Study and optimization of the mechanical properties of PVP/PVA polymer nanocomposite as a low temperature adhesive in nano-joining

Ahmed A. Taher¹, Ayad M Takhakh² and Sabah M Thahab³

¹ Department of Mechanical Engineering, University of Kufa. E-mail: ahmed.abosabeeh@uokufa.edu.iq

² Department of Mechanical Engineering, Al-Nahrain University. E-mail: ayadtakak@yahoo.com

³ Department of Physics, Faculty of Science, University of Kufa. E-mail: sabah.alabboodi@uokufa.edu.iq.

ABSTRACT

This paper presents a study of the effects of temperature, curing time and silver nano-particles (AgNPs) on the concentration of the PVP/PVA nano composites on shear force of adhesive nano-joining (ANJ). Low temperature (LT) ANJ of copper–copper (Cu–Cu) at the temperature values of 200, 215, 225, 240 and 275°C and curing times of 10, 20 and 30 minutes, under fixed pressure (5 MPa) are investigated. Higher shear strength value is found at a temperature value of 227 °C and time extent of 10 minutes. It is noted that for an AgNPs concentration of (5%wt) a maximum value of shear strength (2180N) is reached. These results indicate that resistance will increase with raising of the heat up to 227 °C, then followed by a decrease in strength up to 275 °C at a curing time of 10 minutes. The results have been optimized with Neural Network (NN) for accuracy.

KEYWORDS: Adhesive, Polyvinylpyrrolidone, Temperature, silver, time.

1. INTRODUCTION

The use of a low temperature joining technique (LTJT) has been proposed as a possible alternative to high-temperature applications and has been discussed by Schwarzbauer [1], and others in connection die attachment in power electronics in the late 1980s [2].

There are many advantages connected with the use of Ag particles in LTJT. Silver gives better thermal and electrical conductivities than Sn-Pb or Pb-free joints. Nano-particles occur at temperatures less than the melting temperature of Ag, typically 0.2–0.4 Tm. Once sintered, the Ag joint will have a melting temperature similar
to bulk Ag (961 °C). Based on these properties, nano-Ag has been listed in recent studies as a leading candidate for lead-free die attachment as well as die-attachment for silicon carbide technologies [3].

Our test of mechanical properties of adhesives was conducted at variable temperatures from 200 to 275°C. The aim of the work was to improve the shear strength of adhesive joints using AgNPs as additive materials in the polymer solution, mixed of PVP/PVA nanocomposites, and to examine its effect on the shear strength of adhesive nano-joining of Cu–Cu metals joints.

2. EXPERIMENTAL WORK

2.1. Materials

PolyvinylAlcohol (PVA) with MW of 1750, Polyvinyl pyrrolidone (PVP, K30) with MW of 40,000 and silver nanoparticle powder AgNPs (50 nm) were used in this work as base materials. Polymers with viscosity values of 32 and 26 cps were measured using viscosity measurement instruments for (0.2 M) PVA and (0.008 M) PVP polymer solution respectively. Polymer melting point Tm values of 172°C and 192 °C were measured for PVP and PVA polymers, respectively.

The preparation of adhesive polymers with concentrations of 0.008 M of PVP-K30 and 0.02M of PVA were obtained by dissolving polymers at weights of 16gm and 8gm, respectively, in 50 ml distilled water (DW). The solutions were mixed with a magnetic stirrer for 30 minutes at room temperature for the PVP polymer solution and for 15 minutes at 60 °C for the PVA polymer solution. After this, we performed mixing of the silver and polymers to make adhesive materials.

\[
C_x = n_x V \quad \ldots \ldots \quad (1)
\]

\[
n_x = \frac{g M W}{V} \quad \ldots \ldots \quad (2)
\]

Where: \(C_x\) is the concentration mol. L-1, and \(n_x\) is the number of moles.

\(V = 50 \text{ ml volume of solvent}, \ g = 8 \text{ gm.} \) is the mass of PVP

2.2. Preparation of (Cu–Cu) metal adhesive joining

The single shear lap joints were Copper (Cu) samples with dimensions 25.4 mm x 1.6 mm x 90 mm, having a contact area of 21.7mm x 25.4mm), which were prepared to ASTM D1002 [4]. Figure 1 illustrates.

The (Cu–Cu) metal specimens were pressed at pressure 5 Mpa with heating temperature values of 200, 215, 225, 240, and 275°C [5] for 10 minutes using a press under the heating system, as shown in Figure 2. A pressure of 5 Mpa was chosen in light of published research [6]. The temperature range (200–275 °C) was selected according to the adhesive’s material properties, whereby the melting point of PVP is 172°C and PVA is 192°C.
according to the melting temperature test, while the decomposition temperature reaches 483°C according to the TGA test, so required temperature is within this range.

A mixture 50% from PVP/PVA with nano silver (5%) and a curing time of 10 minutes were used; increasing the curing time above 10 minutes produced weakening and burning of the adhesive material, formation of a black adhered region and cracks in the adhesive material structure as well as reduction in shear strength [7].

![Figure 1. Sample according to ASTM D1002 -01](image1)

![Figure 2. Copper adherents after pressing](image2)

### 2.3 Shear force test

A shear force test of the joint was conducting and measured using a universal tensile testing machine according to ASTM –D1002, with a rate of 1 mm/min-1 , shown in Figure 3.

![Figure 3. Shear test](image3)

An artificial neural network of MATLAB 2015 (ANN) can be used in study of the mechanical properties of different materials, including adhesive materials. In this work, the ANN approach has been used for predicting certain mechanical properties of PVP/PVA with silver as an adhesive material. These properties include shear effect and are tested under combined independent situations that include curing time, sintering temperature and concentration of silver nanoparticles.

### 3. RESULTS AND DISCUSSIONS

Figures 4 and 5 show the thermo gram analysis (TGA) and (DSC) of PVP and PVA respectively, which has been plotted with weight loss as a function of the temperature .These measurements were
conducted for PVP and PVA precursor with a heating rate of 10 °C/min in the temperature range of (40 to 800 °C) for PVP polymer and (50 to 350 °C) for PVA polymer.

It is clear that for PVP polymer the TGA initial weight loss curve is 12% in the temperature range 40–87.9 °C, this is due to loss of OH content. In the differential thermal analysis (DTA) curve, two exothermic peaks were observed at 483.92 °C and 678.08 °C, respectively. The sharp and strong exothermic peak at 483.92°C is due to the combined effect of combustion of organic residuals and
the decomposition of PVP and which is well above the heating temperature employed in the present work. These results are close to those obtained by [8].

Table 1 shows the results of shear force with temperature and ratio (PVP/PVA) at pressure 5 MPa. and curing time 10 minutes.

Table 1 shear force with PVA/PVP

| T (OC) | PVP  | 25%PVA | 50%PVA | 75%PVA | PVA  |
|--------|------|--------|--------|--------|------|
| 200    | 590  | 1002   | 995    | 450    | 510  |
| 215    | 575  | 995    | 1020   | 480    | 490  |
| 225    | 490  | 1320   | 1210   | 720    | 498  |
| 240    | 445  | 1120   | 1250   | 680    | 620  |
| 265    | 0    | 0      | 920    | 715    | 515  |

Our results show that the highest value for shear load was reached at (50%PVP and 50%PVA) which has better distribution in composite structure as shown in Figure 6 and has a dynamic viscosity of 37cP, before and after this ratio (50% PVP/PVA) a nonidentical structure in adhesion zone, caused a decrease in shear force. [9]

![Figure 6. Shear force with temperature.](image)

Table 2 shows the results of shear force with treating temperature, force and concentration of nano silver at pressure 5 MPa.
Table 2. Shear force with concentration of nano silver

| T(°C) | 5%  | 10%  | 15%  | 20%  |
|-------|-----|------|------|------|
| 200   | 1330| 1030 | 1120 | 1040 |
| 215   | 1450| 1000 | 1050 | 1011 |
| 225   | 2180| 525  | 1300 | 623  |
| 240   | 1800| 515  | 1200 | 655  |
| 265   | 700 | 1900 | 1022 | 850  |

This table reveals the best result for shear load at 5% silver, that distribution in composite structure and viscosity 39sc improves with growing concentration (10%, 15% and 20%), leading to adhesive agglomeration when offered to temperature, leading to a nonidentical structure in the adhesion zone, causing a drop in shear force [10] as shown in Figures 7 and 8.

![Fig. 7 shear force with Temp with Nano silver](image)
After this concentration (5%), high viscosity from nano particles causes irregular distribution and weak region and cracks in structure.

Many attempts have found that the best structure of the neural networks for predicting shear loads are the two hidden layer structure, which can be coded as 2-6-7-1. For shear load it is clear that the best training performance is 0.0011663 and 0.99874 for training and 0.99841 for testing samples respectively, as shown in Figure 9) and Figure 10.

Figure 8. shear force with Temperature with and without nano-silver

Figure 9. Training and testing prediction data, PVP/PVA with silver of two hidden layers
Table 3 shows the summary of curing time, temperature, experimental shear force and predicted natural shear force.

Table 3. Experimental and predicted natural shear force.

| Time(min.) | Temp. (°C) | Exp.shear force | Nur.shear force | Err% |
|------------|------------|-----------------|-----------------|------|
| 10         | 200        | 1330            | 1287.833        | 3.170468 |
| 10         | 215        | 1450            | 1465.682        | -1.08149 |
| 10         | 225        | 2180            | 2167.74         | 0.562382 |
| 10         | 240        | 1800            | 1801.62         | -0.09001 |
| 10         | 275        | 700             | 703.8215        | -0.54594 |
| 20         | 200        | 1030            | 1036.46         | -0.62718 |
| 20         | 215        | 1000            | 986.5146        | 1.348544 |
| 20         | 225        | 525             | 549.3018        | -4.62891 |
| 20         | 235        | 515             | 522.8684        | -1.52784 |
| 20         | 250        | 1900            | 1882.917        | 0.89908 |

There is a good result between the experimental and neural networks (ANN) because there is convergence between the training and testing result as shown in Figure 11.
4. CONCLUSIONS

The effect of temperature of adhesive materials on the shear force is show a slight fittings, from 200 to 227°C, however at a 227 to 250°C an increase in the heating time gave little in terms of the shear strength of lap joints. A good value of adhesive joint force was obtained for 50%PVP and 50%PVA at 10 min and 227°C because a viscosity of PVP /PVA increased slightly in direct proportion from 200 to 225°C but decreased during the range 225- to 275°C. A good shear force occurred with 5% nano silver; there are non-identical in the microstructure of adhesive joints for concentration of silver for 10, 15, 20%. There was a degradation of the shear force of lap joints at temperature 275°C, due to crack formation in the microstructure of the adhesive joints.

5. REFERENCES

1. Amandine, M., Wissam, S., Rapha, R., "Die attach using silver sintering. Practical implementation and analysis", European Journal of Electrical Engineering (2013), No. 3-4, p. 293–305.

2. Al-Shammari, B., Al-Fariss, T., Al-Sewailm, F. and Elleithy, R. “The Effect of Polymer Concentration and Temperature on the Rheological Behavior of Metallocene Linear Low Density Polyethylene (mLLDPE.) Solutions” (2011), Journal of King Saud University, Engineering Sciences, 23(1), p. 9–14.

3. Kim S, “Mechanical properties of nano-silver joints as die attach materials”, (2012), Journal of Alloys and Compounds ,514, p. 6–19.
4-ASTM D1002, “Standard Test Method for Apparent Shear Strength of Single-Lap-Joint Adhesively Bonded Metal Specimens by Tension Loading (Metal-to-Metal)”, (2001).

5-Khazaka R, Mendizabal I, Henry D, "Review on joint shear strength of mano-silver paste and its long-term high temperature reliability", (2014), Journal of Electronic Materials, Vol. 43, No. 7, DOI: 10.1007/s11664-014-3202-6.

6- Siow, "Are Sintered Silver Joints Ready for Use as Interconnect Material in Microelectronic Packaging", (2014), Journal of Electronic Materials, DOI: 10.1007/s11664-013-2967-3.

7- Abbas, A., Abdul Kareem F, H., Sabah M. "Experimental study of temperature effect an curing time on the shear strength of adhesive joints by polyvinyl pyrrolidone PVP K30", (2017), Kufa Journal of Engineering, Vol. 8, No. 2, pp. 53–66.

8- Sivaiah, K., Rudramadevi, B. H., Buddhudu, S., Kumar, G. B. and Varadarajulu, A., “Structural, thermal and optical properties of Cu2+ And Co2+: PVP polymer films”, (2010), Indian Journal of Pure & Applied Physics, 48, p. 658–662.

9- Ebnesajjad, S. and Landrock, A. H. Adhesives Technology Handbook, (2007) , pp. 3520–3530, 2nd Edition, United States: William Andrew Publishing,

10-Aubrey, D Polymer Science Handbook, (1972 ), pp.936, Second Edition , North-Holland Pub.