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# 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  $\mu\text{m}$ ) 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

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]		Localized impact. Suitable for testing small bending radius. The bending angle can be well controlled.
folding [9, 11, 13]		Suitable for testing the fixed bending radius. The whole sample is subjected to the same impact.
cylinder assisted cyclic bending		Dynamic bending radius. Fixing the sample stably is difficult.
parallel plate cyclic bending [13-14]		Dynamic bending radius. Challenge in determining bending radius and evaluating its stability.
arc-shape cyclic bending [12, 15]		
cyclic bending [10, 16-18]		

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.

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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  $\mu\text{m}$  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  $^{\circ}\text{C}$  for 45 min and 70  $^{\circ}\text{C}$  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 by 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 extent 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 $\mu\text{m}$
No.2	PET	Al	150 – 165 $\mu\text{m}$
No.3	PLA	Barrier	160 – 170 $\mu\text{m}$
No.4	PET	Barrier	170 – 175 $\mu\text{m}$

TABLE III. MATERIALS USED FOR SC FABRICATION.

Components	Materials	Thickness	Model
Substrate	High heat PLA	100 $\mu\text{m}$	developed by VTT [27]
	PET	125 $\mu\text{m}$	Melinex ST506
Laminated Layer	Aluminum foil	15 $\mu\text{m}$	Nanografi
	Barrier foil	50 $\mu\text{m}$	3M FTB 3-50
Current collector	Graphite ink	100 $\mu\text{m}$	Henkel PF407C
Electrode	Activated carbon	100 $\mu\text{m}$	Kuraray YP-80F
	Chitosan binder		Sigma-Aldrich 50494
Separator	Cellulose paper	40 $\mu\text{m}$	Dreamweaver Titanium 40
Electrolyte	NaCl (aq)	-	NaCl : H <sub>2</sub> O = 1 : 5 ( 99.8% purity)
Sealant	Adhesive tape	130 $\mu\text{m}$	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:

$$r = d * H / 2h \quad (1)$$

### 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.

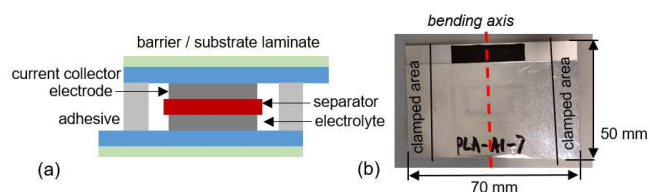


Figure 1. (a) Schematic of printed flexible supercapacitor, (b) a fabricated flexible supercapacitor sample.

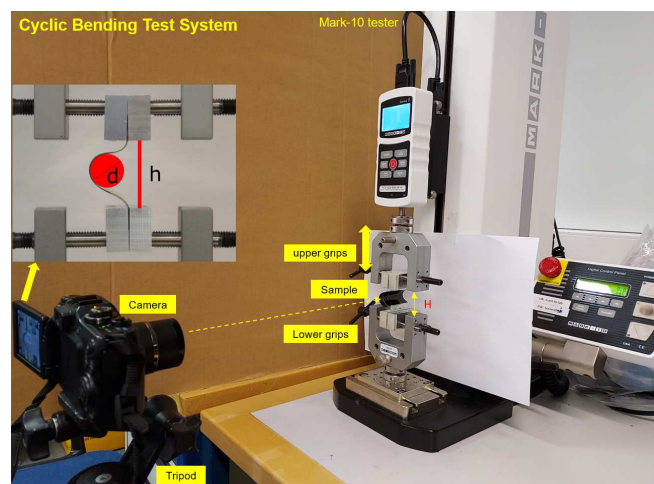


Figure 2. cyclic bending test setup. (Note: The initial distance between lower and upper grips was 50 mm.)

## 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

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/AI	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/AI	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 %

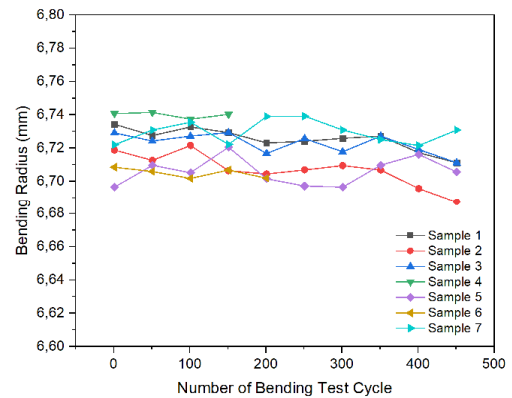


Figure 3. An example of small variations of bending radius of SCs with the increasing number of bending test cycle. (Note: Sample 4 and 6 failed seriously after 200 and 250 cycles of test, after which they were not tested)

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.

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## REFERENCES

- [1] R. Kötz, and M. Carlen, "Principles and applications of electrochemical capacitors," *Electrochimica Acta*, vol. 45, 2483–2498, May 2000.
- [2] J. Keskinen, S. Tuurala, M. Sjödin, K. Kiri, L. Nyholm, T. Flyktman, *et al.*, "Asymmetric and symmetric supercapacitors based on polypyrrole and activated carbon electrodes," *Synthetic Metals*, vol. 203, 192–199, May 2015.
- [3] S. Palchoudhury, K. Ramasamy, R. K. Gupta, and A. Gupta, "Flexible supercapacitors: A materials perspective," *Frontiers in Materials*, vol. 5, 83, Jan. 2019.
- [4] Y-Z. Zhang, Y. Wang, T. Cheng, L-Q. Yao, X. Li, W.Y. Lai, *et al.*, "Printed supercapacitors: materials, printing and applications," *Chem. Soc. Rev.*, vol. 48, 3229–3264, May 2019.
- [5] S. Lehtimäki, *Printed Supercapacitors for Energy Harvesting Applications*. Tampere University of Technology Publication 1463, 2017.
- [6] F. Béguin and E. Frackowiak (ed), *Supercapacitors: Materials, Systems, and Applications*. Wiley-VCH Verlag GmbH & Co. KGaA, March 2013.
- [7] J. Keskinen, *Supercapacitors on Flexible Substrates for Energy Autonomous Electronics*. Tampere University of Technology Publication 1562, 2018.
- [8] S. Shetty and T. Reinikainen, "Three- and Four- Bend Testing for Electronic Packages," *J. Electron. Packag.* vol. 125, 556–556, Dec. 2003.
- [9] R. Zhang, Y. Xu, D. Harrison, J. Fyson, and D. Southee, "A study of the electrochemical performance of strip supercapacitors under bending conditions," *Int. J. Electrochem. Sci.*, vol. 11, 7922–7933, Aug. 2016.
- [10] S.-S. Lee, K-H. Choi, S-H Kim, and S-Y. Lee. "Wearable supercapacitors printed on garments," *Adv. Func. Mater. Weinheim*. vol. 28, Issue 11, 1705571, Jan. 2018.
- [11] H. Wang, M. Totaro, S. Veerapandian, M. Ilyas, M. Kong, U. Jeong, and L. Beccai, "Folding and bending planar coils for highly precise soft angle sensing," *Adv. Mater. Technol.* vol. 5, 2000659, Nov. 2020.
- [12] J. Yeo, G. Kim, S. Hong, M. S. Kim, D. Kim, J. Lee, *et al.*, "Flexible supercapacitor fabrication by room temperature rapid laser processing of roll-to-roll printed metal nanoparticle ink for wearable electronics application," *J. Power Sources*, vol. 246, 562–568, Jan. 2014.
- [13] C-Y. Chen, H. Qin, H-P. Cong, S-H. Yu, "A Highly stretchable and real-time healable supercapacitor," *Adv. Mater.* vol. 31. Issue 19, 1900573, March 2019.
- [14] P. Li, Y. Li, Z. Zhang, J. Chen, Y. Li, and Y. Ma, "Capillarity-driven assembly of single-walled carbon nanotubes onto nickel wires for flexible wire-shaped supercapacitors," *Mater. Sci. Energy Technol.* vol. 1, 91–96, Dec. 2018.
- [15] B. Li, N. Hu, Y. Su, Z. Yang, F. Shao, G. Li, *et al.*, "Direct inkjet printing of aqueous inks to flexible all-solid-state graphene hybrid micro-supercapacitors," *ACS Appl. Mater. Interfaces*, vol. 11, 46044–46053, Nov. 2019.
- [16] C. Kim and C. H. Kim, "Universal testing apparatus implementing various repetitive mechanical deformations to evaluate the reliability of flexible electronic devices," *micromachines*, vol. 9, 492, Sept. 2018.
- [17] D. Lee, H-W. Kim, J-M. Kim, K-H. Kim, and S-Y. Lee, "Flexible/rechargeable Zn–Air batteries based on multifunctional heteronanomaterial architecture," *ACS Appl. Mater. Interfaces*, vol. 10, 22210–22217, June 2018.
- [18] X. Wu, Y. Xu, Y. Hu, G. Wu, H. Cheng, Q. Yu, *et al.*, "Microfluidic-spinning construction of black-phosphorus-hybrid microfibrils for non-woven fabrics toward a high energy density flexible supercapacitor," *Nat. Commun.*, vol. 9, 4573, Nov. 2018.
- [19] T. Happonen, T. Ritvonen, P. Korhonen, J. Häkkinen, and T. Fabritius, "Modeling the lifetime of printed silver conductors in cyclic bending with the Coffin-Manson relation," *IEEE Trans. Device Mater. Reliab.* vol. 16, 25–29, March 2016.
- [20] D. N. Wright, A-S. B. Vardoy, B. D. Belle, and M. V. Taklo, "Bending Machine for Testing Reliability of Flexible Electronics," 2017 IMAPS Nordic Conference on Microelectronics Packaging (NordPac 2017), Gothenburg, Sweden, 18-20 June 2017, 6
- [21] M. Kujala, T. Kololuoma, J. Keskinen, and D. Lupo, "Screen Printed Vias for a Flexible Energy Harvesting and Storage Module," 2018 International Flexible Electronics Technology Conference, pp. 1-6, 2018.
- [22] M. Kujala, T. Kololuoma, J. Keskinen, D. Lupo, M. Mäntysalo, and T. M. Kraft, "Bending reliability of screen-printed vias for a flexible energy module," *npj Flex Electron* 4, 24, September 2020.
- [23] B-J. Kim, H-A-S. Shin, S-Y. Jung, Y. Cho, O. Kraft, I-S. Choi, and Y-C. Joo, "Crack nucleation during mechanical fatigue in thin metal films on flexible substrates," *Acta Materialia*, vol. 61, 3473 – 3481, May 2013.
- [24] J. Lewis, "Material challenge for flexible organic devices," *Materials Today*, vol. 9, 38 – 45, April 2006.
- [25] H. Moon, H. Lee, J. Kwon, Y. D. Suh, D. K. Kim, I. Ha, *et al.* "Ag/Au/Polypyrrole core-shell nanowire network for transparent, stretchable and flexible supercapacitor in wearable energy devices," *Sci. Rep.* 7, 41981, Feb. 2017.
- [26] J. Keskinen, A. Railanmaa, and D. Lupo, "Monolithically prepared aqueous supercapacitors" *J. Energy Storage*, vol. 16, 243 – 249, April 2018.
- [27] E. Luoma, M. Välimäki, T. Rokkonen, H. Säskilähti, j. Ollila, J. Rekilä, and K. Immonen, "Oriented and annealed poly(lactic acid) films and their performance in flexible printed and hybrid electronics," *J. Plast. Film Sheeting*, vol. 37, 429–462, Feb. 2021. K. Elissa, "Title of paper if known," unpublished.