

Study Mechanical and Chemical Properties of prepared Starch – Urea Adhesive Resin

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Abstract

The force of urea-starch origin adhesive was studied on the (wood-wood) surfaces in all experimental to know the convenient it in wood industries. We found the maximum shear stress (0.543N/mm²), Elongation (1.41%) and modulus (2 N/mm²) when curing temperature (25oC) for curing time (7 days). Also the adhesive was cured at temperature (160oC) for different curing time (10, 20, 30, 50 Min.). The result was found the maximum shear stress (0.896N/mm²), Elongation (1.7%) and modulus (1.3 N/mm²) at curing time (10Min.). The effective of adhesive by immerse in water and absolute ethanol also studied. The more effected in adhesive was by water and was increasing the swelling with increased the time. When immerse the adhesive materials in ethanol was very limited affected and the swelling was nearly stable in the period of time from (10Min.) to (50Min.). The weight loss with time was studied at the temperatures of 160 oC. The amount of weight reduction was increased with the time.

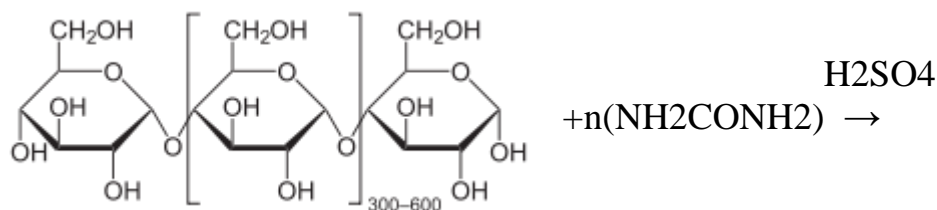
Key words: Starch, Urea, Wood, Adhesive .

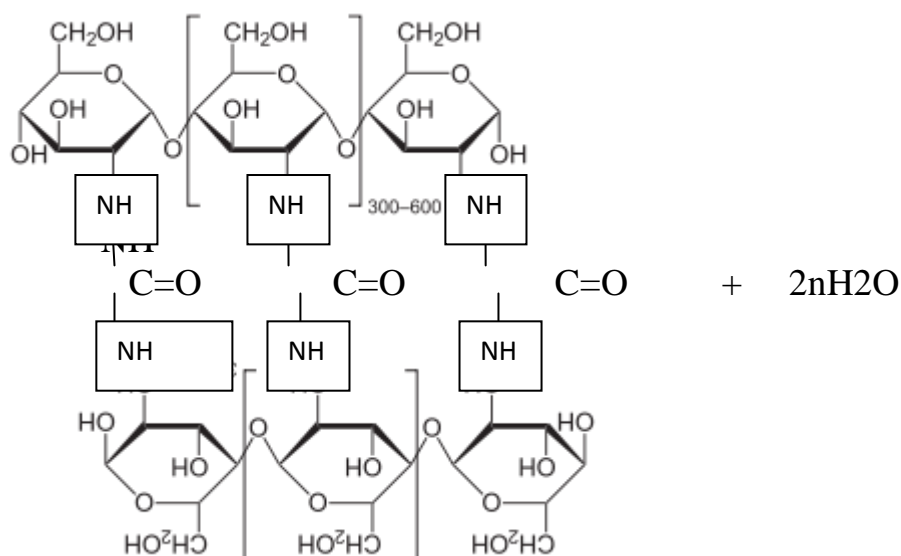
الخلاصة

حضر لاصق نشأ- يوريا ودرست قوته علي سطح الخشب في كل تجارب هذا البحث لمعرفة صلاحية استخدامه في الصناعات الخشبية. ووجد ان اكبر قوة شد هي (٠,٥٤٢٥٠١ نيوتن/ملم^٢) و استطالة (١,٤١ %) و معامل المرونة هو (٢ نيوتن/ملم^٢) عند معالجة اللاصق في درجة حرارة (٢٥ سليزية) ولفترة زمنية مقدارها (٧ أيام). وكذلك عالج اللاصق عند درجة حرارة (١٦٠ سليزية) ولفترات زمنية مختلفة هي (١٠ ، ٢٠ ، ٣٠ ، ٥٠ دقيقة). وجد ان أعظم قوة شد كانت (٠,٨٩٥٧٥٥ نيوتن/ملم^٢) باستطالة مقدارها (١,٧ %) ومعامل مرونة (١,٣ نيوتن/ملم^٢) عند معالجة اللاصق لفترة زمنية مقدارها (١٠ دقيقة). ودرس كذلك مدى تأثير اللاصق عند غمرة بالمذيبات الكيميائية (الماء والكحول الايثيلي المطلق) فوجد ان أكبر تأثير لللاصق كان في الماء حيث يزداد انتفاخ اللاصق مع الزمن، بينما في الكحول الايثيلي كان تأثيره جدا محدود و الانتفاخ تقريبا ثابتا لفترة زمنية من (١٠ دقيقة) الى (٥٠ دقيقة). وايضا درس مدى نقصان وزن اللاصق مع الزمن عند درجة حرارة (١٦٠ سليزية)، فوجد زيادة في نقصان الوزن مع الزمن.

Introduction

The starch chemical formula (C₆H₁₀O₅)_n is a polysaccharide carbohydrate consisting of a large number of glucose units joined together by bonds. The starch react with urea by active groups in starch (-OH) and urea (-NH₂) as shown in the below1.





There has been considerable interest in the starch for enhancing the adhesion, and previous studies have included the wood adhesive was developed by cross linking corn-starch and poly(vinyl alcohol) (PVOH) with hexamethoxy-methylmelamine (Cymel 323). Citric acid was used as a catalyst and latex (UCar 443) was added to improve moisture resistance². The interfacial reaction of the polyethylene (PE)/starch blend system containing the reactive compatibilizer maleated polyethylene (m-PE) was directly characterized by Fourier transform infrared (FTIR) spectroscopy³. The effects of starch origin (potato, corn, and rice starches) and hypochlorite level (NaOCl , 0.8% and 2% w/w) on the structures and physicochemical properties of oxidized starches were investigated. Oxidized starch batters exhibited greater adhesions than did native starch batters, with rice starch batter exhibiting the greatest adhesion⁴. The surface energies were used to estimate the starch/polymer interfacial energy and work of adhesion⁵. Kinetic studies of gelatinization of cornstarch adhesive under conditions simulating those at the corrugator revealed that the rate of advance of the gelatinization front in the starch film was linear with square root of time⁶. The experiment of bacteria adhesion onto starch granules is conducted. It is found that anaerobic saccharolytic bacteria have the highest adhesion ability in their growth and initial stage of stationary phase⁷. A series of starch graft copolymers were prepared from commercial cornstarch products and vinyl monomers and were evaluated as adhesives in pigmented paper coatings⁸. The extension of a centrifuge technique to measure adhesion and friction forces to an ultracentrifuge has been described. The equipment and procedure provide many experimental possibilities of which the adhesion of single particles to flat compacted powder surfaces has been used to measure the adhesion and friction force of starch microspheres to microcrystalline cellulose⁹. Contact angles of water and other liquids were measured on films and extruded ribbons of normal, high amylose and waxy cornstarch using a Chan dynamic contact angle analyzer¹⁰. The effect of processing of cassava methods on the suitability of cassava starch produced for adhesive formulation were

investigated. Out of the three processing methods, the one without steeping gave the best starch for adhesive production in terms of the physico-chemical properties such as swelling power¹¹. The stability of 70 : 24 : 6 w/w/w blends of a lactic acid-based hot melt adhesive (LHM), oxidized potato starch (dried or nondried), and polyethylene glycol (PEG) was studied¹². In the wood processing industry considerable amounts of starch products are used as extender for the gluing with synthetic resin adhesives based on aminoplasts (urea and melamine resins)¹³. Prepare thin-boiling and no gelling adhesives, granular wheat and corn starches were thinned by treatment with aqueous hydrochloric acid or sodium hypochlorite and then hydroxypropylated with propylene oxide¹⁴. A combination of soy flour and isolated soy protein was investigated as the adhesive in pigmented paper coatings that contained dialdehyde starch (DA) as the insolubilizing agent¹⁵. Functionalized metallocene copolymers synthesized from ethylene with 5-hexen-1-ol and ethylene with 10-undecen-1-ol were used as compatibilizers in LDPE/starch and LDPE/dextran blends in order to improve the interfacial adhesion between hydrophobic LDPE and hydrophilic natural polymers¹⁶. The influences of inorganic phosphates, as residuals and/or impurities within starch, on the adhesion of acid-treated cornstarch phosphates to cotton and polyester fibers were investigated for warp sizing¹⁷. The influence of starch carbamoyl ethylation upon the adhesion capability of starch to pure cotton, all polyester, and polyester/cotton blend fiber substrates were investigated¹⁸. The develop a new route for surface functionalization of biodegradable polymers. The method is based on a wet chemistry modification, resulting in etching and/or hydrolysis in order to increase the amount of polar groups, such as hydroxyl ($-\text{OH}$) and carboxylic ($-\text{COOH}$) groups on the surface of the polymer. The polymer used as substrate was a corn starch-ethylene vinyl alcohol biodegradable blend¹⁹.

Experimental

The adhesive content is:

Material	Amount
Starch	50 (g.)
Urea	12 (g.)
Water	420 (ml)
Sulfuric acid(H_2SO_4) (29% W)	1 (Cm ³)

The procedure for attended Starch – Urea adhesive was as following:-

- (1) Add urea to the (140 ml) of water in a pot and heating them to the boiling point temperature. After that the sulfuric acid (H_2SO_4) was added to the mixture.
- (2) Mix starch with (280 ml) of water at room temperature and well stirred until the mixture be homogenous.
- (3) Continuously, add few mixture of starch-water to the mixture of hot urea-water and continuous with well stirring and heating at boiling point temperature until all amount of starch-water mixture finish.

- (4) Lift the adhesive from supplying heating source and let it cooling to room temperature after that we can use it in specimen.
- (5) The wood specimen attended according to the ASTM (D1002) for testing shear stress.
- (6) Let the specimens drying at room temperature for 1 day.
- (7) The specimens were cured at the temperatures and times requirement.
- (8) The specimens leave to cooling at room temperature, after that it makes convenient tests.

Discussion

The force of urea-starch adhesive was studied (by apparatuses Gunt HAMBURG, WP300.20, made in Germany) on the wood surface in all experimental to know the convenient it in wood industries .The adhesive was studied on the wood surface at curing temperature (25oC) for curing time (7 days) to dry it. We found the maximum shear stress (0.542501N/mm²), Elongation (1.41%) and modulus (2N/mm²) as show in fig.(1) and table (1). It should be noted that in uniaxial loading the stress(S), S, is simply the force (F) divided by the cross-sectional area (A) and the strain, E, is the deformation (L) divided by the original length (L_o). The modulus (M) of the material is then the ratio of stress/strain²⁰.

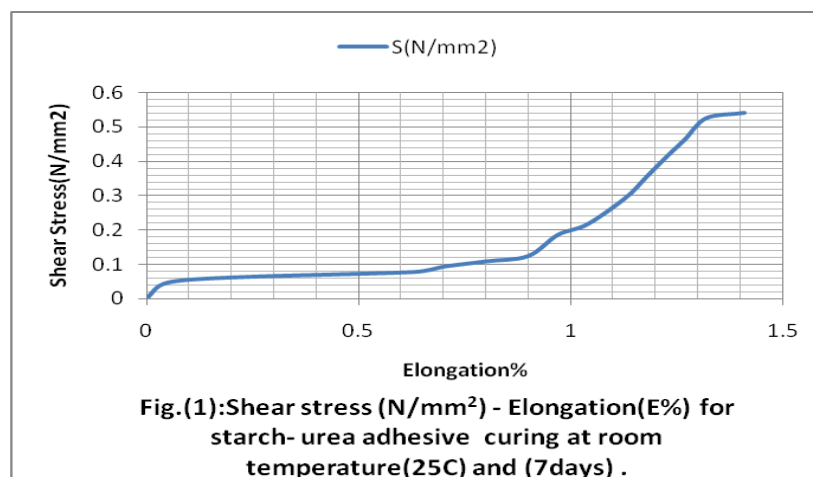
$$\text{Stress, } S = \frac{F}{A}$$

$$\text{Strain(Elongation), } E = \frac{L}{L_o}$$

$$\text{Modulus, } M = \frac{S}{E}$$

Table(1):Shear Stress (N/mm²) – Elongation(%) for starch- urea adhesive curing at room temperature (25 o C) and time(7days).

E[%]	0	0.05	0.21	0.63	0.7	0.8	0.9	0.97	1.04
S(N/mm ²)	0	0.0465	0.062	0.0775	0.093	0.1085	0.124	0.186	0.217
E[%]	1.13	1.18	1.23	1.27	1.32	1.41			
S(N/mm ²)	0.2945	0.3565	0.4185	0.465	0.527	0.5425			



It is found that the adhesive behavior as elastomeric materials²¹ as shown in fig.(1).

When adhesive cure at temperature (160°C) and curing time (10 min.) on the wood surface, it is found the maximum shear stress (0.895755N/mm²), Elongation (1.7%) and modulus (1.3N/mm²) As shown in fig.(2) and table (2) below. Also we can seen from fig.2 the adhesive materials behavior as elastomeric polymer²¹.

Table (2): Shear stress (N/mm²) vs. Elongation (E%) for starch- urea adhesive curing at temperature(160°C) and time(10Min.).

S(N/mm ²)	E[%]	S(N/mm ²)	E[%]
0	0	0.699318	0.86
0.03143	0.04	0.715033	0.96
0.06286	0.08	0.746463	1
0.09429	0.12	0.762178	1.05
0.15715	0.17	0.777893	1.12
0.235725	0.22	0.793608	1.17
0.290728	0.26	0.793608	1.22
0.353588	0.3	0.809323	1.26
0.432163	0.35	0.825038	1.32
0.510738	0.39	0.840753	1.36
0.573598	0.44	0.856468	1.42
0.589313	0.48	0.856468	1.47
0.605028	0.54	0.872183	1.52
0.620743	0.63	0.88004	1.56
0.652173	0.74	0.88004	1.61
0.667888	0.78	0.895755	1.65
0.683603	0.82	0.895755	1.7

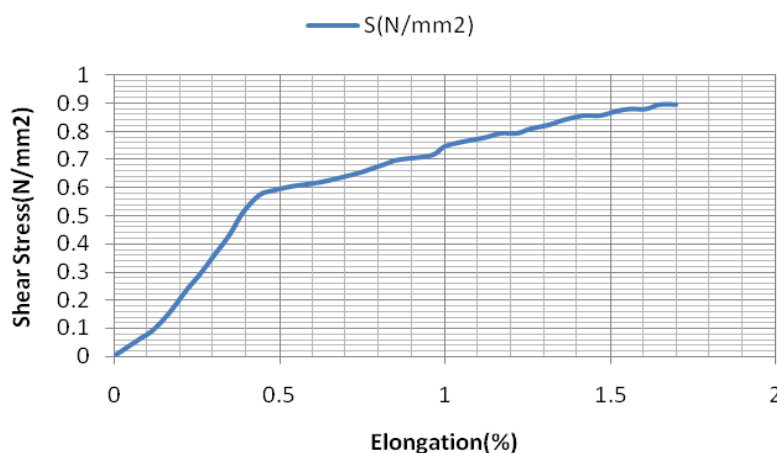
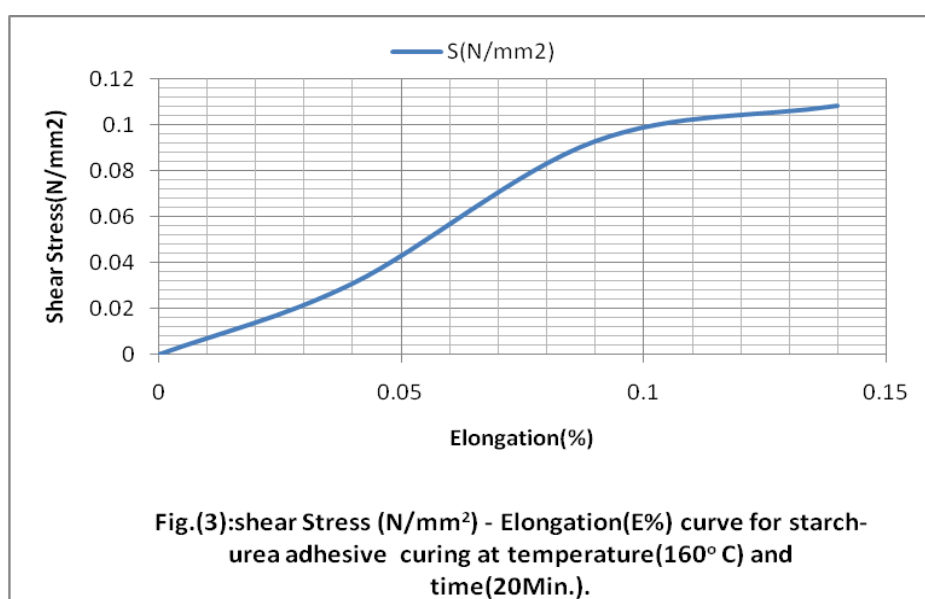


Fig.(2)Shear Stress (N/mm²) - Elongation(E%) curve for starch- urea adhesive curing at temperature(160°C) and time(10Min.).

When curing adhesive at temperature (160°C) and curing time (20Min.) was found the shear stress of adhesive decrease to the (0.1085N/mm²) and also the elongation decrease to the (0.14%) and the modulus was (1.07N/mm²).The high decreasing in shear stress and elongation because the degradation in adhesive materials as shown in Fig.(7) and table (7). From Fig.(3) and table(3) the adhesive materials behavior as elastomeric materials²¹.

Table(3):Shear Stress (N/mm²) vs. Elongation(E%) for starch- urea adhesive curing at temperature(160° C) and time(20Min.).

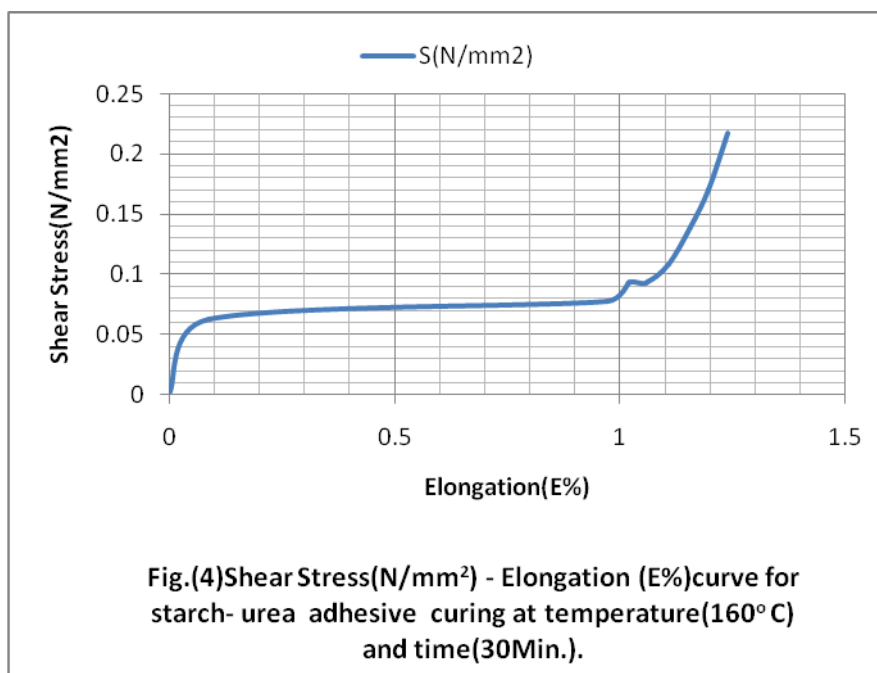
S(N/mm ²)	0	0.031	0.093	0.1085
E[%]	0	0.04	0.09	0.14



When adhesive cure at temperature (160°C) and curing time (30 min.) on the wood surface, it was found the maximum shear stress (0.217N/mm²), Elongation (10.33%) and modulus (2.5N/mm²) As shown in fig.(4) and table (4) below. Also we can seen from fig.2 the adhesive materials behavior as elastomeric polymer²¹.

Table(4) shear Stress (N/mm²) vs. Elongation(E%) for starch- urea adhesive curing at temperature(160° C) and time(30Min.).

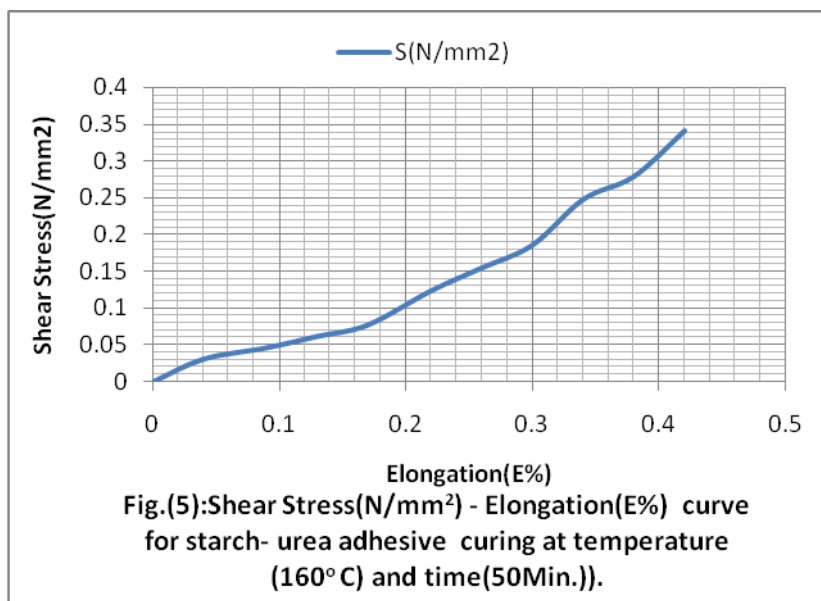
S(N/mm ²)	E[%]
0	0
0.062	0.088
0.0775	0.972
0.093	1.02
0.093	1.06
0.1085	1.108
0.1395	1.156
0.1705	1.196
0.217	1.24



The adhesive cure at temperature (160oC) and curing time (50 min.) on the wood surface and the result was the maximum shear stress (0.341N/mm²), Elongation (0.42%) and modulus (0.75N/mm²) As shown in Fig.(2) and Table (2) below. Also we can see from Fig.5 the adhesive materials behavior as brittle polymer²¹.

Table(5):Shear Stress (N/mm²) vs. Elongation (E%)for starch- urea adhesive curing at temperature(160o C) and time(50Min.).

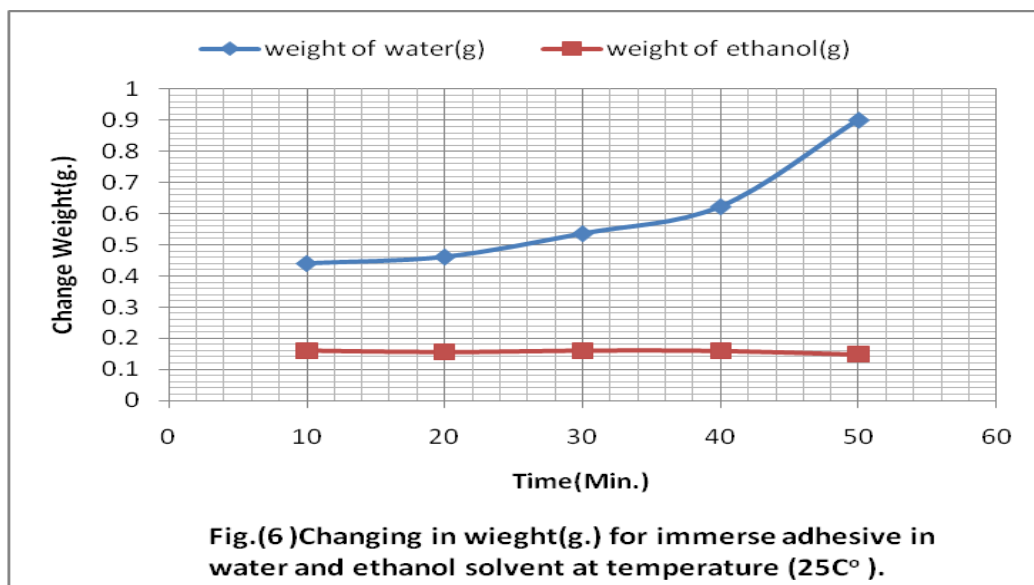
S(N/mm ²)	E[%]
0	0
0.031	0.04
0.0465	0.09
0.062	0.13
0.0775	0.17
0.124	0.22
0.155	0.26
0.186	0.3
0.248	0.34
0.279001	0.38
0.341001	0.42



The effective of adhesive by immerse in water and absolute ethanol also studied. The more effected in adhesive was by water and was increasing the swelling with increasing the time as shown in Fig.(6) and Table(6) below. But when immerse the adhesive materials in ethanol it was very limited effected and the swelling was nearly stable in the period of time from (10Min.) to (50Min.) as shown in Fig.(6) Table (6).

Table(6):Changing in weight(g.) for immerse adhesive in water and ethanol solvent at temperature (25Co).

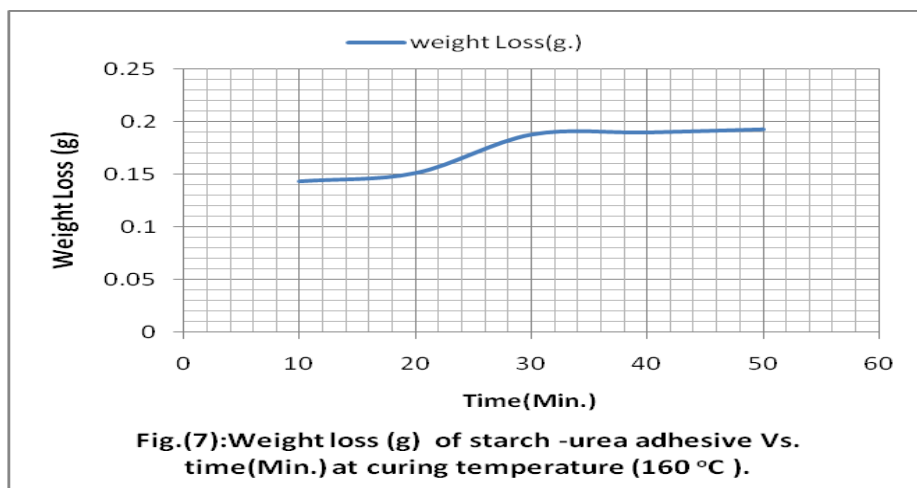
Ethanol weight (g)	Water weight (g)	Time(min.)
0.16	0.44	10
0.156	0.461	20
0.16	0.536	30
0.159	0.623	40
0.149	0.902	50



The weight loss versus time was studied at the temperatures of 160 oC. It can be seen that the amount of weight reduction increases with the time as shown in Fig.(7) and Table (7). Also the best result of adhesive strength at curing temperature (160 oC) in (10Min.) that because less cracking in adhesive with more oriented the chains of polymer²² as shown in Fig.(7) and Table(7).

Fig.(7):Weight loss (g) of starch -urea adhesive Vs. time(Min.) at curing temperature (160 oC).

Weight Loss (g.)	Time(Min.)
0.143	10
0.151	20
0.188	30
0.19	40
0.193	50



Conclusion

The maximum shear stress and elongation was found at curing temperature (160 oC) and curing time (10Min.) and they equal to the (0.896N/mm²), (1.7%) respectively.

Very limited effected was found in adhesive material when immerse adhesive material in absolute ethanol. But affected it in water solvent was greater effected and swelling increase with time increasing.

The adhesive degraded when left it in temperature (160 oC) and the degradation was increased with the time.

References

- (1) Thomas M. Devlin "text Book of Biochemistry with clinical correlations"6th edition, wiley-liss, P=650, 2006.
- (2) Syed H. Imam, Lijun Mao, Liang Chen , Richard V. Greene" Wood Adhesive from Crosslinked Poly(Vinyl Alcohol) and Partially Gelatinized Starch: Preparation and Properties" Starch , Volume 51 Issue 6, Pages 225 – 229,1999.
- (3) Seong Ik Yoo , Tae Yeon Lee , Jin-San Yoon , Ik-Mo Lee , Mal-Nam Kim , Han Sup Lee" Interfacial adhesion reaction of polyethylene and starch blends using maleated polyethylene reactive compatibilizer",Journal of Applied Polymer Science,Volume 83 Issue 4, Pages 767 – 776 ,2001.
- (4) Daris Kuakpetoon, Ya-Jane Wang" Characterization of Different Starches Oxidized by Hypochlorite",Starch ,Volume 53 Issue 5, Pages 211 – 218,2001.
- (5) G. Biresaw , C. J. Carriere" Correlation between mechanical adhesion and interfacial properties of starch/biodegradable polyester blends"Journal of Polymer Science Part A: Polymer Chemistry,Volume 39 Issue 9, Pages 920 – 930,2001.
- (6) M. Inoue, P. Lepoutre" Kinetics of gelatinization of cornstarch adhesive",Journal of Applied Polymer Science,Volume 31 Issue 8, Pages 2779 – 2789,2003.
- (7) Hsi-Hua Wang , Tsyy-Wen Chiou, Jyh-Ping Hsu" Anaerobic saccharolytic bacterial adhesion to raw starch granules", Biotechnology and Bioengineering,Volume 29 Issue 9, Pages 1122 – 1126,2004.
- (8) M. E. Carr , Peoria" Preparation and Application of Starch Graft Poly(vinyl) Copolymers as Paper Coating Adhesives",Starch,Volume 44 Issue 6, Pages 219 – 223,2006.
- (9) Fridrun Podczek, J. Michael Newton" Development of an ultracentrifuge technique to determine the adhesion and friction properties between particles and surfaces",Journal of Pharmaceutical Sciences,Volume 84 Issue 9, Pages 1067 – 1071, 2006.
- (10) J. W. Lawton," Surface Energy of Extruded and Jet Cooked Starch",Starch ,Volume 47 Issue 2, Pages 62 – 67,2006.
- (11) Samson O. Agboola, John O. Akingbala, Gbikeloluwa B. Oguntimein," Starch,Volume 42 Issue 1, Pages 12 – 15,2006.

- (12) Saara Inkinen , Mikael Stolt, Anders Södergård" Stability studies on blends of a lactic acid-based hot melt adhesive and starch",Journal of Applied Polymer Science,Volume 110 Issue 4, Pages 2467 – 2474,2008.
- (13) Lore Plath," The Role of Starch Products in Glueing of Wood Materials by Synthetic Resin Adhesives",Starch ,Volume 24 Issue 9, Pages 306 – 312,2006.
- (14) Koo Min Chung , Professor Paul A. Seib , " Thin-Boiling and Nongelling Adhesive Prepared from Maize and Wheat Starches", Starch,Volume 43 Issue 11, Pages 441 – 446,2006.
- (15) F. B. Weakley, M. E. Carr, C. L. Mehlretter," Dialdehyde Starch in Paper Coatings Containing Soy Flour-Isolated Soy Protein Adhesive",Starch,Volume 24 Issue 6, Pages 191 - 194, 2006.
- (16) Ana María Domínguez, Raúl Quijada, Mehrdad Yazdani-Pedram" Use of Functionalized Metallocene Copolymers from Ethylene and Polar Olefins as Compatibilizers for Low-Density-Polyethylene/Starch and Low-Density-Polyethylene/Dextran Blends", Volume 291 Issue 8, Pages 962 – 971,2006.
- (17) Zhifeng Zhu , Zheqiong Cheng," Effect of Inorganic Phosphates on the Adhesion of Mono-phosphorylated Cornstarch to Fibers",Starch,Volume 60 Issue 6, Pages 315 – 320,2008.
- (18) Zhifeng Zhu , Peihua Chen," Carbamoyl ethylation of starch for enhancing the adhesion capacity to fibers",Journal of Applied Polymer Science,Volume 106 Issue 4, Pages 2763 – 2768,2007.
- (19) I. B. Leonor , H.-M. Kim , F. Balas , M. Kawashita , R. L. Reis , T. Kokubo , T. Nakamura," Alkaline treatments to render starch-based biodegradable polymers self-mineralizable",Journal of Tissue Engineering and Regenerative Medicine,Volume 1 Issue 6, Pages 425 – 435,2008.
- (20) R.J. Crawford,"PLASTICS ENGINEERING", P=62, 3rd edition, Butterworth-Heinemann, Oxford,UK,2002.
- (21) Lawrence E. Nielsen, Robert F. Landel"Mechanical properties of polymers and composites",2nd edition, Marcel Dekker, Inc., 1994.
- (22) Ana A. Taker,Trans.By Dr.Akram A. Mohammed"Physical Chemistry For Polymer",page=224-228, Mosal university editions,Mosal,Iraq,1984.