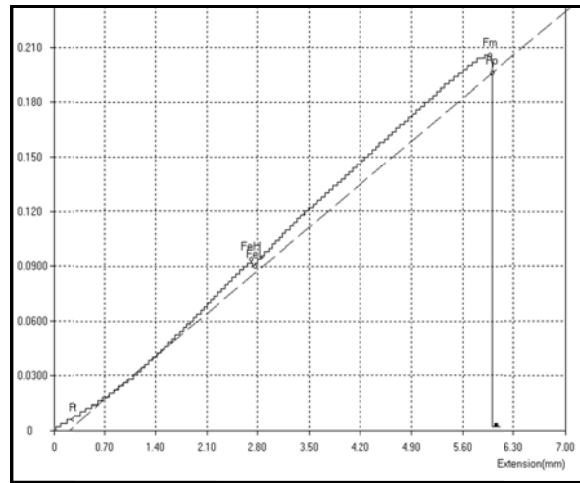
(A) Natural leather standard



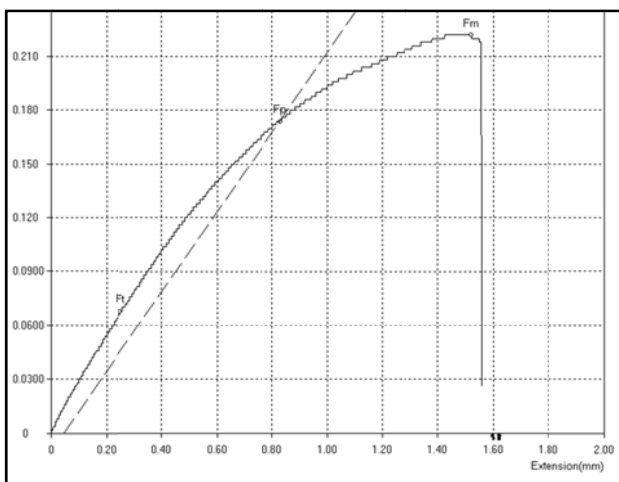
(B) PVA



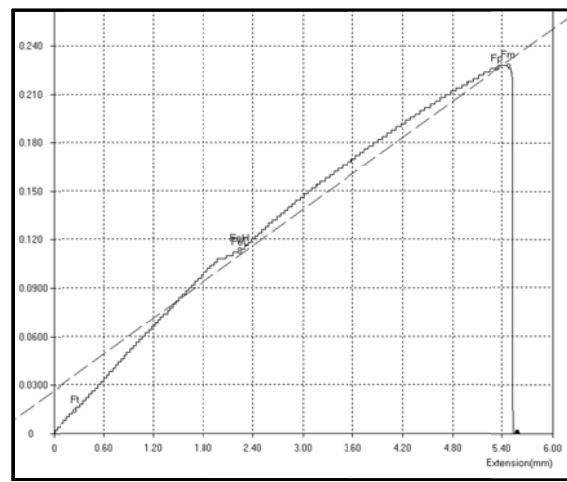
(C) 1.5%SiO₂\PVA



(D) 2.5%SiO₂\PVA

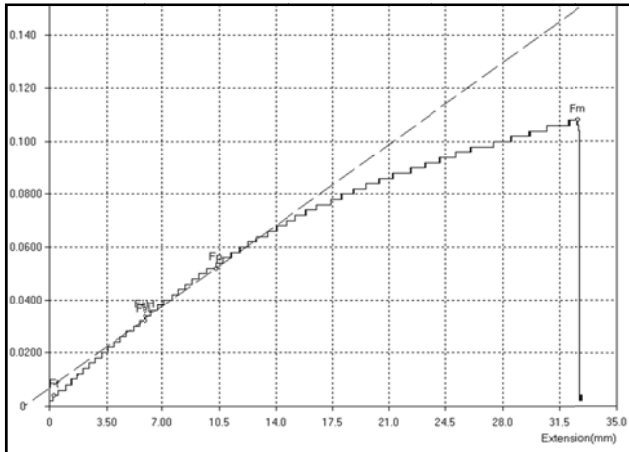


(E) 3.5%SiO₂\PVA

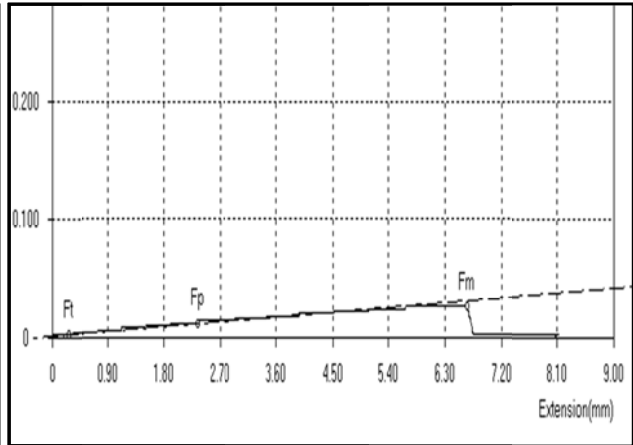


(F) 4.5%SiO₂\PVA

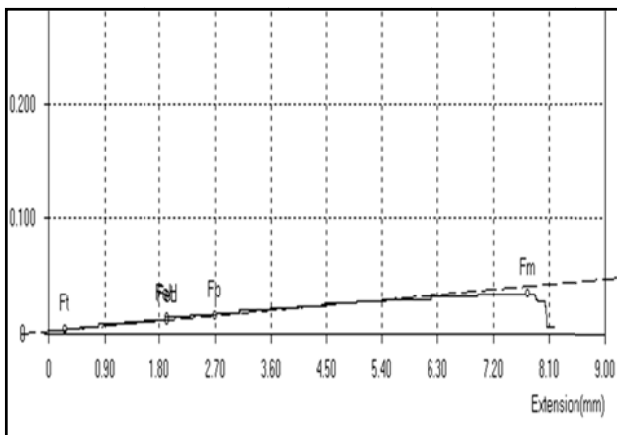
Fig. 10: The load-extension curves of natural leather.



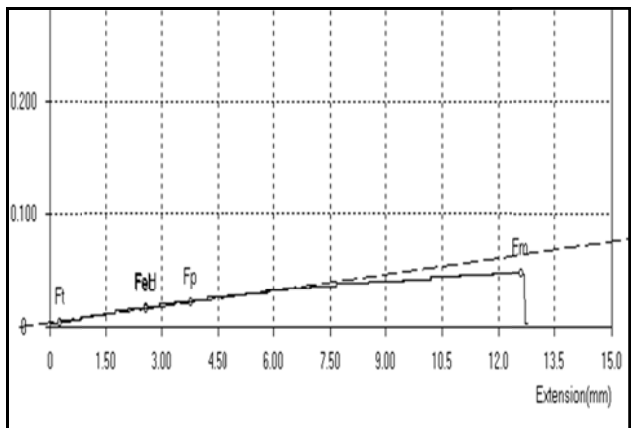
(A) Vulcanized rubber standard



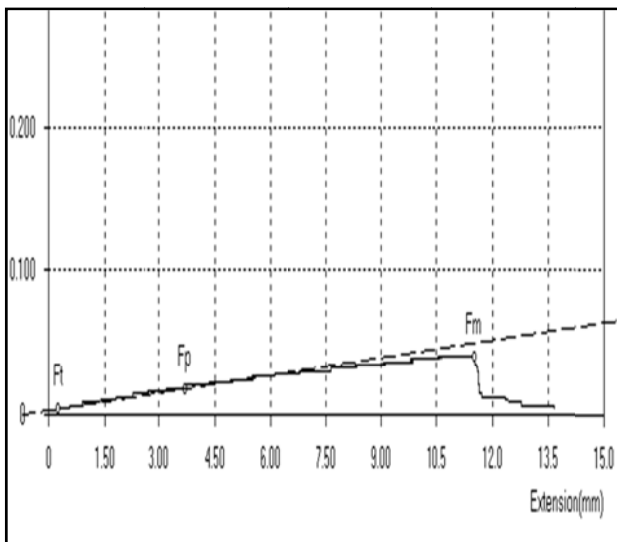
(B) PVA



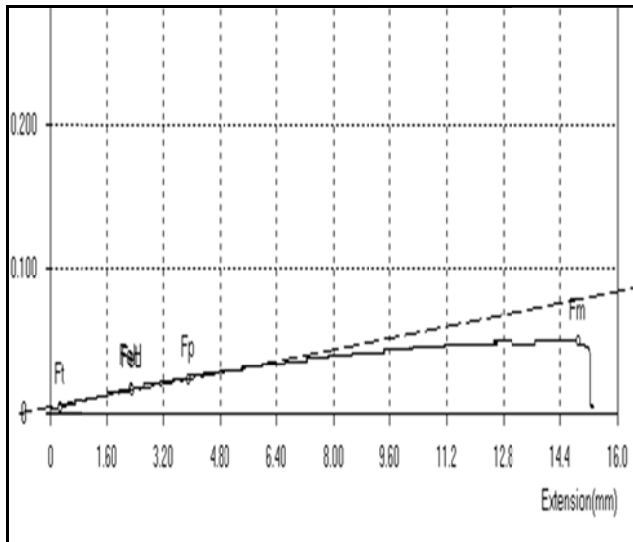
(C) 1.5%SiO₂/PVA



(D) 2.5%SiO₂/PVA

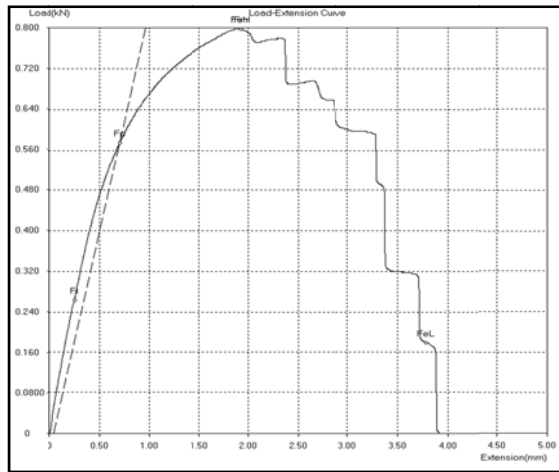


(E) 3.5%SiO₂/PVA

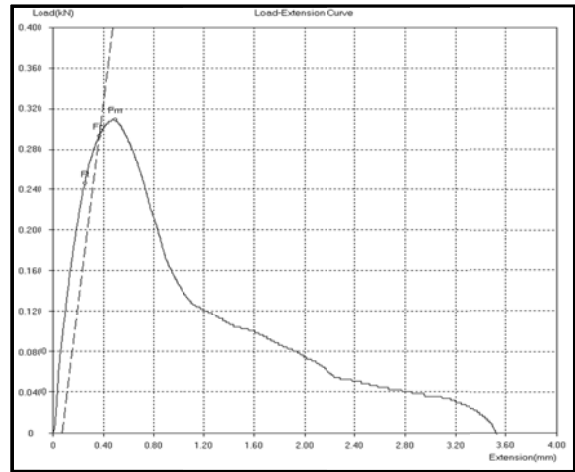


(F) 4.5%SiO₂/PVA

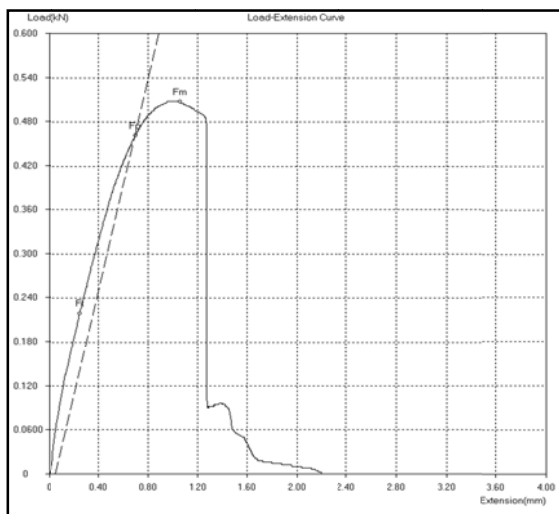
Fig. 11: The load-extension curves of vulcanized rubber.



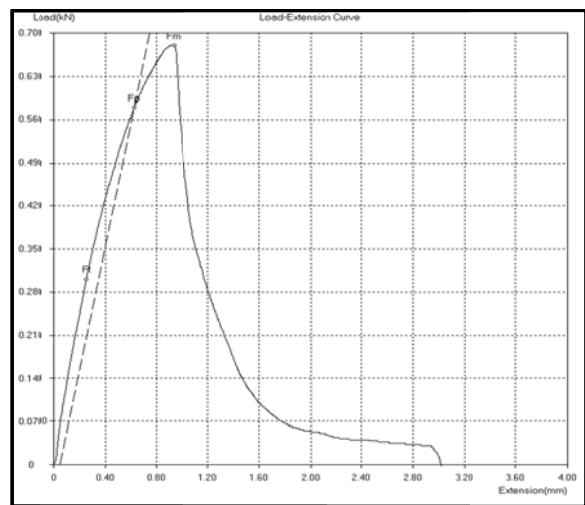
(A) Cartoon standard



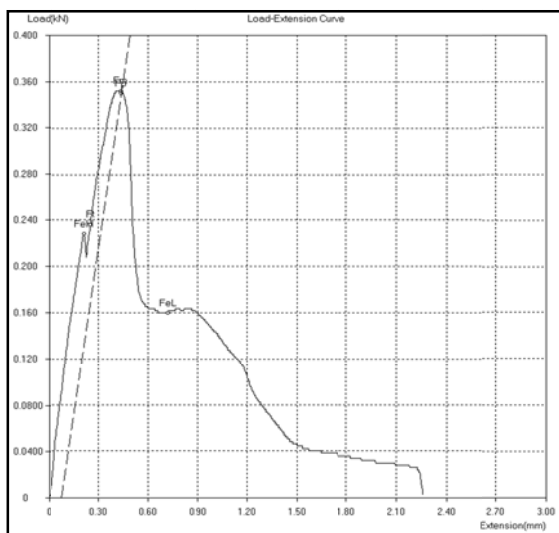
(B) PVA



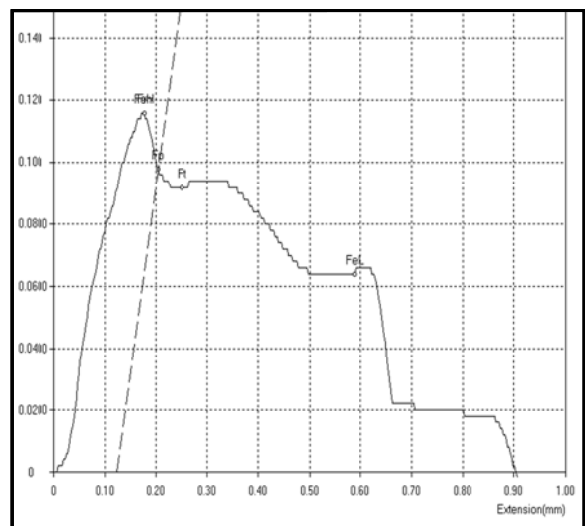
(C) 1.5%SiO₂/PVA



(D) 2.5%SiO₂/PVA



(E) 3.5%SiO₂/PVA



(F) 4.5%SiO₂/PVA

Fig. 12: The load-extension curves of cartoon samples.

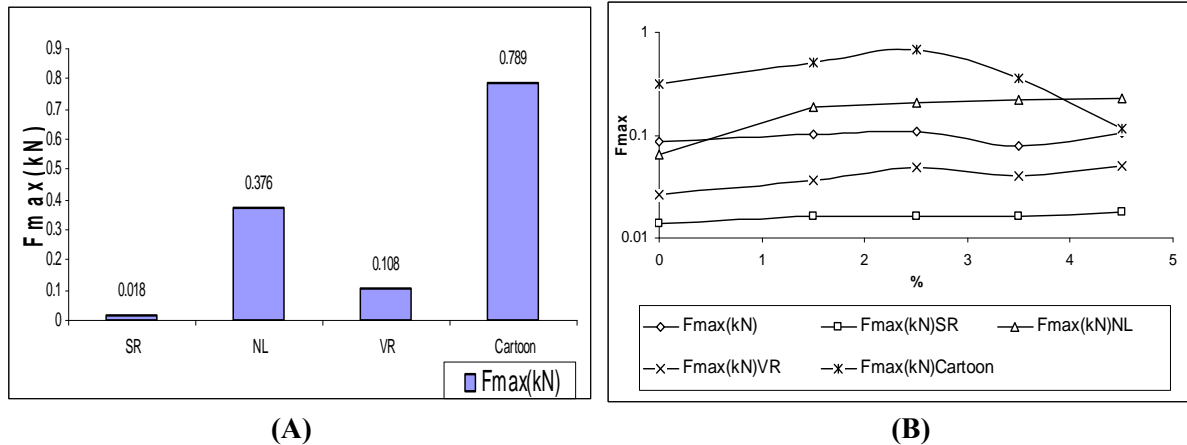


Fig. 13: (A) F_{max} of adherent materials. (B) F_{max} of PVA before and after SiO_2 addition at different concentration.

2. Shear strength

From the Fig.(14A-14E) to (17A-17E) it was seen the shear strength of PVA and SiO_2 /PVA at different concentration as adhesive material and Sponge Rubber, Natural Leather, Vulcanized rubber and Cartoon as adherent material. It was found that the adhesive strength increased with SiO_2 ratio, which increased for sponge rubber and cartoon adherent. Shear strength and elongation at break are the basic characteristic relations to mechanical properties of adhesives. The shear strength of adhesion also induces failures in external surface veneers. If there is higher shear strength also higher veneer failure [11]. Fig. 15A-15E shows the shear strength of PVA. As adhesives material containing different ratio of SiO_2 (0-4.5 wt%); The shear strength of PVA as adhesive increased from (0.074 MPa) to (0.346MPa) at 2.5% SiO_2 for Natural Leather adherents. The results shows that the maximum shear force that's capable of separating or de-bonding joined of Natural Leather pieces glued together was evidence of strong interaction between SiO_2 and PVA adhesive through hydrogen bonding. The results were in a good agreement with Bhatnagaretal [12]. The increase

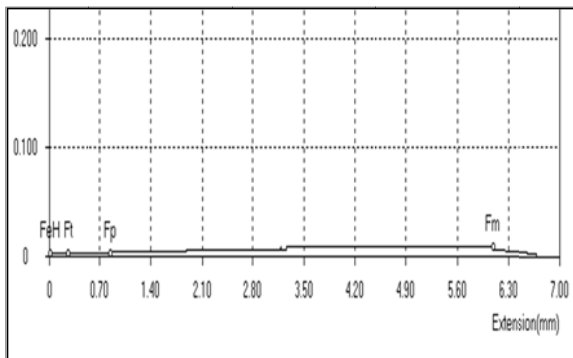
of SiO_2 ratio at more than 2.5% SiO_2 caused glasterining of the SiO_2 matrix, which really decrease the effective content of SiO_2 these ratio exhibited lower shear strength compare to samples of the lower ratio [13]. In Fig.(16A-16E) it was seen the shear strength of PVA and SiO_2 /PVA at different concentration with Vulcanized Rubber as adherent. It was found there was unsystematic change in shear strength, and the maximum shear strength was at 2.5%. Thus could be explained by the different behavior of adhesive when loaded in shear and intension in the presence of defect such as voids. In tension, once a crack is triggered next to a void, the sample fails was attributed to the high stress concentration. In shear even of a crack is triggered, the remaining area is capable of further deformation, especially if adhesives is ductile [14]. Tensile adhesive tests involve a complex mixture of tensile and shear forces, which render difficult the investigation of the results. Moreover; it has been explained extensively in literature Berndt, C.C. and C.K. Lin[15] that this test cannot be related to fracture mechanic characteristics like toughness, as the crack propagation is spontaneous and depends on the critical of low size at

the interface [14-17]. Fig. 18 shows hear strength of the samples involved.

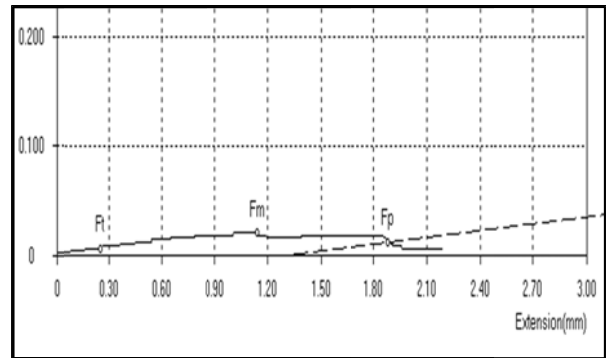
Conclusions

In general the tensile strength increased with SiO₂ concentration from 1.5% to 4.5%, so the sample involved can be used as the adhesive

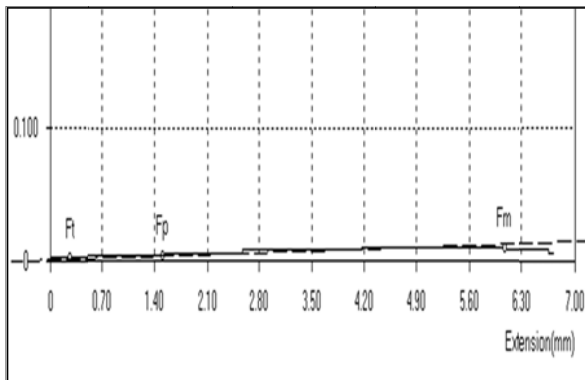
material. It was found the adhesive strength increased with SiO₂ for sponge rubber, and cartoon adherents, whereas the results showed maximum shear strength for Natural Leather and Vulcanized rubber at 2.5% SiO₂, so it can be used as the adhesive material.



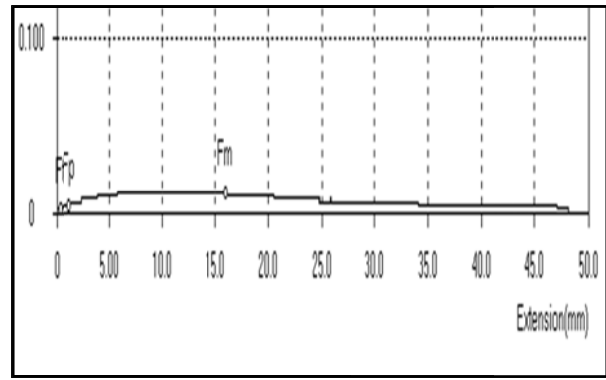
(A) PVA



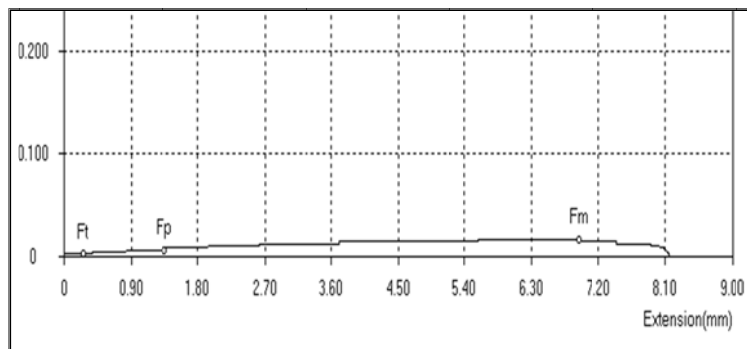
(B) 1.5%SiO₂\PVA



(C) 2.5%SiO₂\PVA

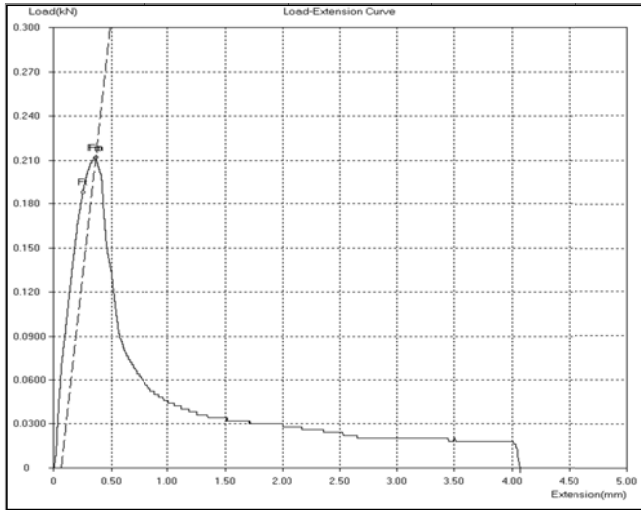


(D) 3.5%SiO₂\PVA

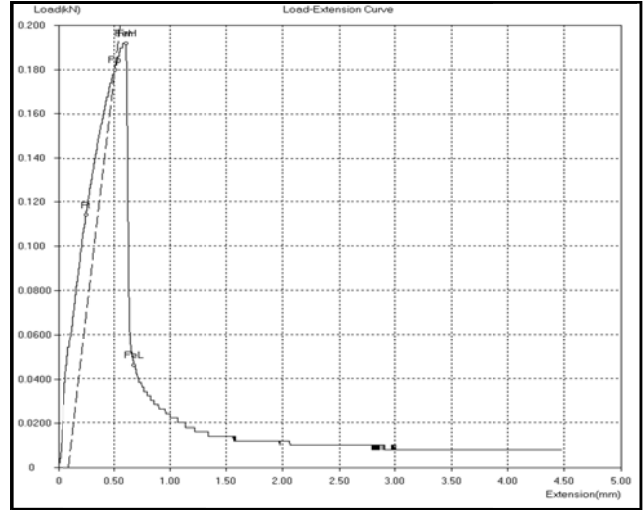


(E) 4.5%SiO₂\PVA

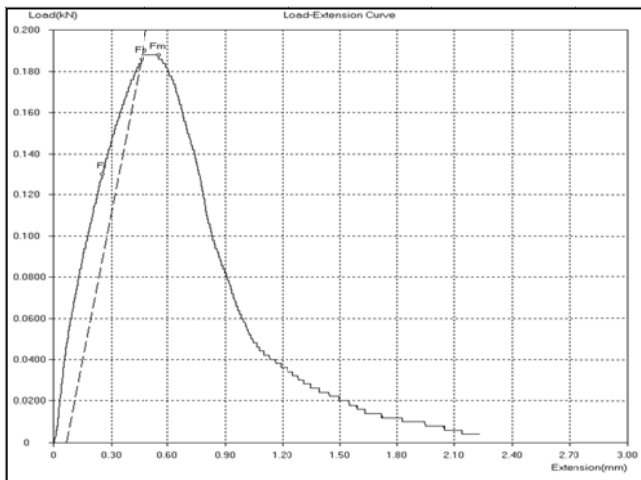
Fig. 14: Load-extension curves of sponge rubber samples.



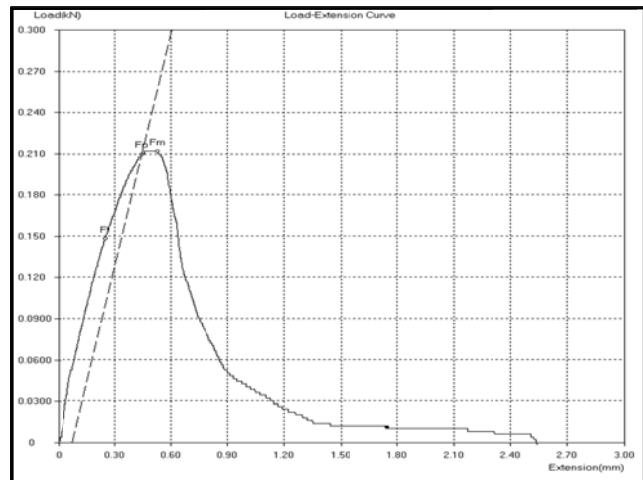
(A) PVA



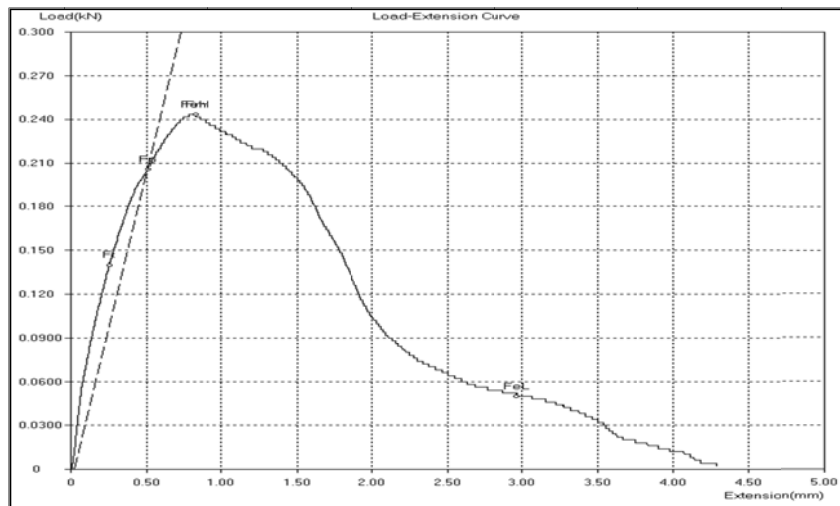
(B) 1.5%SiO₂/PVA



(C) 2.5%SiO₂/PVA

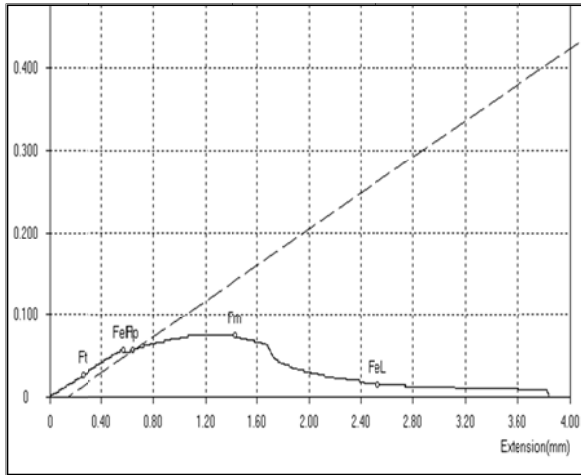


(D) 3.5%SiO₂/PVA

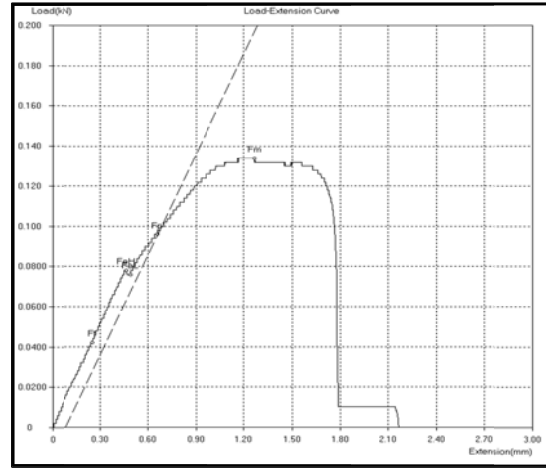


(E) 4.5%SiO₂/PVA

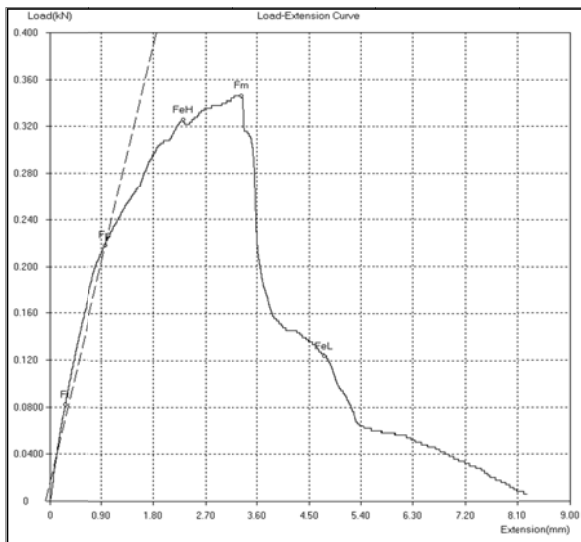
Fig. 15: Load-extension curves of natural leather samples.



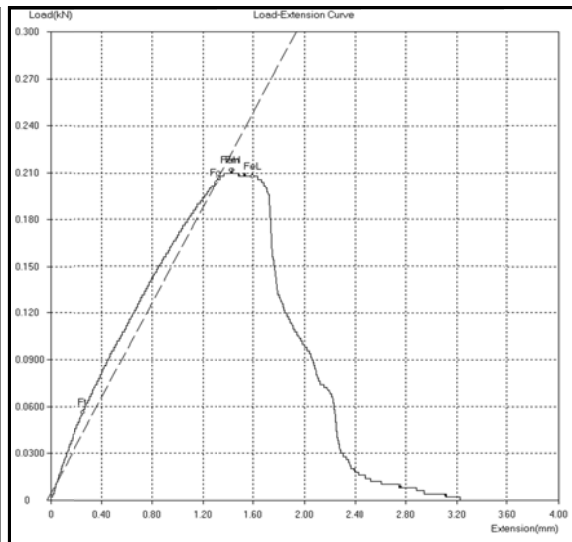
(A) PVA



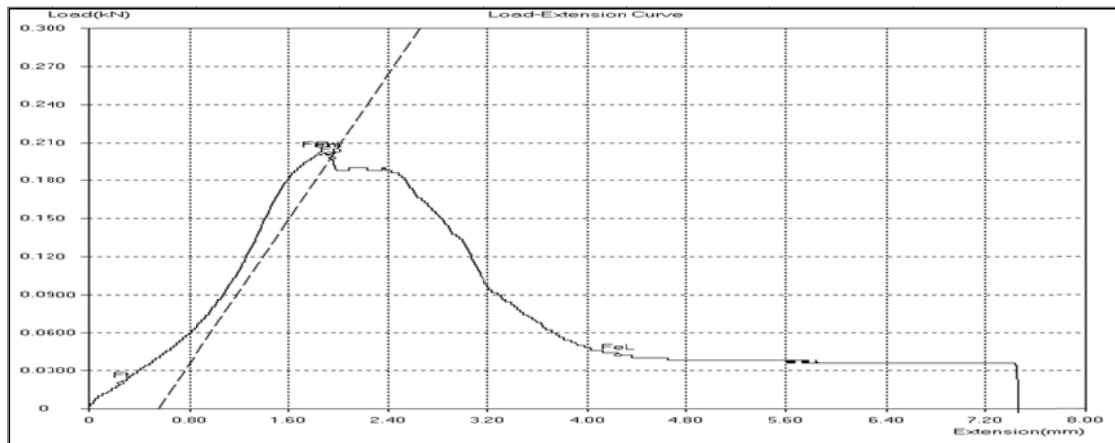
(B) 1.5%SiO₂/PVA



(C) 2.5%SiO₂/PVA

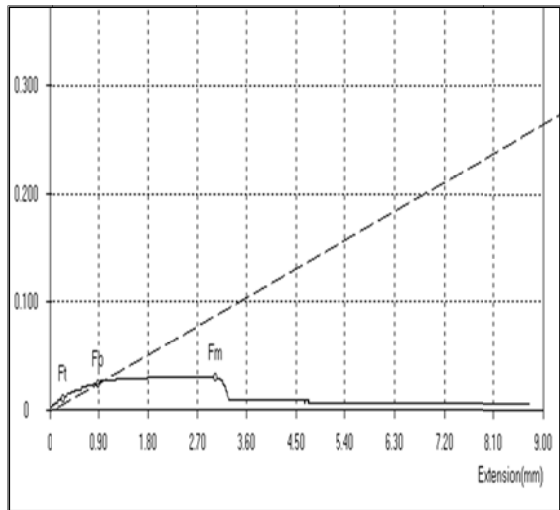


(D) 3.5%SiO₂/PVA

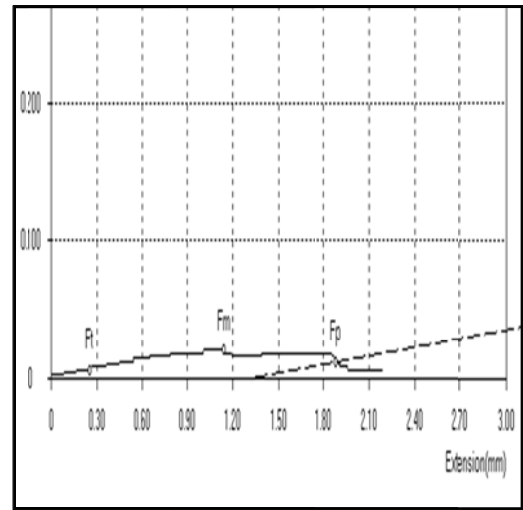


(E) 4.5%SiO₂/PVA

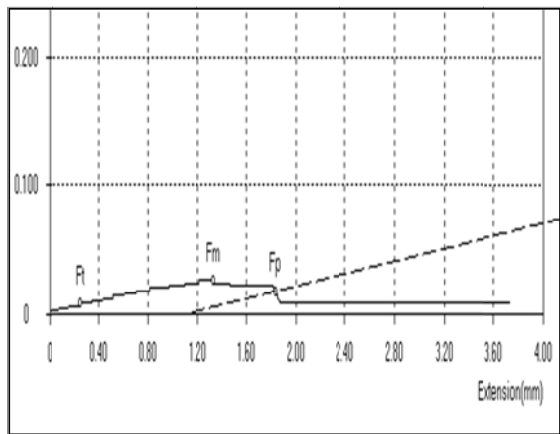
Fig. 16: Load-extension curves of vulcanized rubber samples.



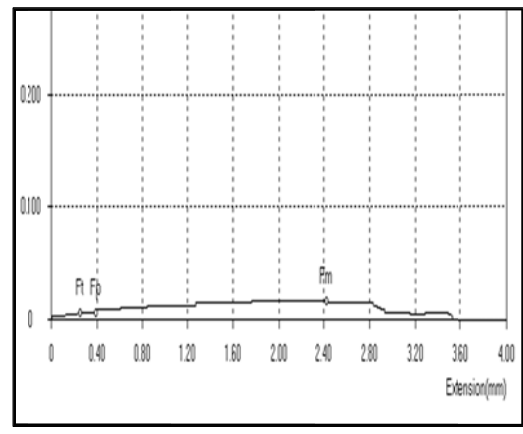
(A) PVA



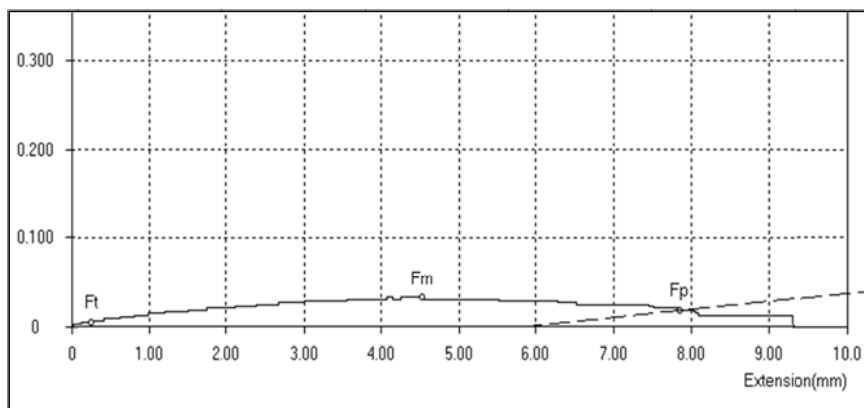
(B) 1.5%SiO₂\PVA



(C) 2.5%SiO₂\PVA



(D) 3.5%SiO₂\PVA



(E) 4.5%SiO₂\PVA

Fig. 17: Load-Extension curves of cartoon sample.

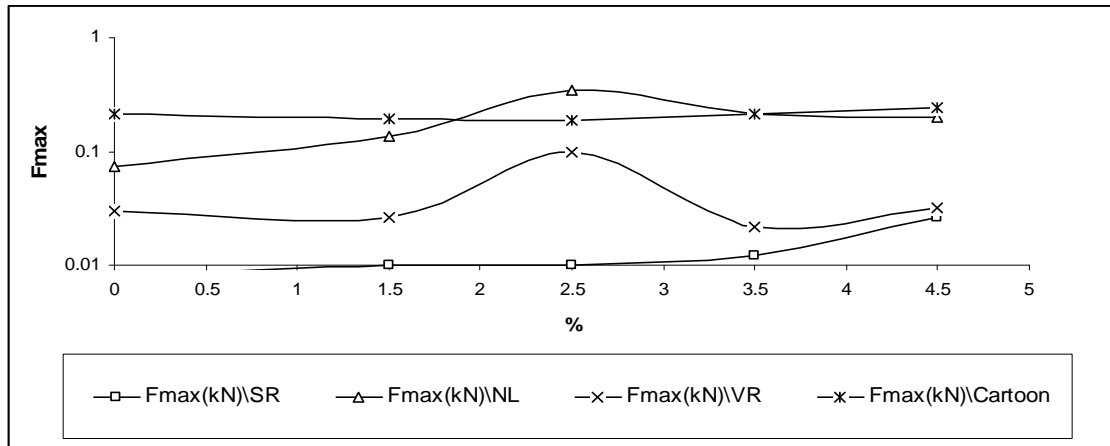


Fig. 18: Shear strength of the samples involved.

Table 1: Main mechanical properties of SR, NL, VR, cartoon samples for $F_{max}(kN)$.

Sample	Mechanical Properties			
	Fmax (kN)\SR	Fmax (kN)\NL	Fmax (kN)\VR	Fmax (kN)\Cartoon
Standard	0.018	0.376	0.108	0.789
0%	0.014	0.064	1.026	0.310
1.5%	0.016	0.186	0.036	0.508
2.5%	0.016	2.206	0.048	0.680
3.5%	0.016	0.222	0.04	0.352
4.5%	0.018	0.228	0.05	0.116

Table 2: Main mechanical properties of SR, NL, VR, cartoon sample for ultimate tensile strength (MPa).

Sample	Mechanical Properties			
	Ultimate tensile strength (MPa)\SR	Ultimate tensile strength (MPa)\NL	Ultimate tensile strength (MPa)\VR	Ultimate tensile strength (MPa)\Cartoon
Standard	0.482	18	3	13.5
0%	0.482	3	0.49	5.15
1.5%	0.24	8	1.02	8.36
2.5%	0.44	8	1.04	11.3
3.5%	0.44	9	1.44	5.88
4.5%	0.5	10	1.5	1.94

Table 3: Main mechanical properties of SR, NL, VR, cartoon sample for shear strength.

Sample	Mechanical Properties			
	Fmax (kN)\SR	Fmax (kN)\NL	Fmax (kN)\VR	Fmax (kN)\Cartoon
0%	0.008	0.074	0.03	0.212
1.5%	0.01	0.134	0.026	0.192
2.5%	0.01	0.346	0.1	0.188
3.5%	0.012	0.212	0.022	0.212
4.5%	0.026	0.202	0.032	0.244

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