

# PREPARATION AND PROPERTY OF Ni/Ag-Cu POWDER COMPOSITE CONDUCTIVE PASTE

## PRIPRAVA IN LASTNOSTI Ni/Ag-Cu KOMPOZITNE PREVODNE PASTE V PRAHU

Fuqi Lian, Xiaolei Su\*, Yi Liu

School of Materials Science & Engineering, Xi'an Polytechnic University, Xi'an 710048, China

Prejem rokopisa – received: 2022-01-07; sprejem za objavo – accepted for publication: 2022-03-21

doi:10.17222/mit.2022.360

A composite conductive paste was prepared using Ni powder and Ag-Cu powder as the conductive fillers, and modified polyurethane resin (PUR) as the matrix. The conductive coatings were cured after being applied onto polyethylene terephthalate (PET) films. Results show that when the mass ratio of the Ag-Cu powder, Ni powder and resin matrix was 13 : 2 : 5, the resistivity of the prepared conductive paste reached a minimum value of  $7.92 \times 10^{-4} \Omega\cdot\text{cm}$  and the binding force between the conductive coating and PET film was 20.1 N. In addition, the prepared coatings also exhibited good corrosion resistance in a neutral corrosive environment. Even after being bent 50 times, they showed no significant change, still exhibiting good corrosion resistance and bending performance.

Keywords: conductive paste, polyethylene terephthalate, thin film, modified polyurethane resin, corrosion resistance

Avtorji v pričujočem članku opisujejo pripravo prevodne kompozitne paste z uporabo prahov Ni in Ag-Cu kot električno prevodnega polnila in matrice iz modificirane poliuretanske smole (PUR; angl.: polyurethane resin). Prevodne prevleke so toplotno obdelali in jih nato nanесли na polimerne filme oziroma tanke trakove iz polietilen tereftalata (PET; angl.: Polyethylene terephthalate). Rezultati raziskave so pokazali, da so pri masnem razmerju prahov Ag-Cu, Ni in matrične smole 13 : 2 : 5 dobili prevodno pasto z najmanjšo prevodnostjo  $7.92 \times 10^{-4} \Omega\cdot\text{cm}$ . Pri tem je bila vezavna sila med prevodno pasto in PET filmom 20,1 N. Poleg tega so pripravljene prevleke imele dobro odpornost proti koroziji v nevtralnem korozijskem okolju. Po 50-kratnem pregibanju polimernih filmov s prevodno prevleko še ni prišlo do vidne spremembe odpornosti proti koroziji ali pokanja prevleke med pregibanjem.

Ključne besede: prevodna pasta, polietilen tereftalat, tanek film, modificirana polivretanska smola, korozijska odpornost

## 1 INTRODUCTION

The most important raw material in high-temperature sintered ceramics is the conductive paste, which is a new functional material that integrates materials, chemical industry, fluids, and electronic component technology.<sup>1</sup> Conductive paste is primarily made up of a conductive filler and an organic carrier, and the conductive properties of the paste are mostly determined by the conductive filler.<sup>2</sup> Filler properties such as resistivity, morphology and particle-size distribution have a substantial impact on conductivity.<sup>3</sup> Conductive paste has become an indispensable material for electronic components, and it is popular due to its environmental protection, non-polluting, simple production process, and a low curing temperature. Traditional solders pollute the environment, pose a health risk, and cannot meet modern needs.<sup>4</sup> Although precious metals like Au, Ag, and Pd as conductive fillers allow a great precision and reliability, their high prices limit their use.<sup>5</sup> Due to their low costs and ease of production, Cu, Ni, and Al as well as other base metals, have become the preferred conductive fillers.<sup>6</sup>

By combining the benefits of several conductive particles, the overall performance of conductive paste can be improved. The Ni-base metal was utilized as the filler in a conductive paste, which showed good chemical stability, solder resistance and no particle migration.<sup>7</sup> The conductive paste that contains precious metal Ag exhibits strong adhesion, electrical conductivity, chemical stability, a high production cost, and Ag<sup>+</sup> migration.<sup>8</sup> Cu paste has the advantages of being environmentally friendly, low in cost, and easy to manufacture, but it has the disadvantage of being easily oxidized, and the conductivity decreases after oxidation.<sup>9,10</sup> As a new conductivity functional material, Ag-Cu powder can address issues such as high cost, Cu-powder oxidation, and Ag<sup>+</sup> migration.<sup>11</sup> A precious and base-metal particle filler in composite conductive paste not only has good conducting qualities but it also lowers the preparation cost.

S. Y. Zhang et al. investigated and optimized the resistivity and shear strength of nano-Ag-Cu powder particles mixed with conductive adhesive. The conductive filler content was optimized to 23.5 % volume, the conductive-adhesive resistivity was  $5.159 \times 10^{-6} \Omega\cdot\text{cm}$ , and the shear strength was 6.121 MPa.<sup>12</sup> S. N. Liu et al. produced Ni-powder particles with a diameter of 11–48 nm in an aqueous medium. Synthetic nano-Ni powder and mi-

\*Corresponding author's e-mail:  
su\_x\_lei@163.com (Xiaolei Su)

cro-Ni powder were used as conductive particles, and Ni powder conductive paste with a resistance of  $1.22 \times 10^{-4} \Omega\text{-cm}$  was prepared via screen printing and high-temperature sintering.<sup>7</sup> S. J. Peighambardoust et al. prepared conductive paste, using epoxy resin as the matrix, Ag-Cu and Ag decorated graphene as the conductive filler, thus achieving good thermal stability. The electrical conductivity, shear strength and stability were investigated. The volume resistivity reached  $1.60 \times 10^{-2} \Omega\text{-cm}$ .<sup>13</sup> The problem of aging, poor mechanical properties, large volume resistivity, and low bond strength significantly limited its application although it had many advantages.<sup>14</sup>

PET films exhibit outstanding mechanical qualities, thermal stability and good adhesion properties. A conductive paste coated on the PET films has good characteristics. Ag-Cu powder was prepared using a thermal curing process, and the Ag-Cu powder with 20 w/% Ag content and Ni powder as fillers, modified polyurethane resin (PUR) as the matrix, propylene glycol methyl ether acetate (PMA) and dimethyl ester (DBE) as the solvents. The above-prepared conductive paste was then applied to PET films. The conductive-paste conductivity, corrosion resistance, bending performance and conductivity mechanism were studied.

## 2 EXPERIMENTAL PART

### 2.1 Materials

Modified polyurethane resin (PUR), propylene glycol methyl ether acetate (PMA), dimethyl ester (DBE) and a curing agent (HF-3285) were purchased from Polycarbon Synthetic Materials Co., Ltd., Guangzhou, China. The coupling agent was KH550 produced by Yousuo Chemical Technology Co., Ltd., Jinan, Shandong, China.

The PET film was purchased from Guangdian Film Materials Co., Ltd., Hangzhou, Zhejiang, China. The Cu powder was purchased from Yousuo New Materials Co., Ltd., Beijing, China.  $\text{AgNO}_3$  (98 %) was purchased from Guanghua Technology Co., Ltd., Guangzhou, China. The emulsifier was NP-5, produced by Fuyu Fine Chemical Co., Ltd., Tianjin, China. NaOH, DTPA, NaCl and diluted  $\text{H}_2\text{SO}_4$  were provided by Hongyan Reagent Factory, Hedong, Tianjin, China. The Ni powder was purchased from Jingxing Welding Material Co., Ltd., Nangong, Hebei, China. All the above reagents were analytically pure.

### 2.2 Preparation of the conductive coating

Ball-milled 63  $\mu\text{m}$  dendritic Cu powder yielded a flake Cu powder. For pickling, 30 g of ball-milled Cu powder was placed in a beaker and 5 %  $\text{H}_2\text{SO}_4$  was added. Meanwhile, the liquid was agitated to ensure that all of the oxides on the surface were pickled. After pickling, the Cu powder was washed to neutralize it. The beaker was filled with a certain amount of NP-5 and DTPA as well as deionized water in a 1 : 10 solid-to-liquid ratio. To make a Cu-powder suspension, the treated Cu powder was placed into a beaker and mechanically agitated for 30 min. A 15 g/L  $\text{AgNO}_3$  solution (DTPA :  $\text{AgNO}_3 = 1 : 2$ ) was prepared. After stirring the suspension, the  $\text{AgNO}_3$  solution was slowly added and allowed to react for 30 min. The Ag-Cu powder solution was produced, filtered and dried.

The preparation technique for an ecologically safe, non-toxic, non-polluting conductive paste formulation (the ratio of the organic carrier in the paste is PUR : DBE : PMA = 10 : 7 : 3) is as follows. First, certain amounts of DBE, curing agent HF-3285 and coupling agent KH550 (the amounts of the curing agent and cou-

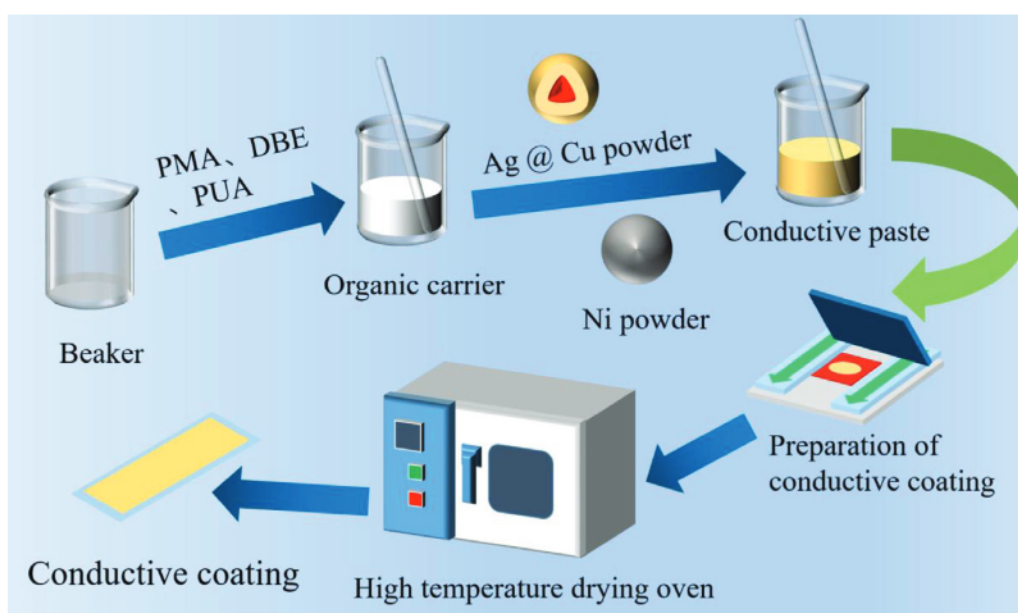


Figure 1: Preparation flow chart of the composite conductive paste



pling agent are 3 w/% and 1 w/%, respectively) are thoroughly mixed, then added to the PUR beaker and stirred to prepare the organic carrier; this is then mixed well with the Ag-Cu powder and Ni powder, and a small amount of PMA is added to adjust the paste viscosity. The paste is then scraped onto the PET sheet and dried for 30 min at 130 °C to create a Ni/Ag-Cu powder conductive coating. **Figure 1** depicts the conductive-paste preparation process.

**Table 1:** Composition of the composite conductive paste

Sn. No.	Ni powder (w/%)	Ag-Cu powder (w/%)	matrix resin (w/%)
1	0	75	25
2	10	65	25
3	20	55	25
4	30	45	25
5	40	35	25
6	50	25	25

### 2.3 Performance testing

The surface resistance of the coating was tested with a DC low-resistance tester, and the bonding strength between the conductive coating and the substrate was tested with an automatic scratch tester. The morphology of the conductive coating was observed with a cold field emission scanning electron microscope. The particle size of the conductive filler was measured with a laser particle-size analyser. The corrosion of the conductive coating in a neutral corrosion environment was analysed using an electrochemical workstation.

#### 2.3.1 Electrical conductivity

The coating was cut into cuboids with dimensions of (3.4 × 1.1 × 0.005) cm (length × width × thickness) for resistance value measurement with the DC low-resistance tester, and then the resistivity was calculated with Equation (1):

$$\rho = RS/L \quad (1)$$

where  $\rho$  is the surface resistivity ( $\Omega\cdot\text{cm}$ ),  $R$  is the resistance value measured with the DC low-resistance tester ( $\Omega$ ),  $L$  is the length (cm) and  $S$  is the cross-sectional area ( $\text{cm}^2$ ).

#### 2.3.2 Electrochemical performance

The coating was cut into cuboids with dimensions of (10 × 10 × 0.005) cm and the working area was 1.0  $\text{cm}^2$ . The electrochemical workstation was used to test the corrosion resistance of the coating in a 3.5 % NaCl solution. The three-electrode system was adopted. The conductive coating was the working electrode, the Ag/AgCl (saturated KCl) electrode was the reference electrode, and the carbon rod was the auxiliary electrode. The frequency range of the EIS test was over 0.01–100 kHz and the AC voltage was 5 mV. The polarization-curve scanning rate was 10  $\text{mV}\cdot\text{s}^{-1}$  and the scanning positioning

was –0.4 V to 0.3 V. The above tests were carried out at room temperature.

#### 2.3.3 Corrosion performance

The coating was immersed in a neutral environment for 168 h, then the surface was dried with filter paper. The microstructure of the samples was observed with a metallographic microscope and the resistance was measured. Then, the resistivity after corrosion was calculated with Equation (1).

#### 2.3.4 Bending performance

The conductive paste was cut into a sample with dimensions of (3.4 × 1.1 × 0.005) cm, then the resistance of the conductive coating was tested by attaching it to a cylindrical object with a diameter of 0.8 cm in the positive direction (conductive side outward). The resistance test was performed every 10 bends for a total of 50 bends on the coating. The square resistance of the coating for every 10 bends was calculated with Equations (1) and (2):

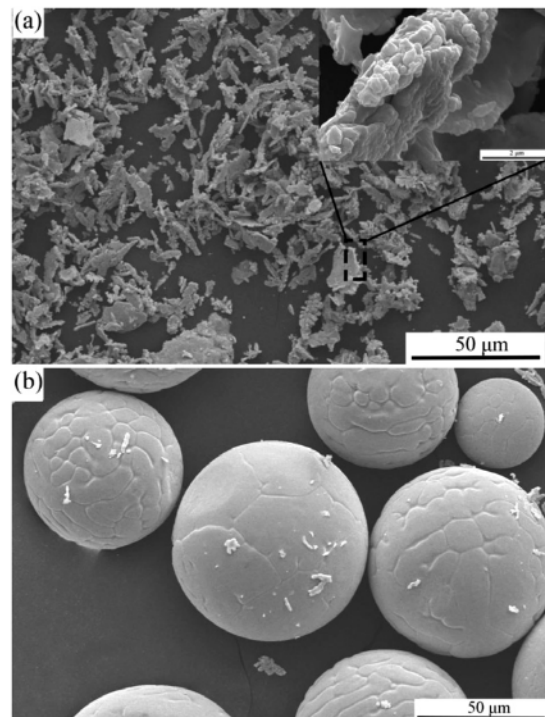
$$R_s = \rho/t \quad (2)$$

where  $R_s$  is the volume resistance ( $\Omega\cdot\text{sq}^{-1}$ ),  $\rho$  is the surface resistivity ( $\Omega\cdot\text{cm}$ ),  $t$  is the coating thickness.

## 3 RESULTS AND DISCUSSION

### 3.1 Characterization of the Ni and Ag-Cu powder

PUR has good mechanical qualities, wear resistance, high strength, low adhesion and poor heat resistance.<sup>15</sup> The wear resistance, mechanical characteristics, high



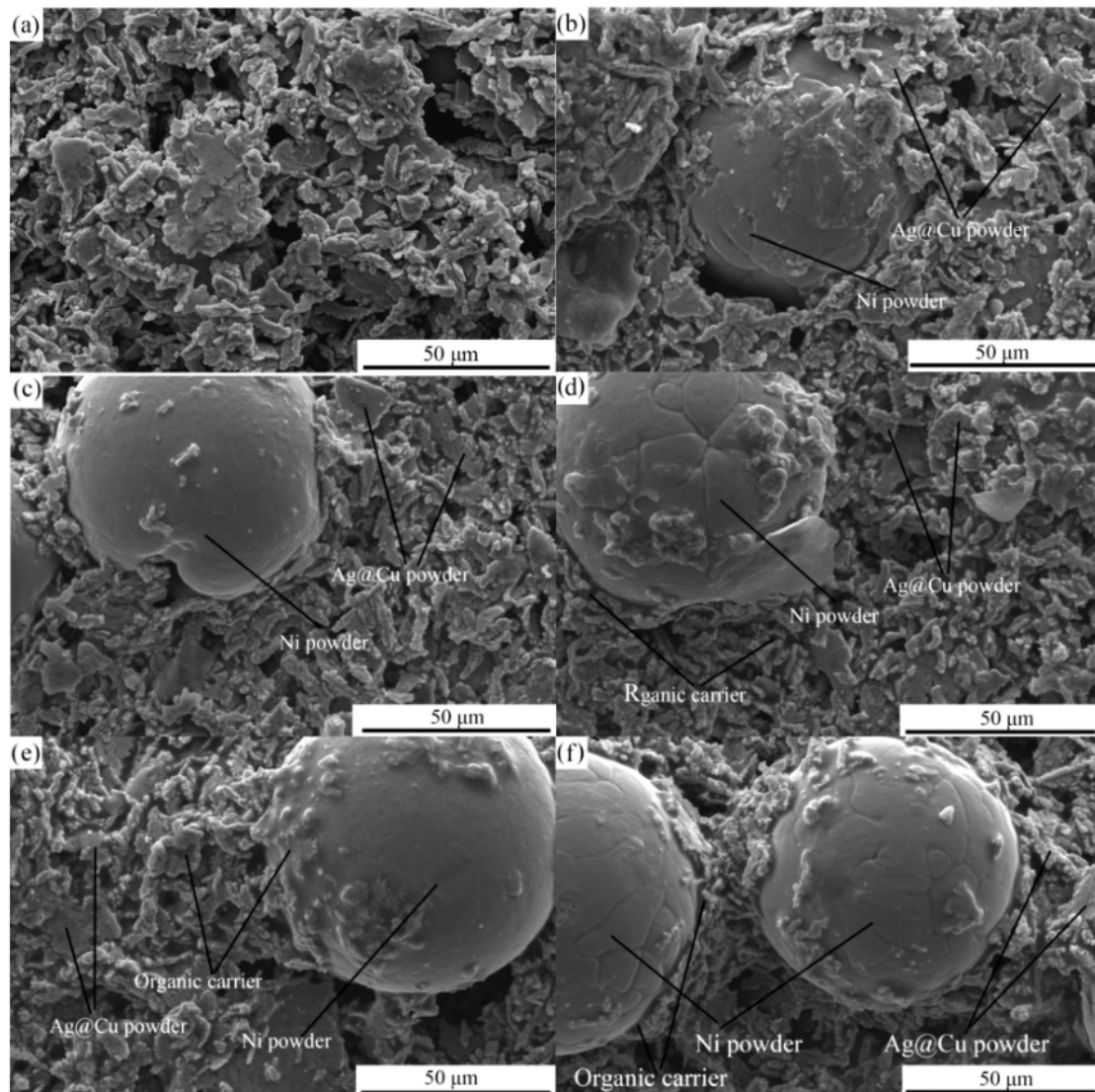
**Figure 2:** Characterization of different conductive fillers in conductive paste: a) SEM image of Ag-Cu powder, b) SEM image of Ni powder

strength and high adhesion are all the advantages of the modified resin. The composite conductive paste was filled with Ag-Cu powder with 20 w/% Ag and Ni powder in the experiment. The microstructures of the Ag-Cu powder and Ni powder, shown in **Figure 2**, were flakey and spherical, respectively. The Ni powder exhibits good dispersibility and no agglomeration, while the in-house-made Ag-Cu powder has good dispersibility and no agglomeration. Using a laser particle-size analyser, the average particle sizes of Ag-Cu powder and Ni powder were 10.18  $\mu\text{m}$  and 50  $\mu\text{m}$ , respectively.

### 3.2 SEM characterization

A SEM was used to examine the surface micro-morphology of the composite conductive coatings with different Ni-powder concentrations. **Figure 3** depicts the coating-surface micro-morphology.

It can be observed from **Figure 3** that with an increase in the Ni-powder content, the number of Ni powder spheres increases continuously. When the content of Ni powder is 10 w/% and 20 w/%, the spherical Ni powder is sparsely distributed in the Ag-Cu conductive paste. Curing the composite paste can form a good conductive network or conductive path inside the coating, which exhibits a good conductive ability. When the content of Ni powder is (30, 40 and 50) w/%, the spherical Ni powder is densely distributed in the Ag-Cu conductive paste, and the Ni powder is a conductive filler with a small range of agglomeration in the coating. After solidification, the composite paste does not form a complete conductive network or conductive path in the coating, and the conductive ability decreases continuously. In particular, when the Ni powder content is 40 w/% and 50 w/%, the conductive network in the coating after the solidification of the composite paste is incomplete, resulting in a sharp decrease in the conductivity.



**Figure 3:** SEM of conductive paste with different Ni powder contents: a) 0 w/%, b) 10 w/%, c) 20 w/%, d) 30 w/%, e) 40 w/%, f) 50 w/%

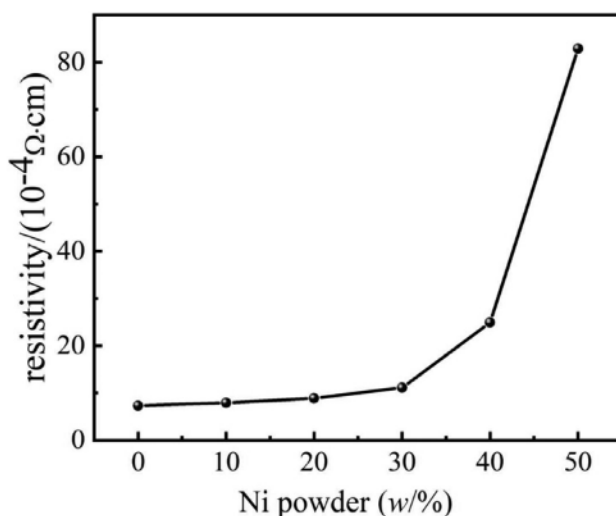


### 3.3 Effect of the conductive-filler content on resistivity

The conductivity of the paste mainly comes from the conductive fillers. Compared with the point contact of the spherical fillers, an addition of a sheet filler can increase the contact area and contact probability, thereby improving the conductivity of the paste. The composite paste is prepared with several types of conductive particles and they can combine their respective advantages so that the overall performance of the paste is improved. **Figure 4** shows the resistivity of the composite conductive paste with different Ni-powder percentages.

According to the seepage theory for conductive paste,<sup>12,14</sup> the interface contact of the conductive filler is the key factor for the paste resistivity. The conductive path is formed by the contact of the fillers, making the paste electrically conductive. Before the paste is cured, the conductive particles are in an isolated state and not in contact with each other. During the curing process, the volatilization of organic solvents and the cross-linking volume shrinkage of the resin shorten the distance between the conductive particles so that an interface contact occurs between the fillers, thus showing a certain degree of conductivity. With the increase in the amount of conductive particles, the resistance change of the paste is discontinuous. When the filler content reaches a certain critical value, the interface contact between the conductive particles can form a complete conductive network, the conductive paste mutates and the semiconductor also mutates into a conductor. The critical value of the sudden change in the resistivity of the paste is called the percolation threshold.

**Figure 4** shows that when the Ni powder content is 10 w/% and the Ag-Cu powder content is 65 w/%, the resistivity of the paste is  $7.92 \times 10^{-4} \Omega\cdot\text{cm}$ ; when the Ni powder content is 40 w/%, the resistivity of the paste sharply increases, to  $24.9 \times 10^{-4} \Omega\cdot\text{cm}$ . When the Ni powder content is 50 w/%, the resistivity of the coating is  $82.9 \times 10^{-4} \Omega\cdot\text{cm}$  and the resistivity of the paste sharply

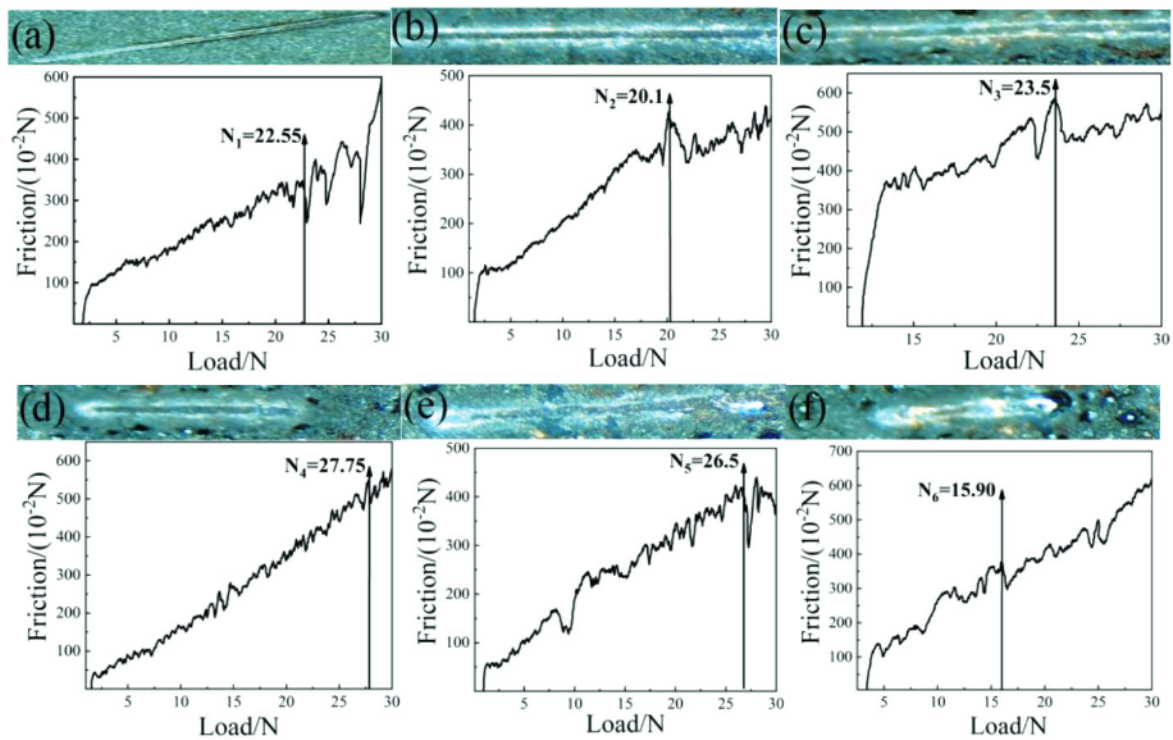


**Figure 4:** Resistivity of conductive paste with different Ni-powder contents

increases. The figure shows that with the increase in the Ni-powder content, the resistivity of the paste increases gradually. When the conductive particles are the Ag-Cu powder, the resistivity of the paste is  $7.3 \times 10^{-4} \Omega\cdot\text{cm}$ , and the resistivity of the paste contains 10 w/% and 20 w/% of the Ni powder is  $7.92 \times 10^{-4} \Omega\cdot\text{cm}$  and  $8.90 \times 10^{-4} \Omega\cdot\text{cm}$ . Compared with the single conductive filler Ag-Cu powder conductive paste, the resistivity of the composite filler conductive paste exhibits little change and can thus reduce the production cost of the paste while meeting the practical application requirements.

### 3.4 Influence of the conductive-filler content on adhesion

**Figure 5** is a graph of the bonding strength and scratches of the Ni-powder composite conductive coating with different contents. The scratch morphology left by the scratch tester on the surface of the conductive coating was identified with a metallographic microscope. When the scratch loading reaches the limit of the coating, the conductive coating peels off or falls off from the PET-film substrate, and the appearance of the scratch changes as the load becomes the critical load. When the loading continues to increase and it increases to a certain extent, it drops again. The bonding force between the coating and the substrate is low. In the process of scratching, the structure of the coating is squeezed and accumulated and the coating falls off. The binding force between the conductive coating and the PET-film substrate increases first and then decreases. When the content of Ni powder is 30 w/%, the binding force between the conductive coating and the PET-film substrate is N4, and the binding force reaches the highest value. The coating surface begins to come off and the tissue slips as the dynamic stress approaches the critical load and the load continues to rise. The coating function fails as the coated surface continues to fall off. The depth of the scratch gradually rises as the applied load is increased. When the content of Ni powder is 10 w/% and 20 w/% and the loading is applied again, the friction force increases sharply. When the loading is continued, the friction force fluctuates continuously and there is a large fluctuation. When the content of Ni powder is 40 w/% and 50 w/% and the loading is applied, the friction force increases. The structure of the coated surface is severely compressed and piled up when the friction force varies greatly; the scratches on the surface are neat and clear of burrs, and no substrate is revealed at the ends of the scratches. An excessive Ni-powder content causes poor bonding force between the coating and the PET-film substrate, low resin viscosity, a gap between the filler and interface, poor interface compatibility and low bonding strength of the coating with the substrate.



**Figure 5:** Characterization of adhesion performance of conductive coatings with different Ni-powder contents: a) 0 w%, b) 10 w%, c) 20 w%, d) 30 w%, e) 40 w%, f) 50 w%

### 3.5 Study on the corrosion resistance of conductive paste

The composite conductive coatings with different contents of Ni-powder were immersed in a 3.5 % NaCl solution. After being placed in it at room temperature for 168 h, the corroded conductive coatings were taken out, the water on the surface was dried with filter paper for observation and the coating surface fell off and wrinkled. The micromorphology of the conductive coating after corrosion is shown in **Figure 6**. The resistivity of the composite conductive coating before and after the immersion for 168 h is shown in **Figure 6h**.

The coating has many small defects due to its structure. During curing, the resin solvent volatilizes and shrinks, resulting in many microdefects in the coating, such as uneven distribution of the filler density, micropores and cracks. Macroscopic defects such as scratches on the coating surfaces are inevitable. These macroscopic or microscopic defects provide channels for the infiltration of corrosive media. In the process of immersion corrosion, the paste shrinks, the solvent volatilizes when it is solidified, and many pores or gaps appear inside the coating. The corrosive medium penetrates the coating through these parts so that the coating is corroded and various corrosion phenomena appear on the coating surface. It can be seen from **Figure 6** that with the increase in the Ni-powder content, surface defects such as holes and pits appear in the coating in different ranges after the immersion corrosion for 168 h. From

**Figure 6h**, it can be found that the resistivity of the composite filler coating with 10 w% and 20 w%.

Ni powder changed little after the corrosion, while the resistivity of the composite filler coating with (30, 40, and 50) w% Ni powder changed greatly after the corrosion. With the increase in the Ni-powder content, the resistivity change of the composite filler coating after the immersion for 168 h showed an increasing trend. This is because with the increase in the Ni-powder content, the corrosion resistance of the composite filler conductive coating decreases gradually, and the conductivity in a neutral corrosion environment also decreases gradually. After corrosion, the resistivity of the composite conductive paste coating with a Ni-powder concentration of 10 w% is not significantly different from that of a single conductive filler Ag-Cu conductive paste. When the content of Ni powder is 10 w% and 20 w%, the composite coating has good corrosion resistance after curing.

### 3.6 Study of the electrochemical corrosion resistance

The electrochemical-impedance-spectroscopy (EIS) curves and potentiodynamic-polarization curves of the Ni-powder composite conductive coatings with different Ni-powder concentrations in the 3.5 % NaCl solution are shown in **Figure 7**.

**Figure 7a** shows the polarization curves for the composite conductive coatings in the electrolytic solution. It can be seen that the self-corrosion potential of the coating with 30 w% Ni-powder content is the smallest, and



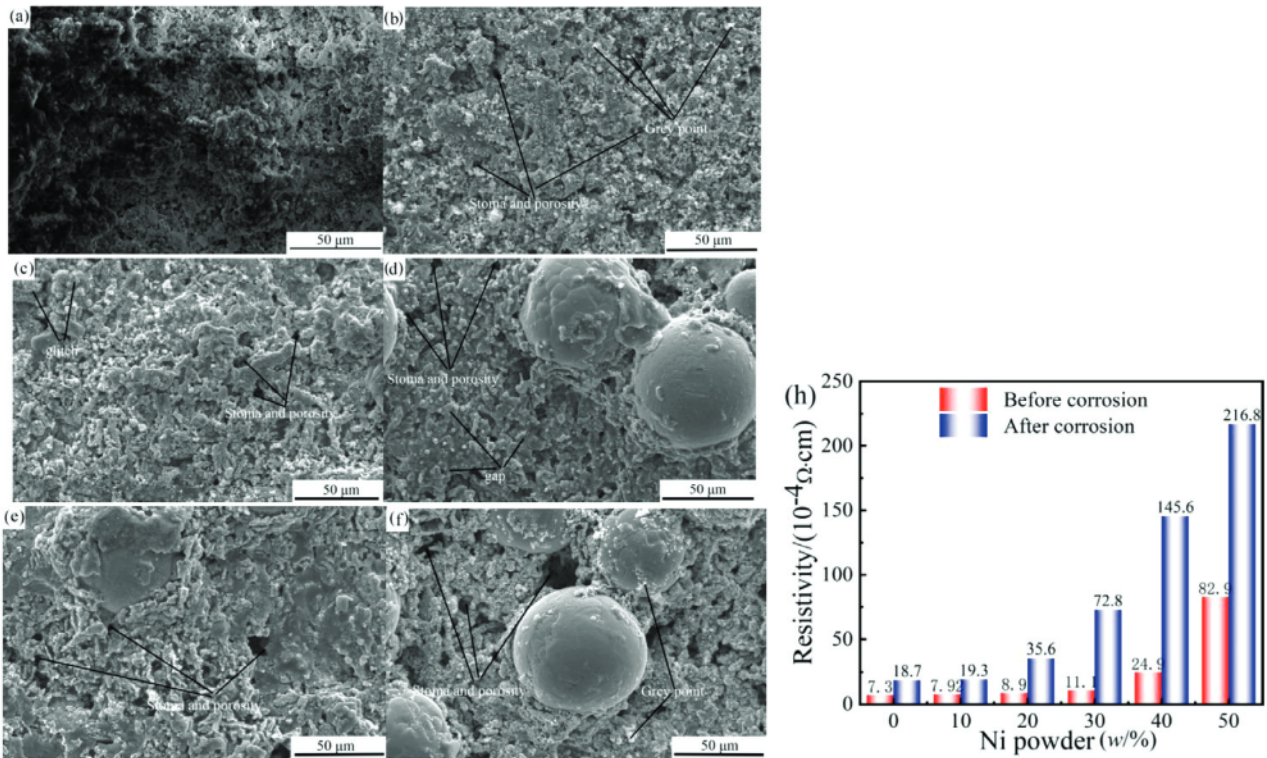


Figure 6: SEM and resistivity of the conductive coating after corrosion in neutral environments: a) Ni powder 0 w%, b) Ni powder 10 w%, c) Ni powder 20 w%, d) Ni powder 30 w%, e) Ni powder 40 w%, f) Ni powder 50 w%, h) resistivity of conductive coating after corrosion w/w

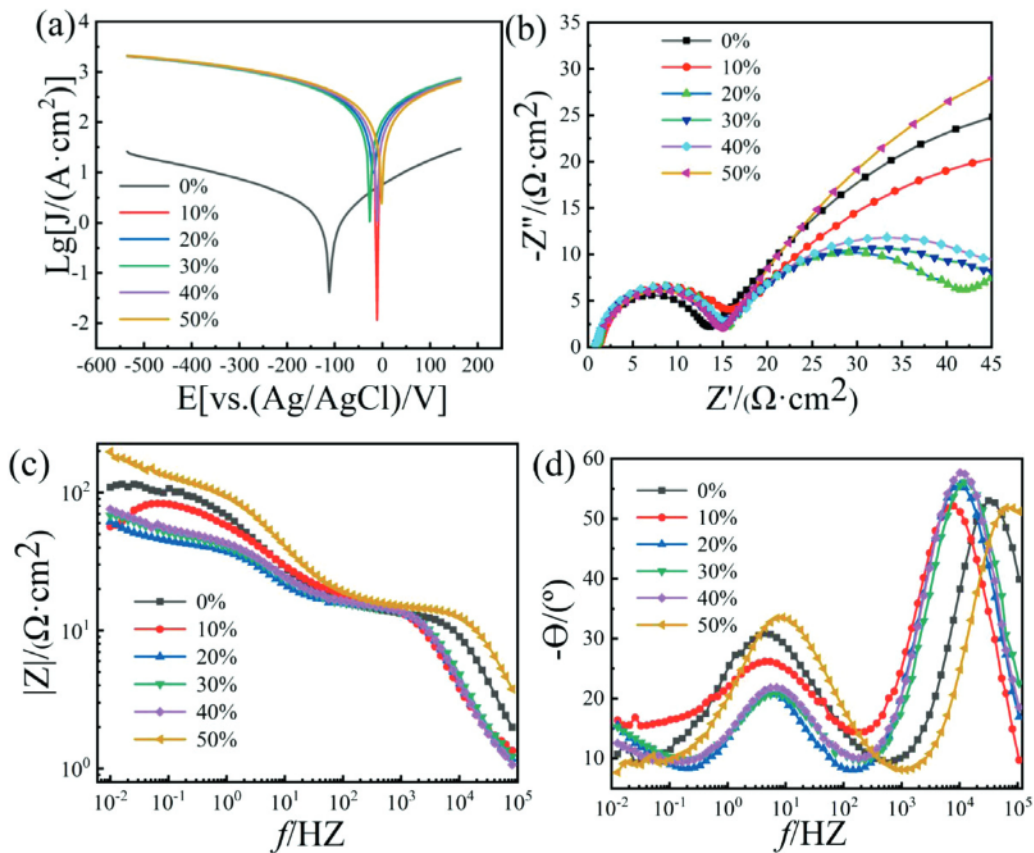


Figure 7: Polarization curves and impedance curves of composite conductive coating with different Ni-powder contents: a) polarization curve, b) Nyquist diagram, c) Bode amplitude/frequency, d) Bode phase/frequency

that of the coating with 50 w/% Ni-powder content is the largest. The self-corrosion potential values for the coatings with 10 % and 40 % Ni-powder content overlap, and the self-corrosion potential value for the coating with 20 w/% Ni-powder content is between those for 10 w/% and 30 w/%. The corrosion current density of the coating with 10 w/% Ni-powder content is the smallest, and the corrosion current density of the coating with 20 % Ni-powder content is the largest. The corrosion-current-density values for the coatings with Ni powder content are similar. The corrosion current density and self-corrosion potential values for the single conductive filler Ag-Cu conductive coating are lower, and the self-corrosion potential value is lower than that for the composite conductive coating with 10 w/% Ni-powder content. The smaller the corrosion current density, the slower is the corrosion rate of the coating and the best is the corrosion resistance of the coating; the higher the self-corrosion potential, the greater is the inertness of the coating surface and the less likely the coating is to react with the corrosive medium.<sup>16–18</sup> Therefore, the coating with 10 w/% Ni powder has the best corrosion resistance.

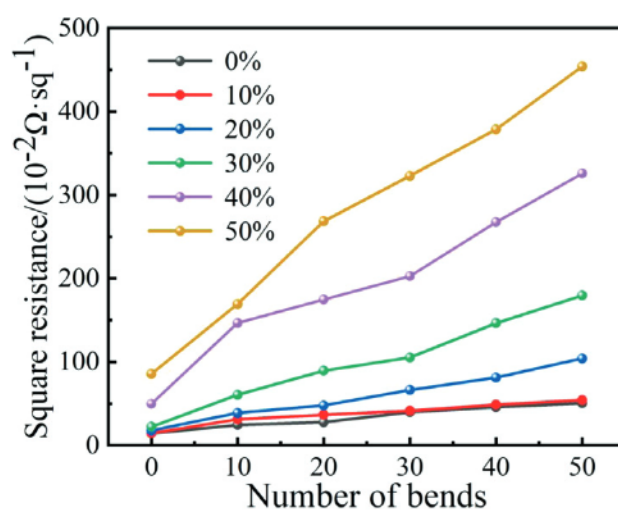
The impedance spectra of the composite conductive coatings in the electrolytic solution are depicted in **Figures 7b to 7d**. The coating exhibits the capacitive-arc overall frequency, as shown in **Figure 7b**. The smaller radius of the curvature of the capacitive arc, the smaller is the impedance. The impedance of the coatings with 50 w/% and 10 w/% Ni-powder contents is the largest, and the impedance of the coating with 20 w/% Ni-powder content is the smallest. **Figure 7c** shows the relationship between the impedance modulus  $|Z|$  and frequency  $f$  of the composite conductive coating. With the increase in the Ni-powder content, the impedance modulus  $|Z|$  of the coating decreases, but with the increase of frequency  $f$ , the impedance modulus  $|Z|$  decreases gradually, and decreases sharply in the high-frequency region. This shows that with the increase in the Ni-powder content, the barrier property of the coating decreases, which accelerates the diffusion of the corrosive medium and reduces the corrosion resistance of the coating.

The relationship between the frequency  $f$  and the phase angle  $\theta$  of the composite conductive coating is given in **Figure 7d**. It can be seen from the figure that as the frequency  $f$  increases, the phase angle  $\theta$  of the coating decreases first and then increases in the low-frequency region, and increases first and then decreases in the middle and high-frequency regions. The phase angle  $\theta$  eventually tends toward a stable value. In the low-frequency region, the phase angle migrates to the small angle direction, and in the middle and high-frequency region, the phase angle collectively migrates to the large angle direction. The barrier performance of the coating is improved, the diffusion of the corrosive medium is inhibited, and the corrosion resistance of the conductive coating is improved. When compared to a single conductive filler Ag-Cu conductive coating, the impedance modulus

$|Z|$  and phase angle  $\theta$  of the conductive coating with a Ni-powder concentration of 10 w/% are not significantly different regarding the frequency  $f$ , while the capacitive reactance arc curvature radius is slightly lower. Therefore, the conductive coating with 10 w/% Ni powder has the best corrosion resistance.

### 3.7 Effect of the conductive-filler content on bending performance

**Figure 8** is a graph showing the change in the square resistance of conductive coatings with different Ni-powder contents under different bending times. The flexible mechanical properties of the conductive coating are mainly reflected in the bending performance of the coating. The smaller the changes in the square resistance of the conductive coating with the increase in bending times, the better are the flexible mechanical properties of the coating. The figure shows that the square resistance increases with the increase in the bending times. When the Ni-powder content is 10 w/%, the square resistance of the coating after 50 bends changes to  $0.399 \Omega \cdot \text{sq}^{-1}$ . When the Ni-powder content is (20, 30 and 40) w/%, the square resistance of the coating after 50 bends changes to  $0.864 \Omega \cdot \text{sq}^{-1}$ ,  $1.576 \Omega \cdot \text{sq}^{-1}$  and  $2.76 \Omega \cdot \text{sq}^{-1}$ . When the content of Ni powder is 50 w/%, the resistance of the coating after 50 bends changes to  $2.882 \Omega \cdot \text{sq}^{-1}$ . With the increase in the Ni-powder content and bending times, the change value for the coating square resistance shows an upward trend. The reason is that with the increase in the Ni-powder content, the binding force between the paste and the PET film substrate first increases and then decreases, and the bending performance of the coating gradually decreases. After being folded 50 times, the square resistance of the conductive coating with the Ni-powder content of 10 w/% is nearly entirely overlapped as compared to the single conductive filler Ag-Cu conductive coating. The coating with 10 w/% Ni powder



**Figure 8:** Resistance of composite conductive coating varies with the number of bends



exhibits good flexibility and bending performance. When the Ni-powder content amounts to 50 w/%, the adhesion between the paste and the PET film substrate is the poorest, and so are the flexibility and bending performance of the coating.

#### 4 CONCLUSIONS

The created composite conductive paste performs best when the ratio between the Ag-Cu powder, Ni powder and resin is 13 : 2 : 5. The resistivity of the PET film to the substrate is  $7.92 \times 10^{-4} \Omega\text{-cm}$ , and the bonding force between the PET film and the substrate is 20.1 N. It also has excellent corrosion resistance and bending properties. Conductive pastes with various Ni-powder concentrations were subjected to electrochemical corrosion testing. According to the studies, the corrosion resistance of the composite conductive coating with a Ni-powder content of 10 w/% is the best.

#### Acknowledgment

This work was supported by the Natural Science Foundation of Shaanxi Province, China (Grant No. 2022JQ-319).

#### 5 REFERENCES

- Y. Xia, P. Cai, Y. Liu, J. Zhu, Z. Xiao, A low-cost and high-efficiency electro-thermal composite film composed of hybrid conductivity fillers and polymer blends matrix for high-performance plate heater, *Journal of Electronic Materials*, 50 (2021) 6, 3084–3094, doi:10.1007/s11664-021-08873-0
- X. K. Ji, X. C. Jin, C. Hou, G. S. Xiao, X. F. Shu, Loading rate response on shear mechanical properties of conductive adhesive with different silver contents, *Journal of Adhesion Science and Technology*, 34 (2020) 18, 1935–1946, doi:10.1080/01694243.2020.1749405
- P. E. Lopes, D. Moura, L. Hilliou, B. Krause, P. Pötschke, H. Figueiredo, R. Alves, E. Lepleux, L. Pacheco, M. C. Paive, Mixed carbon nanomaterial/epoxy resin for electrically conductive adhesives, *Journal of Composites Science*, 4 (2020) 3, 105, doi:10.3390/jcs4030105
- Q. H. Tian, D. Deng, L. I. Yu, X. Y. Guo, Preparation of ultrafine silver powders with controllable size and morphology, *Transactions of Nonferrous Metals Society of China*, 28 (2018) 3, 524–533, doi:10.1016/S1003-6326(18)64685-0
- W. Gang, W. Hui, Y. Cui, J. Bai, Preparation of micro-sized and monodisperse crystalline silver particles used for silicon solar cell electronic paste, *Journal of Materials Science*, 25 (2014) 1, 487–494, doi:10.1007/s10854-013-1613-4
- Z. Liu, X. L. Qi, H. Wang, Synthesis and characterization of spherical and mono-disperse micro-silver powder used for silicon solar cell electronic paste, *Advanced Powder Technology*, 23 (2012) 2, 250–255, doi:10.1016/j.apt.2011.03.004
- S. N. Liu, S. K. Tam, K. M. Ng, Dual-reductant synthesis of nickel nanoparticles for use in screen-printing conductive paste, *Journal of Nanoparticle Research*, 23 (2021) 3, 1–12, doi:10.1007/s11051-021-05191-8
- T. H. Kim, H. Kim, H. J. Jang, N. Lee, S. H. Lee, Improvement of the thermal stability of dendritic silver-coated copper micro-particles by surface modification based on molecular self-assembly, *Nano Convergence*, 8 (2021) 1, 1–8, doi:10.1186/s40580-021-00265-8
- H. Y. Dong, X. Li, Y. Dong, S. H. Guo, L. F. Zhao, A novel preparation method of electrically conductive adhesives by powder spraying process, *Materials*, 12 (2019) 17, 2793, doi:10.3390/ma12172793
- X. G. Cao, H. Y. Zhang, Fabrication and performance of silver coated copper powder, *Electronic Materials Letters*, 8 (2012) 4, 467–470, doi:10.1007/s13391-012-1110-6
- J. J. Kim, H. W. Lee, V. V. Dabhade, S. R. Kim, W. T. Kwon, D. J. Choi, H. Kim, Y. Kim, Electro magnetic interference shielding characteristic of silver coated copper powder, *Metals & Materials International*, 16 (2010) 3, 469–475, doi:10.1007/s12540-010-0619-1
- S. Y. Zhang, X. Q. Qi, M. Yang, Y. Cao, T. S. Lin, P. He, K. W. Paik, A study on the resistivity and mechanical properties of modified nano-Ag coated Cu particles in electrically conductive adhesives, *Journal of Materials Science: Materials in Electronics*, 30 (2019) 10, 9171–9183, doi:10.1007/s10854-019-01246-8
- S. J. Peighambaroust, H. Rikhtegar, P. P. Mohammadzadeh, A. Mirmohseni, Electrically conductive epoxy-based nanocomposite adhesives loaded with silver-coated copper and silver-coated reduced graphene oxide nanoparticles, *Polymers for Advanced Technologies*, 30 (2019) 8, 1996–2004, doi:10.1002/pat.4632
- A. Mik, B. Ebc, A. Jhl, Improved sinter-bonding properties of silver-coated copper flake paste in air by the addition of sub-micrometer silver-coated copper particles, *Journal of Materials Research and Technology*, 9 (2020) 6, 16006–16017, doi:10.1016/j.jmrt.2020.11.069
- Y. Shi, Y. Wu, Z. Zhu, Modification of aqueous acrylic-polyurethane via epoxy resin postcrosslinking, *Journal of Applied Polymer Science*, 88 (2003) 2, 470–475, doi:10.1002/app.11750
- B. Han, S. Zhu, W. W. Dong, Y. F. Bai, H. Ding, Y. L. Luo, P. Di, Improved electrochemical corrosion resistance of hot-press sintered WC-Al<sub>2</sub>O<sub>3</sub> composites with added TiC in alkaline solutions, *Ceramics International*, 47 (2021) 22, 32168–32178, doi:10.1016/j.ceramint.2021.08.109
- Y. Y. Li, T. S. Cao, D. Y. Wang, J. Zhao, C. M. Bao, Y. K. Song, X. M. Meng, C. Q. Cheng, Electrochemical corrosion of FV520B stainless steel in solutions bearing hydrogen bromide and acetic acid at high temperature from 130 °C to 200 °C, *Corrosion Engineering, Science and Technology*, 56 (2021) 7, 639–647, doi:10.1080/1478422X.2021.193330
- S. P. Rosoiu, S. Costovici, C. Moise, A. Petica, L. Anicai, T. Visan, M. Enachescu, Electrodeposition of ternary Sn-Cu-Ni alloys as lead-free solders using deep eutectic solvents, *Electrochimica Acta*, 32 (2021) 19, 24152–24167, doi:10.1016/j.electacta.2021.139339