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Materials Design of Electrically Conductive Pastes for Stretchable Device Packaging

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Abstract

Materials design concept to suppress increase in electrical resistivity of stretchable printed wires using elastomer-based conductive pastes during a cyclic tensile test is discussed. Silver (Ag) micro-flakes were used as the main conductive fillers in the conductive pastes. Microstructure of the pastes was modified using Ag micro- and nanoparticles to obtain the conductive pastes with bimodal filler-size-distributions. Stretchable wire samples were printed on an elastic textile substrate using these conductive pastes. The electrical resistivity of the wires was measured after curing. As a result, increase in electrical resistivity of the wires during a uniaxial tensile test at 10 % strain was found to be significantly suppressed by using the bimodal conductive pastes.

1. Introduction

Recently, stretchable microsystems [1] have attracted great interest for developing smart devices such as wearable and patchable healthcare devices. To establish stretchable microsystems, novel materials and processing for forming stretchable wires and electrodes are require to be developed. Then, several types of conductive materials, including metals (solid [2] and liquid [3]), organic polymers (electrically conductive polymers), conductive-coated fibers and electrically conductive pastes [4,5], have been used to form stretchable wires and electrodes.

This work examines about stretchable wires and electrodes prepared using the electrically conductive pastes. The electrically conductive pastes composed of elastomer-based binder containing metallic fillers can exhibit an extensive stretchability. Therefore, additive printing processes such as screen-printing using these pastes are examined to form stretchable wires and electrodes.

Although stretchable wires and electrodes are successfully prepared by printing these pastes, several problems are still remained. For example, electrical resistivity of the printed wires and electrodes varied during mechanical deformation. The variation of the electrical resistivity exhibits time-dependency that is caused by the viscoelastic deformation behavior of the elastomer binder. In addition, their electrical resistivity is increased due to fatigue damage during repeated mechanical deformation [6].

In the present work, materials design for improving fatigue resistance of stretchable wires and electrodes printed with elastomer-based conductive pastes is discussed. To improve the fatigue resistance of the stretchable conductive pastes, viscoelasticity of the elastomer-based binder is needed to be controlled. Advanced stretchable conductive pastes can be developed through two different approaches. One is molecular designs for the elastomer-based binder. For example, polymeric microstructure composed of soft and hard segments is designed to control deformability of the conductive pastes when we use polyurethane-based elastomers as the binder. In addition, variation in electrical conductivity due to mechanical deformation is influenced by geometrical factors and size-distribution of conductive fillers. Therefore, control of these factors of fillers is another approach to obtain advanced stretchable conductive pastes.

Here, the effect of filler-size-distribution of fillers on electrical conductivity of polyurethane-based conductive pastes containing silver (Ag) fillers was investigated. Fatigue of stretchable printed wires was found to be suppressed by using the conductive pastes with bimodal filler-size-distributions.

2. Experimental procedure

Preparation of conductive pastes

A polyurethane-based binder (Matsui Shikiso Chemical Co. Ltd., Kyoto, Japan) was selected for preparing conductive pastes. Three types of Ag fillers including micro-flakes (3-10 μm), micro-particles (2-3 μm) and nanoparticles (50 nm) were used. These Ag fillers were mixed into the polyurethane-based binder to prepare the bimodal pastes (flakes/micro-particles and flakes/nanoparticles) up to ~80 wt% (total) Ag loading.

The electrical resistivity of these pastes was measured using the four-point probe method after curing at 100 ºC for 1200 s.

Preparation of stretchable printed wires

The bimodal conductive pastes were screen-printed on a stretchable textile substrate to obtain the printed wire samples for a uniaxial cyclic tensile test.

First, the bottom insulative layer was printed on the substrate using the polyurethane-based binder containing no Ag fillers. After curing at 100 ºC for 1200 s, the conductive pastes with Ag loading were printed on the insulative layer to form a strip line (2mm x 45mm x 30μm). Then, the conductive pastes were cured at 100 ºC for 1200 s.

Uniaxial cyclic tensile test with electrical resistance measurement

A uniaxial cyclic tensile test was conducted at 10 % strain at 0.3 Hz and ambient temperature. The electrical resistance of the printed wire samples was concurrently measured during the tensile test. In this work, the fatigue lifetime of the printed wires was defined as the number of cycles required to reach 1 kΩ at 10 % elongation.

After the cyclic tensile test, the samples were annealed at 100 ºC for 1200 s to examine recovery of electrical resistance of the printed wires.

3. Results and discussion

Electrical resistivity of stretchable printed wires printed with the bimodal conductive pastes

Figure 1 shows electrical resistivity of the printed wire samples (with no elongation) using the bimodal pastes, as a function of micro-particle content within the total filler loading. Because the electrical resistance of the wires exhibited a minimum value, we can recognize appropriate composition of fillers for the bimodal conductive pastes. The appropriate composition for the bimodal pastes was different depending on the size of the Ag particles. In the case of the bimodal pastes containing the micro-particles, the minimum resistivity was obtained at 20 wt% of micro-particles within the total filler loading. By contrast, the electrical resistivity of the bimodal pastes was minimized by the addition of only 3 wt% nanoparticles within the total filler loading.

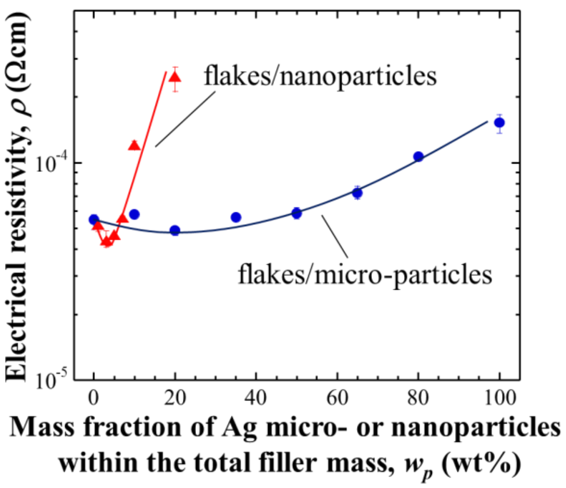

Fig. 1 Electrical resistivity of printed wire samples using bimodal conductive pastes with no tensile strain, as a function of mass fraction of Ag micro- or nanoparticles within the total filler loading.

Fatigue behavior of stretchable wires using single- and bimodal pastes

Variation in the electrical resistance of stretchable wires using the single-modal paste containing Ag flakes and the bimodal pastes was measured during a uniaxial cyclic tensile test. Figure 2 shows the electrical resistance of the wire samples at 10 % strain (loading state) during the cyclic tensile test. A significant increase in electrical resistance was observed during the initial ~1000 cycles of the tensile test. However, the electrical resistance at the loading state became stable beyond ~1000 cycles. In the case of the sample using the single modal pastes, the electrical resistance increased again above ~3000 cycles.

In contrast, the electrical resistance of the samples using the bimodal pastes remained stable at least until 10000 cycles. Therefore, fatigue resistance of the stretchable printed wires can be improved using bimodal pastes.

The improvement of fatigue resistance of the stretchable wires is thought to be achieved by controlling mobility of the polymer chains (deformability) of the binder. To discuss effect of filler-size distribution on deformability of the binder, additional characterizations of the stretchable wires, such as uniaxial tensile test and stress relaxation test, were conducted. As a result, the Ag micro-particles suggested to exhibit different effect with the nanoparticles.

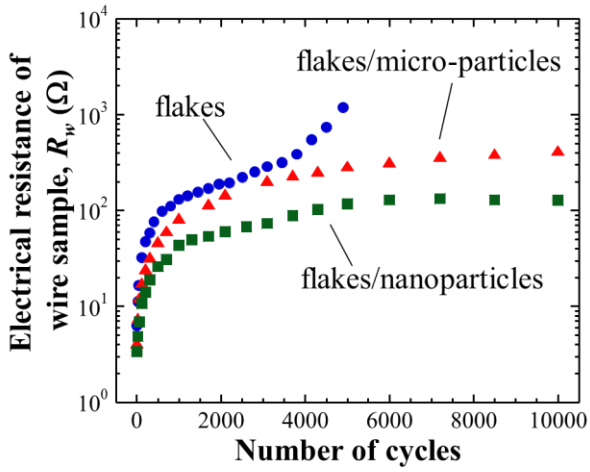

Fig. 2 Variation in electrical resistance of stretchable printed wire samples using single- and bimodal conductive pastes at the loading state during a uniaxial cyclic tensile test at 10 % strain.

4. Conclusions

Variation in electrical conductivity of stretchable wires printed on a stretchable textile substrate using polyurethane-based conductive pastes containing Ag fillers was investigated during a uniaxial cyclic tensile test. As a result, use of the conductive pastes with bimodal filler-size-distributions such as micro-flakes/micro-particles and micro-flakes /nanoparticles is found to be effective to improve fatigue resistance.

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