

# Ultra-low Temperature Curable Conductive Silver Adhesive with different Resin Matrix

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**Abstract.** The ultra-low temperature curable conductive silver adhesive with curing temperature less than 100 °C needed urgently for the surface conductive treatment of piezoelectric composite material due to the low thermal resistance of composite material and low adhesion strength of adhesive. An ultra-low temperature curable conductive adhesive with high adhesion strength was obtained for the applications of piezoelectric composite material. The microstructure, conductive properties and adhesive properties with different resin matrix were investigated. The conductive adhesive with AG-80 as the resin matrix has the shorter curing time (20min), lower curing temperature (90°C) and higher adhesion strength (7.6MPa). The resistivity of AG-80 sample has the lower value ( $2.13 \times 10^{-4} \Omega \cdot \text{cm}$ ) than the 618 sample ( $4.44 \times 10^{-4} \Omega \cdot \text{cm}$ ).

## 1. Introduction

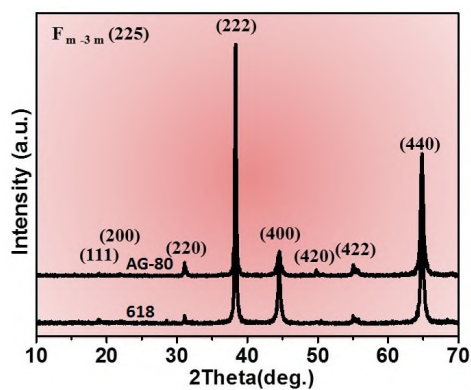
The surface conductive treatment is important to the dielectrics when they used in the devices. Piezoelectric composite materials are widely used in transducers for acoustics, medical and other fields [1-4]. During the preparation of the transducer, the adhesion strength of the electrode layers for the piezoelectric composite transducers is a difficult research point due to the polymer components in the composite material cannot bear high temperature, requiring low curing temperature and short curing time conductive adhesive which also shows strong conductive ability and adhesion strength. Conductive silver adhesive had been widely used in the field of IT and materials [5-12]. In recent decades, with the rapid development of semiconductor devices, conductive silver adhesive developed rapidly. M Bouguettaya [10] reported the Intrinsic Conducting Polymer (ICP) used to prepare conductive adhesives, providing excellent electrical and thermal properties for carbon black and polyaniline. X S Li [12] prepared silver nanowires as conductive adhesive for the conductive filler by alcohothermal method, and it has good electrical and mechanical properties. However, currently reported conductive silver adhesive showing the curing temperature of 150 ~ 300 °C [13-20]. It is not low enough for the piezoelectric composite material. In this paper, to meet the application of piezoelectric composite materials, the ultra-low temperature (90°C) fast curing (30min) with high adhesion strength conductive silver adhesive were studied. 618 and AG-80 were chosen to be the resin matrix.



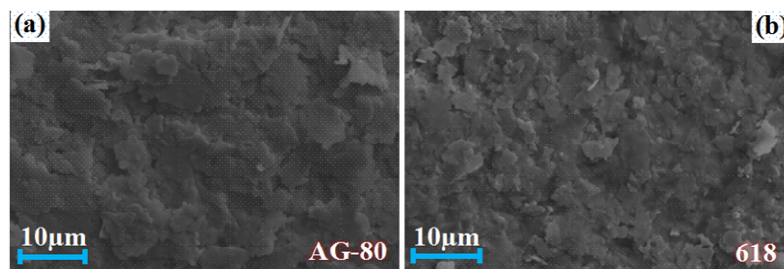
## 2. Experimental section

The resin matrix solution is firstly prepared by Epoxy Resin (AG-80), curing agent Ethylenediamine (EDA), Dibutyl Phthalate (DBP, C<sub>16</sub>H<sub>22</sub>O<sub>4</sub>, Analytical Pure), Carboxylated- terminated liquid acrylonitrile rubber (CTBN, Analytical Pure), Silane Coupling Agent (SCA-403, Analytical Pure) and accelerator Diethylene Glycol Butyl Ether (DGBE, Analytical Pure) in proportion of 100:160:10:10:10:10. Then add acetone (Analytical Pure), prepared resin matrix solution, and nano-silver (100 nm flaky pellets) in proportion of 10:30:60 to obtain the final ultra-low temperature curable conductive silver adhesive. The powder X-ray diffraction analysis was carried out at the 4B9A beamline at the Beijing synchrotron radiation facility (BSRF) with an X-ray wave length of 0.15406nm. The surface morphological was observed using a Scanning Electron Microscope (SEM: EVO-18, ZEISS, Germany). The component analysis was carried out by Energy Dispersive Spectrometer (EDS: EVO-18, ZEISS, Germany). The thermal analysis was carried out by thermogravimetric analysis (SDT Q600, TA, USA) and differential scanning calorimetry (DSC: SDT Q600, TA, USA).

## 3. Results and discussions

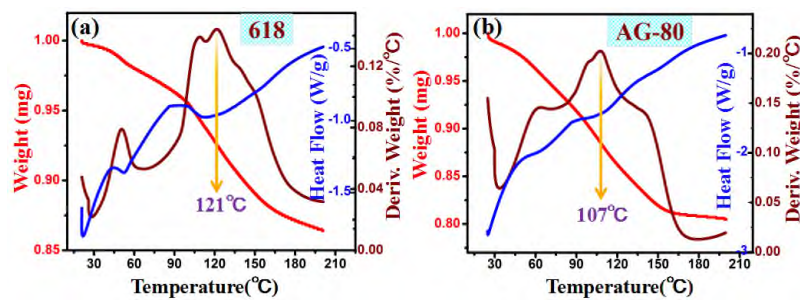


**Figure 1.** The synchrotron radiation X-Ray Powder Diffraction patterns of cured ultra-low curable conductive silver adhesive with resin matrix of 618 and AG-80, respectively.



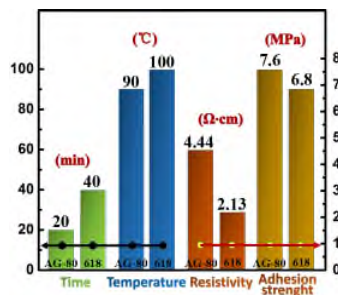
**Figure 2.** The SEM image of cured ultra-low temperature curable conductive adhesive with resin matrix of AG-80 and 618, respectively.

Figure 1 shows the synchrotron radiation X-Ray Powder Diffraction patterns of cured ultra-low curable conductive silver adhesive with resin matrix of 618 and AG-80, respectively. Both two samples show the single Ag phase with space group of Fm -3 m (225) (PDF No. 42-0479). Figure 2 shows the SEM image of cured ultra-low temperature curable conductive adhesive with resin matrix of AG-80 and 618, respectively. It can be seen that after the curing, the conductive surface shows good sintered representation. The whole surface of sample formed a good conductive path, in other words, the silver flake grain arranged neatly, and connected tightly. The sample with 618 as resin matrix has relative neater arrangement. As the characteristics of the large specific surface area, high surface free energy, high activity, various internal crystal defects in nano-silver particles, the condensation reaction of organic, and the thermal movement of silver particles made adjacent nano-silver particles move and gather during heat-curing. The conductive adhesive formed larger structure by crystal boundary migration and edge blending of crystal particles.



**Figure 3.** The thermal analysis results of the conductive silver adhesive with resin matrix of 618 and AG-80, respectively.

Figure 3 shows the thermal analysis (TGA, DTG, DSC) results of the conductive silver adhesive with resin matrix of 618 and AG-80, respectively. Cured products of AG-80 shows weight loss significantly in the 70 °C to 150 °C, and gradually slowed down after 150 °C until the weight loss rate reached 17%, and then the curing reaction tends to complete. Cured products of 618 shows weight loss significantly in the range of 100 °C to 175 °C. Until the weight loss rate reached 13%, the curing tends to complete. This is due to the epoxy groups of AG-80 is more than 618, and the activity is much higher than 618. The reaction releases a lot of heat, which can make sure fast curing at low-temperature. The weight loss rate of cured product of AG-80 and 618 reached the maximum at 107 °C and 121°C, respectively. The initial increase of heat release indicates the beginning of crosslinking of the resin. With the increase of the heating rate, the exothermic amplitude is also increasing. The initial decomposition temperature of cured product of AG-80 and 618 are 50 °C and 70°C. In addition, it can be seen that thermal stability of 618 epoxy resin is poor.



**Figure 4.** The process parameters, conductive property and adhesion property of the conductive adhesive with resin matrix of 618 and AG-80, respectively.

Figure 4 shows the process parameters, conductive property, and adhesion strength of cured ultra-low curable conductive silver adhesive with resin matrix of 618 and AG-80, respectively. It can be found that the conductive adhesive with AG-80 as the resin matrix has the shorter curing time, lower curing temperature and higher adhesion strength. As described above, it is due to the difference of epoxy group between 618 and AG-80. The resistivity testing following “ASTM D257-2007” standard. It can be seen from figure 5 that the resistivity of 618 sample has the lower value  $2.13 \times 10^{-4} \Omega \text{ cm}$  than the AG-80 sample ( $4.44 \times 10^{-4} \Omega \text{ cm}$ ). It may be because the better micromorphology of 618.

#### 4. Conclusions

In summary, an ultra-low temperature curable conductive adhesive with different resin matrix (AG-80 and 618) were investigated for surface conductive treatment of piezoelectric composite material. Both two samples show the single Ag phase with space group of  $F_{m-3m}(225)$  (PDF No. 42-0479), and the surface with conductive treatment formed a good conductive path in all direction. The conductive adhesive with AG-80 as the resin matrix has the shorter curing time, lower curing temperature and higher adhesion strength. The resistivity of 618 sample has the lower value  $2.13 \times 10^{-4} \Omega \text{ cm}$  than the AG-80 sample ( $4.44 \times 10^{-4} \Omega \text{ cm}$ ).

#### References

- [1] Jang C, Sharifi M, Palmese, G R, Abrams C F 2014 *Polymer*, **55**(16) 3859-68.

- [2] Lan T, Kaviratna P D, Pinnavaia T J 1995 *Chem. Mater.* **7**(11) 2144-50.
- [3] Sang J L, Sohn H, Hong J W 2010 *J. Nondestruct. Evaluat.* **29**(2) 75-91.
- [4] Sato S, Yamada K, Inagaki N 2006 *Med. Biol. Eng. Comp.*, **44**(5) 353-62.
- [5] Pettersen S R, Redford K, Njagi J, Kristiansen H, Helland S, Kalland E, Goia D, Zhang Z, He J 2017 *J. Electr. Mater.* 1-11.
- [6] Rane S B, Seth T, Phatak G J, Amalnerkar D P, Das B K 2003 *Mater. Lett.* **57**(20) 3096-100.
- [7] Zhao S Y, Li X, Mei Y H, Lu G Q 2016 *J. Electr. Mater.* **45**(11) 1-11.
- [8] Peng P, Hu A, Gerlich A P, Zou G, Liu L, Zhou Y N 2015 *Acs. Appl. Mater. Interf.* **7**(23) 12597.
- [9] Kwon J, Cho, H, Eom H, Lee H, Suh Y D, Moon H, Shin J, Hong S, Ko S H 2016 *Acs. Appl. Mater. Interf.* **8**(18) 11575.
- [10] Bouguettaya M, Védie N, Chevrot C 1999 *Synth. Met.* **102**(1-3) 1428-31.
- [11] Marcq F, Demont P, Monfraix P, Peigney A, Laurent C, Falat T, Courtade F, Jamin T 2011 *Microelectron. Reliabil.* **7** 1230-4.
- [12] Li X S, Xiang X Z, Wang L, Bai X J 2016 *Rare Met.* 1-5.
- [13] Morris J E 2017 *Polymers*, **3**(1) 191-7.
- [14] Nakayama K, Nagai A, Iida N 2007 *Mater. Trans.* **48**(3) 594-9.
- [15] He S, Zhang X, Yang B, Xu X, Chen H, Zhou C 2017 *Chinese Phys. B*, **26**(7) 426-32.
- [16] Park S D, Yoo M J, Kang N K, Park J C, Lim J K, Kim D K 2004 *Macromol. Res.* **12**(4) 391-8.
- [17] Lu C A, Lin P, Lin H C, Wang S F 2006 *Japanese J. Appl. Phys.* **45**(9A) 6987-92.
- [18] Bai J G, Zhang Z Z, Calata J N, Lu G Q 2006 *IEEE Trans. Comp. Pack. Tech.* **29**(3) 589-93.
- [19] Gural P S, Jenniskens P 2007 *Japanese J. Appl. Phys.* **46**(1) 251-5.
- [20] Heckmann T, Souvignat T, Naccache D 2017 *Dig. Investigat.* **21** 53-64.

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