Assessment of a Cyclic Bending Test Method for Printed Flexible Supercapacitor

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***Abstract*— Printed flexible supercapacitor (SC) is seen as an attractive alternative to replace batteries as energy storage unit in energy autonomous sensors. This paper assesses a cyclic bending test method for printed flexible SC. The test is evaluated using four material stacks for printed SC in five different bending radii. The measurement system analysis (MSA) found that the calculated bending radii under all test conditions exhibit variation within a range of 0.3 mm, and the variation takes up less than 8% of bending radius. The measurement system is subjected to 9% total variation, which is within acceptable range. The variation was mainly caused by the uneven thickness distribution across the SC due to its structure. Thus, the variation caused by the test method and measurement is even smaller. In addition, the SC's bending radius subjected to even smaller variation in cyclic bending test. These indicate that this test method is highly reliable and repeatable for evaluating flexible SC.**

Keywords—cyclic bending test; printed electronics; flexible electronics; supercapacitor

I. INTRODUCTION

With the approaching era of energy autonomous sensors, SC has been increasingly concerned energy storage device for powering sensors for its advantages in high power density, long shelf life, quick charging capability, and good cycling stability [1-3]. Furthermore, printed electronics provides simple, low-cost, and eco-friendly methods for fabricating SC [4]. For SC, thick films (on order of 100 µm) of active material are needed, whereas the print resolution is not so critical [5-6], and therefore, doctor-blade coating [5, 7] is an ideal fabrication method regarding these issues due to its good control of the printed layer's thickness, simplicity, and low-temperature operation.

To evaluate the robustness of flexible SC against mechanical impacts, researchers have used some bending tests together with electrical characterization [8-18]. Cyclic bending tests, as presented in Table I, have been commonly used to flexible electronics, as they apply repeated impact and enables to evaluate higher reliability. The 3-point, 4-point, and cyclic folding test methods are not suitable for testing a wide range of bending radius. Cylinder-assisted test has also been used as it can control the bending radius precisely. Happonen *et al*. [19] used it to test the bending reliability of flexible conductors. Wright *et al*. [20] designed

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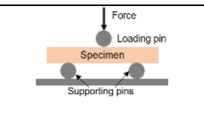
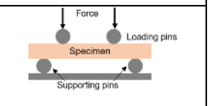
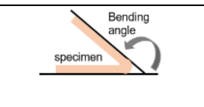
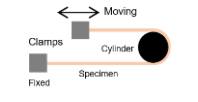
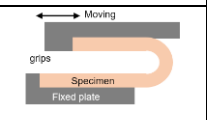
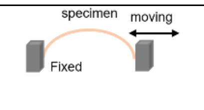
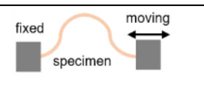
TABLE I. DESCRIPTION OF CYCLIC BENDING TEST METHODS

Methods	Schematic	Description
3-point bending test [8]		Good for testing stiffer materials with large bending radius. Not suitable to light samples, samples with low stiffness. Not suitable for testing small bending radius. Samples fixture can be challenging
4-point bending test [8]		
folding [9, 11, 13]		Localized impact. Suitable for testing small bending radius. The bending angle can be well controlled.
cylinder assisted cyclic bending		Suitable for testing the fixed bending radius. The whole sample is subjected to the same impact.
parallel plate cyclic bending [13-14]		
arc-shape cyclic bending [12, 15]		Dynamic bending radius. Fixing the sample stably is difficult.
cyclic bending [10, 16-18]		Dynamic bending radius. Challenge in determining bending radius and evaluating its stability.

a test machine in which the flexible electronics are against cylinder with constant tension for reliability study. Another method uses two parallel plates to control bending radius, which has been used to study the reliability of printed vias [21-22], thin metal films [23], and flexible devices [24]. However, these methods do not simulate the case when the device is bent without being against any object.

The last two methods in Table I overcome this limitation. Yao *et al.* [12] and Moon *et al.* [25] used arc-shape bending method to study the electrical properties of flexible devices with the increasing number of test cycle. However, it was only for a single device with simple stacking, which make the bending radius measurement easy by controlling the distance between two clamps. However, for testing different devices, when the stacking and material are different, their bending radii are different even when the clamps distance is identical, which is confirmed by the results in Table V. The last method in Table I overcomes this problem, whereas determining the bending radius is difficult in this method.

In this paper, we introduce a method to determine the bending radius in this cyclic bending test, and to assess its repeatability and accuracy using MSA and cyclic test. It aims at recommending this verified method for testing flexible SC.

II. Materials and Methods

A. *Sample Fabrication*

The four substrate-layer laminates for fabricating SCs are described in Table II. They were made with 25 µm thick EL-92734 adhesive using Drytac JM26 tabletop laminator at VTT. The materials used in SC fabrication are presented in Table III. The current collector and electrode were printed onto the laminated substrate using a doctor blade coater (mtv messtechnik) with polyimide film mask, and cured in the oven at 95 ℃ for 45 min and 70 ℃ for 30 min, respectively. The details of fabrication process were reported by Keskinen *et al*. [2, 26]. The face-to-face sandwich structured SCs were assembled using a customized alignment tool. The structure of SCs and a sample are presented in Fig. 1. Designing safety areas in SC for clamping is needed for avoiding the strong impact of small bending radius caused be clamping.

B. *Test Setup*

The cyclic bending test was conducted using ESM 303 Mark-10 motorized tension test stand, as presented in Fig. 2. The sample was firstly fixed by the clamps but without significant tension. The lower testing grips were kept static whereas the upper testing grips moved at the speed of 350 mm/min. The bending radius is defined by the movement of upper grips. The higher and lower limits were set to define the moving trajectory of the upper grips, the lower limit determines to what extend the flexible sample would be bent, which determine the bending radius. The number of bending test cycles was to be set in a way that it exhibits the failure process clearly.

TABLE II. Four Substrate and Layer Configuration of SC.

	Substrate	Layer	Thickness of Substrate-Layer Laminate
No.1	PLA	Al	130 – 145 µm
No.2	PET	Al	150 – 165 µm
No.3	PLA	Barrier	160 – 170 µm
No.4	PET	Barrier	170 – 175 µm

TABLE III. Materials Used for SC Fabrication.

Components	Materials	Thickness	Model
Substrate	High heat PLA	100 µm	developed by VTT [27]
	PET	125 µm	Melinex ST506
Laminated Layer	Aluminum foil	15 µm	Nanografi
	Barrier foil	50 µm	3M FTB 3-50
Current collector	Graphite ink	100 µm	Henkel PF407C
Electrode	Activated carbon	100 µm	Kuraray YP-80F
	Chitosan binder		Sigma-Aldrich 50494
Separator	Cellulose paper	40 µm	Dreamweaver Titanium 40
Electrolyte	NaCl (aq)	-	NaCl : H2O = 1 : 5 (99.8% purity)
Sealant	Adhesive tape	130 µm	3M 468MP-200MP

To determine the bending radius, the sample was imaged using a Canon G11 camera when the upper grips moved to the position of low limit in the first cycle. This is the point when the sample is bent to its smallest curvature and subjected to strongest impact under the testing condition. The camera was held by a tripod, the height and focusing were adjusted for optimal image quality. A circular shape was fit to the bent round shape to determine the bending diameter, as shown in Fig. 2. The ratio of the drawn circle diameter d (in pixel) and the distance between upper and lower grips h (in pixel) is used to compute the bending radius r (in mm), since the real distance between the upper and lower grips H (in mm) is known. Thus, the real bending radius r (in mm) can be computed by the equation:

$$r = d * H / 2h \tag{1}$$

C. *Experiment*

To assess the proportion of the variation caused by the measurement and operation, and to evaluate the repeatability of the test method, a MSA was carried out. The SCs with four laminated substrates described in Table II were tested when H = 30, 25, 20, 15, and 10 mm, respectively, in random order, and each case was tested for three times to study the variation of bending radius and the test repeatability. It is important that for every repetition, the sample is removed from the clamps and fixed to them again, which is to take full consideration of variations caused by operation. The fixed height, instead of the fixed bending radius, was used because the SCs fabricated with different substrates exhibit different bending radius under the same height, thus controlling the height is easier for operation.

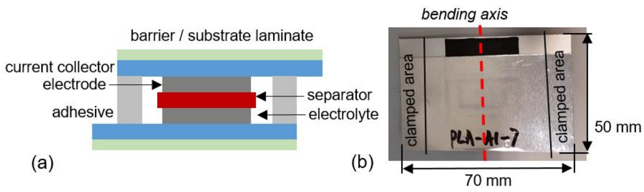


Figure 1. (a) Schematic of printed flexible supercapacitor, (b) a fabricated flexible supercapacitor sample.

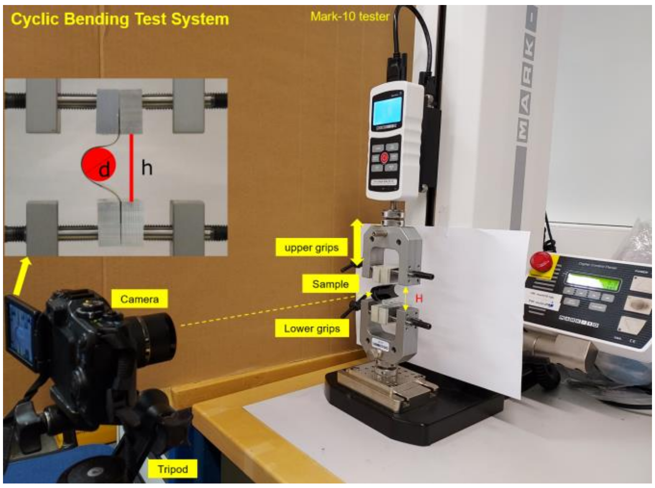


Figure 2. cyclic bending test setup. (Note: The initial distance between lower and upper grips was 50 mm.)

III. Results and Discussion

A. *Gage R&R Study for Bending Radius*

Gage analysis was performed with one operator, 4 types of SC samples, and 5 bending distances. The calculated bending radii were analyzed using Minitab. The results show that the process performance was subjected to 9% total variation, which is within the acceptable range (< 10%) by criterion. The results of variation and standard deviation are presented in Table IV. This level of variation indicates that the test method has high repeatability.

B. *Variations in Calculated Bending Radius*

The bending radii of SCs under each test condition were measured three times, and the variations of the calculated bending radii are presented in Table V. The bending radius variations of 15/20 test conditions were lower than 0.2 mm, and under almost all test conditions, the variations were within 0.3 mm. Considering their average bending radii, the variation range of bending radius takes less than 8% of actual bending radius, with 17/20 test conditions the proportion is less than 5%, which is a rather low level of proportion. This proves that the bending radius is subjected to small variation in this test method, and the test is highly repeatable.

The variation can be from measurement system, initial differences in flexible SC samples caused by variation in material thickness, operation, and imaging. However, it is mainly caused by the uneven thickness distribution of the SC sample due to its structure. Since the central area of the SC consists of printed layers of current collectors, electrodes, and separator paper, which lead to higher thickness than the edge areas. When SC was bent, it was slightly tilted rather than being a standard round, which was shown in the images. Otherwise, the variations caused by measurement would be rather small. Therefore, we can confidently believe that the proportion of variations in bending radius caused by the test method would be actually smaller than 9%.

C. *Bending Radius During Cyclic Bending*

Repeatability of the bending radius during the cycling bending test was assessed by measuring the bending radii of seven samples as a function of number of test cycle. The bending radius was measured in 50 cycle intervals. The results show that the variations are rather small. An example is presented in Fig. 3, which demonstrate the bending radii of the 7 printed flexible SCs with PLA/barrier laminated substrate in 500 cycles test. 2/7 samples survived electrically after 500 cycles test. The bending radii exhibit a variation in the range of 0.03 mm, which is significantly smaller than the variation caused by SC's uneven thickness distribution, setup, and operation. This indicates the high stability in bending radius during test.

IV. Conclusions

This paper assesses the repeatability of a cyclic bending test method that is suitable for evaluating the reliability with a purpose of failure analysis of printed flexible SC. The variables (SC substrate & bending distance) were integrated into 20 test conditions, and the bending radius under each test condition was measured 3 times. The conclusions can be drawn from MSA and cyclic test are: (1) The total variation in bending radius measurement system is within acceptable range, the measurement is repeatable. (2) The bending radius for the specific device must be defined, as the bending radii of different devices are different under the same bending distance. (3) The bending radius during cyclic bending can almost be kept the same, at least within 451 cycles. Thus, this cyclic bending test method is verified to be repeatable and reliable for evaluating printed flexible SC.

TABLE IV. STANDARD DEVIATION AND VARIATION BY SOURCE

Source	Standard Deviation	Study Variation
Total Gage	0.093	8.97%
Repeatability	0.093	8.97%
Part-to-Part	1.032	99.60%
Study Variation	1.036	100.00%

TABLE V. RANGE OF BENDING RADIUS VARIATION UNDER DIFFERENT TEST CONDITIONS

Laminated Substrate	Height (mm)	Range of bending radius variation	Average bending radius	Proportion of variation over bending radius
PLA/Al	10	0.08 mm	4.51 mm	1.77 %
	15	0.18 mm	4.95 mm	3.63 %
	20	0.29 mm	4.86 mm	5.97 %
	25	0.15 mm	5.39 mm	2.78 %
	30	0.02 mm	6.95 mm	0.29 %
PET/Al	10	0.19 mm	4.49 mm	4.23 %
	15	0.17 mm	5.41 mm	3.14 %
	20	0.29 mm	5.41 mm	5.36 %
	25	0.15 mm	6.29 mm	2.38 %
	30	0.13 mm	6.96 mm	1.87 %
PLA/barrier	10	0.33 mm	4.34 mm	7.60 %
	15	0.19 mm	5.45 mm	3.49 %
	20	0.10 mm	6.29 mm	1.59 %
	25	0.11 mm	6.38 mm	1.72 %
	30	0.11 mm	7.73 mm	1.42 %
PET/barrier	10	0.14 mm	4.59 mm	3.05 %
	15	0.21 mm	4.88 mm	4.30 %
	20	0.10 mm	5.36 mm	1.87 %
	25	0.25 mm	6.56 mm	3.81 %
	30	0.06 mm	7.31 mm	0.82 %

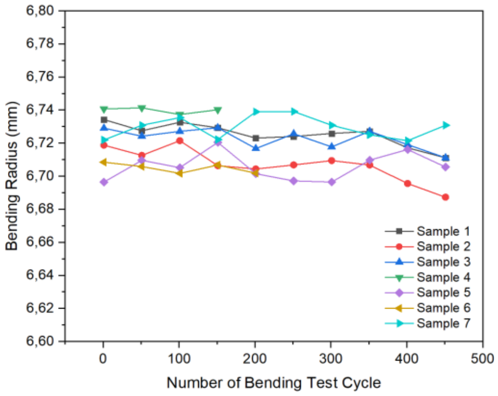


Figure 3. An example of small variations of bending radius of SCs with the increasing number of bending test cycle. (Note: Sample 4 and 6 failed seriously after 200 and 250 cycles of test, after which they were not tested)

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