Flexible Hard Coating (Flex9H®) for Foldable Display Cover Plastic Film

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Abstract

The desire for foldable display seems to be heading toward its zenith. However, its mass production is still impeded by a few technical challenges, specifically, development of flexible cover window films. This invited paper demonstrates innovative flexible hard coating (Flex9H®) for foldable display cover plastic films, which exhibits glass-like pencil hardness, plastic-like flexibility and excellent optical transparency simultaneously.

Author Keywords

Flexible hard coating; Siloxane hybrid material; Foldable display; Cover window film

1. Introduction

Foldable display has received much attention in the last few years as a next-generation technology which can defeat a conflict between downsizing of device and widening of screen [1-3]. Despite the promising future and advances in key enabling technologies, however, the transition toward foldable displays from rigid ones has encountered several technical issues. One of the most decisive hurdles is the identification of solid substitutes for the toughened glass, which has been a prime option as the cover window of rigid displays due to its excellent scratch-resistance and overall protection of the screen.

In foldable displays, the substitute for the toughened glass is likely to be a combination of a plastic film and flexible hard coating. The coating should be optically transparent, and more importantly, it must be as hard as glass to guarantee scratch-resistance and yet be sufficiently flexible to allow a high level of deformation without fracture; for example, the required radius of curvature for foldable displays is less than a few millimeters.

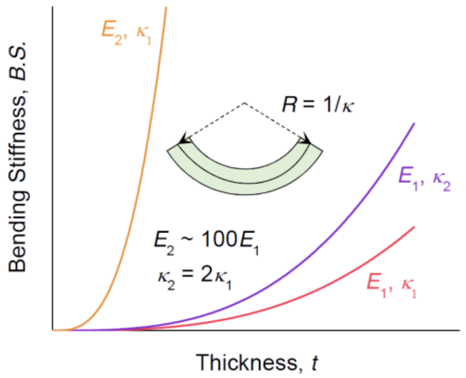

Figure 1. Schematic of bending stiffness of films according to modulus (*E*), radius of curvature (*R*) and thickness (*t*).

Furthermore, in order to easily fold and unfold the display, the coating material should be required to have a plastic-like low modulus and its thickness should be reduced if possible as shown in Figure 1 in accordance with the well-known equation for bending stiffness (*B.S.*) of a film [4];

$$B.S. \approx \frac{Et^3}{12(1-\nu^2)R}$$

, where ν is Poisson's ratio. In other words, hard coating materials for foldable displays exhibit glass-like wear resistance, a plastic-like modulus and a high failure strain limit with outstanding optical transparency. Besides, when it is coated on the plastic substrate, it must be as thin as possible. The technical difficulties in designing such coating materials reside in the fact that, in general, hardness and flexibility are mutually exclusive, that is, hard materials are mostly stiff and brittle, even if they are thin.

Here, we provide insights into the design of a transparent coating of an epoxy-siloxane hybrid nanocomposite that exhibits exceptional levels of both scratch-resistance and flexibility. Our coating material, which is named Flex9H®, in combination with a plastic film can serve as a protective cover window in a foldable display, one of the most viable futuristic forms of flexible display.

2. Discussion and Conclusion

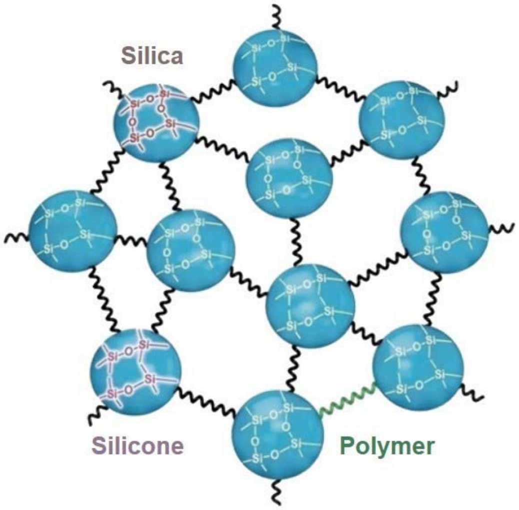

Figure 2. Schematic illustration of the structural concept of Flex9H®.

Figure 2 shows the design principle of Flex9H® coating material, which consists of siloxane nano-building blocks densely linked by cycloaliphatic epoxy polymer tethers. The siloxane networks, which are the smallest elements on the nanometer length scale, were fabricated by sol-gel condensation of alkoxysilanes with cycloaliphatic epoxy functional groups. In order to realize innovative characteristics which come from silica's strength, silicone's elastic resilience and polymer's flexibility, the siloxane nano-building blocks were maximally cross-linked by polymer networks, synthesized using UV-initiated cationic ring-opening polymerization (CROP) of cycloaliphatic epoxide groups. In general, the CROP of multi-functional molecules such as our system results in low degree of cross-linking due to the isolation of growing chains.

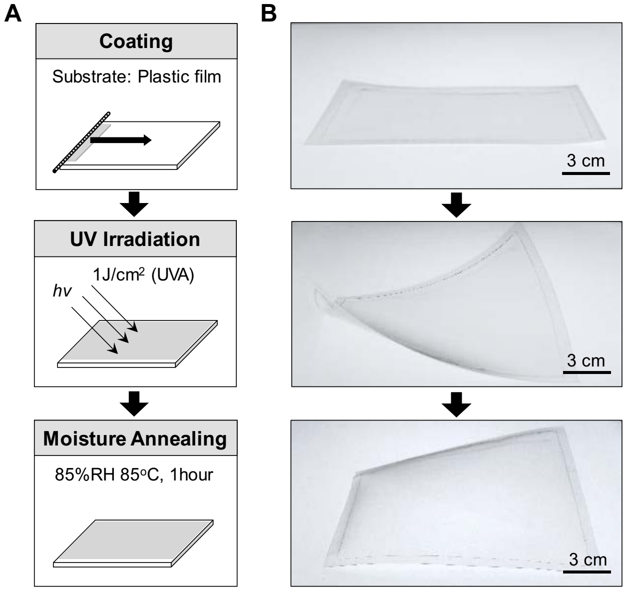

Figure 3. Unique fabrication procedure of Flex9H® coating. A) Schematic flowchart of the curing process of Flex9H® coating. B) Curl change during the fabrication of Flex9H® coating film.

By designing unusual curing procedure which is a two-step process, *i.e.*, UV irradiation and moisture annealing as shown in Figure 3A, we increased the epoxy conversion to the maximum level [5]. The major difference from conventional curing methods is water molecules are intentionally injected into the coating material by moisture annealing after UV curing. The injected H_2O molecules play the important role as chain transfer agents, which can move active sites of the isolated growing chains to neutral epoxides. Based on this strategy of fabrication process, resultant Flex9H® simultaneously obtains all of the benefits of silica, silicone and polymer structure, that is, wear resistance, high-level of elastic resilience and flexibility respectively. Figure 4 shows qualitative stress-strain curve of Flex9H® coating, exhibiting glass-like strength, no plastic deformation and plastic-like elastic strain limit.

Furthermore, there is an additional technical advantage in our curing process. Conventional UV-curable coating systems usually show volume shrinkage after UV irradiation due to the cross-linking, which sometimes results in severe tensile stress in the coating layer and eventually leads to the failure by cracks of the coating. In our system, however, the injected H_2O molecules, which chemically react with the cationic active epoxides, permanently expand volume and lead to compensation for volume shrinkage by the UV curing as shown in Figure 3B. By using our curing mechanism and characteristic, the degree of curl of Flex9H® coating film can be controlled according to the intended use.

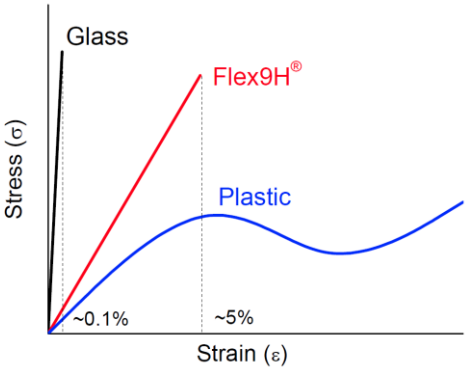

Figure 4. Schematic of stress (σ)-strain (ε) behavior of Flex9H® coating.

In order to investigate the potential of Flex9H® for flexible hard coatings, Flex9H® with varying the coating thickness was fabricated on a 50-μm-thick colorless polyimide (CPI) film which was chosen as the plastic substrate due to its better mechanical reliability compared with other plastics. Figure 5 shows the representative image of Flex9H® coated on CPI film which has a flat surface without curl and is optically transparent in the visible range, as characterized by the UV-Vis spectrometer; the transmittance at 550nm is about 90%.

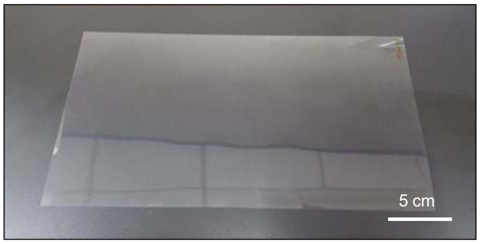

Figure 5. A photograph of Flex9H® coated on CPI film.

The scratch resistance of Flex9H® coated on CPI film was evaluated through pencil hardness tests with a constant applied load of 1 kgf according to KS M ISO 15184 and the results are shown in Figure 6. It should be noted that Flex9H® coating at a thickness of 40 μm was rated with a pencil hardness of 9H, the highest hardness index, comparable to that of the glass surface.

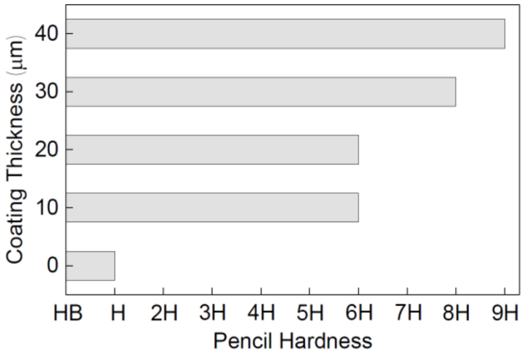

Figure 6. Plot of pencil hardness versus Flex9H® coating thickness (substrate: 50-μm-thick CPI Film).

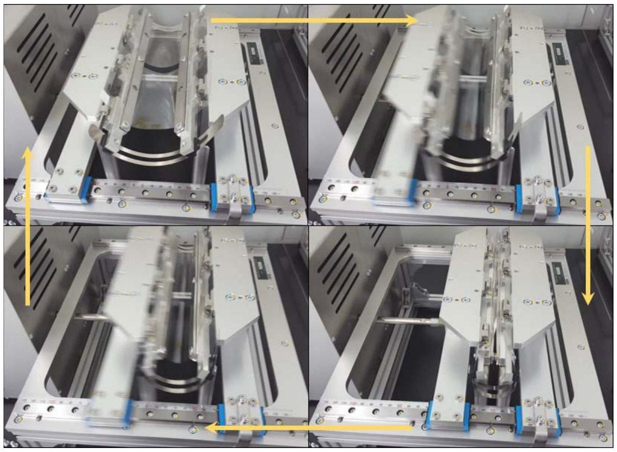

Figure 7. Dynamic folding test for evaluation of inward or outward foldability of the Flex9H® coating films.

Furthermore, the flexibility under a dynamic folding condition was confirmed by using a tension free U-shape folding test machine, of which operation process is described in Figure 7. According to the practical purpose of application, the folding condition can be largely divided into two distinctive folding modes, *i.e.*, inward- and outward-folding. In case of the inward-folding mode, an external compressive stress is applied to the coating surface. On the contrary, the outward-folding causes an external tensile stress. In general, most of hard coating materials are more vulnerable in tension mode due to the low level of elongation. The maximum strain on the coating surface can be easily estimated using the following equation [6];

$$\varepsilon = -\frac{y}{R}$$

, where y is the distance from the neutral axis and R is radius of curvature at the neutral axis. Therefore, in the outward-folding mode, the coating thickness is extremely important and should be thin as possible to realize small folding radius. Actually, Flex9H® coatings on CPI films showed excellent flexibility under the dynamic inward-folding condition with a bending radius of <1mm over more than 100,000 cycles without permanent deformation regardless of the coating thickness (<50 μm). In the outward-folding mode, Flex9H® coatings on CPI films were optimized and showed a pencil hardness of >6H and a bending radius of <5mm over more than 10,000 cycles.

Another important point for the actual commercialization of coating materials is related to the stability against heat, moisture and ultra-violet (UV) light. Heat and moisture resistance was checked by storage of the Flex9H®coated on CPI film in a temperature/humidity test chamber with a constant condition of 85% RH at 85 °C for 120 hours. UV stability was also confirmed by exposing the Flex9H® coated on CPI film for 72hrs to a UV-B lamp of which power is 20W. The distance between the film and the lamp was kept at a constant 20 cm. After the two reliability tests, we could not find any significant degradation in mechanical properties, *i.e.*, pencil hardness and dynamic foldability of Flex9H® coating. Besides, the values of yellow index (YI) of Flex9H® coating on CPI film before and after the reliability tests were measured using spectrophotometer in accordance with ASTM D1925. In case of the heat/moisture reliability test, there was no meaningful change of YI. Also, the UV stability test resulted in trivial increase of < 1.0 in YI.

Table1. The specification of Flex9H® coating for foldable display cover plastic films.

Pencil hardness	9H (Inward-folding) >6H (Outward-folding)
Foldability	<1mm (Inward-folding) <5mm (Outward-folding)
Optical transmittance	>90%
Haze	<1.0
Yellow Index (YI)	<1.0
Heat and moisture resistance	85 °C/85% RH for 120 hrs
UV resistance	UV-B (20W, 20cm) for 72 hrs ΔYI<1.0

3. Impact

The development of optical coating materials that simultaneously exhibit outstanding scratch resistance (hardness) and resist a high level of deformation (flexibility) is a fundamental prerequisite for the implementation of foldable displays. In this paper, we demonstrate that such a coating can be fabricated from the epoxy-siloxane hybrid nanocomposite by a simple, mass-producible method. Our findings can contribute to the mass commercialization of high performance foldable displays.

4. Acknowledgements

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