



# Cellulose electro-active paper: from discovery to technology applications

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Cellulose electro-active paper (EAPap) is an attractive material of electro-active polymers family due to its smart characteristics. EAPap is thin cellulose film coated with metal electrodes on both sides. Its large displacement output, low actuation voltage, and low power consumption can be used for biomimetic sensors/actuators and electromechanical system. Because cellulose EAPap is ultra-light weight, easy to manufacture, inexpensive, biocompatible, and biodegradable, it has been employed for many applications such as bending actuator, vibration sensor, artificial muscle, flexible speaker, and can be advantageous in areas such as micro-insect robots, micro-flying objects, micro-electromechanical systems, biosensors, and flexible displays.

**Keywords:** cellulose, electro-active paper, actuator, piezoelectrics, smart material

## INTRODUCTION

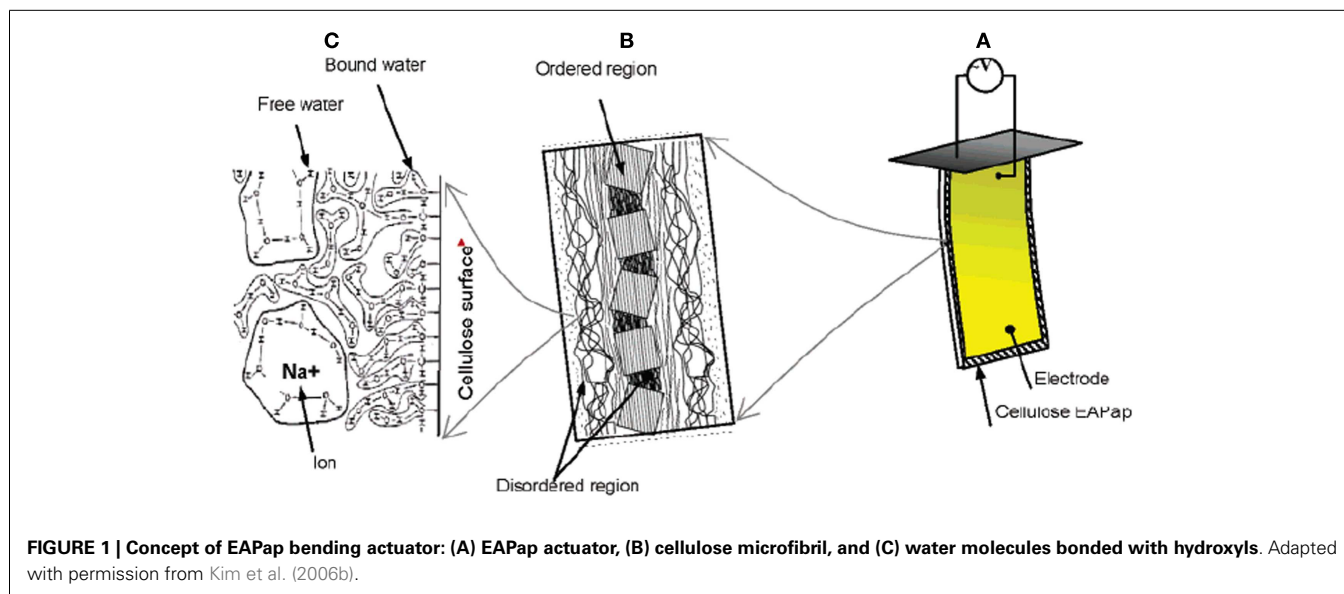
In last few decades, electro-active polymers (EAPs) have received significant attention due to their ability to fabricate large displacement actuators. Cellulose is an endless source of raw materials for environmentally friendly and biocompatible products, and its offshoot are used for optical films, coatings, laminates, pharmaceuticals, and textiles. The shear piezoelectricity in polymers of biological origin such as cellulose and collagen was reported very early in 1950; Fukada experimentally demonstrated the piezoelectric coefficients of wood and verified that oriented cellulose crystallites are responsible for the observed piezoelectricity (Fukada, 2000). The piezoelectric effect exhibited by cellulose film depends on various factors, type of wood, environmental conditions, and orientation of samples. Regardless of these early studies, the prospects of cellulose as an ultra light weight material for electromechanical transduction applications have not been fully explored. The early investigation on EAPap actuator characterized by chemically treated paper with thin electrodes on its both sides shows reasonable performance (Kim and Seo, 2002). When  $2 \text{ MVm}^{-1}$  of excitation electric field was applied to the paper actuator, more than 3 mm tip displacement was observed from the 30 mm long paper film. However, the actuation frequency was low and force output of the EAPap actuator was also small. The force and frequency bandwidth was improved by variation in thickness in the EAPap material at the cost of low actuation voltage and large bending displacement (Yun et al., 2007). In order to enhance the performance of EAPap actuator, conducting polymer coating on EAPap was suggested, which was previously gold electrodes coated (Deshpande et al., 2005). The electrode thickness plays a key role in the performance of actuator. Other techniques employed to achieve smart actuators were use of a single-walled carbon nanotube/polyaniline coated cellulose electro-active paper (EAPap) hybrid actuator and

by mixing multi-walled carbon nanotubes and cellulose (Yun et al., 2006; Yun and Kim, 2007). Carbon nanotubes-mixed cellulose-EAPap actuator exhibited the highest mechanical power output. The investigation on in-plane strain under electric fields reveals that high Young's modulus of EAPap is promising for its use as artificial muscle. The electrical power consumption was  $<8 \text{ mW/cm}^2$ , which is beyond the safety limit of microwave power in air, which direct to the possibility for achieving microwave driven EAPap actuators (Kim et al., 2007).

The potential of cellulose EAPap for smart application is promising. Addition of poly(ethylene oxide)-poly(ethylene glycol) (PEO-PEG), conductive polymer, and carbon nanotubes coatings improved the displacement output and force output of EAPap actuators (Mahadeva et al., 2009a). Cellulose EAPap can be characterized as biosensor since it is biodegradable, biocompatible, capable of broad chemical modification, and hydrophilic (Kim et al., 2008d). The direct piezoelectric effect extends its possibility to be used as self powered vibration and strain sensor for structural health monitoring, energy scavenging transducers, flexible speaker in acoustic applications, and beam vibration control.

## EAPap DISCOVERY AS SMART MATERIAL

Kim et al. (2006b) reported the discovery of cellulose as a smart material that can be used for biomimetic sensor/actuator devices and micro-electromechanical systems. This smart cellulose was termed as EAPap. Considering the cellulose structure and processing of cellulose-based EAPap, its actuation mechanism is combination of ion migration and dipolar orientation. **Figure 1** depicts the concept of EAPap bending actuator. Cellulose EAPap is a film of regenerated cellulose and morphologically, regenerated cellulose consists of ordered and disordered regions. The ordered domains are composed of crystalline and amorphous sections, and water



molecules attached to hydroxyl groups can be found in large disordered regions. During the fabrication process, sodium ions were injected in the fiber. When electric field was applied, these ions migrate to anode, selective ionic, and water transport across the polymer, which direct to bending of actuator. Consequently, the large ordered region is responsible for the dipolar orientation, leading to permanent polarization, which results into piezoelectric behavior.

### ELECTROMECHANICAL BEHAVIOR AND PROPERTIES

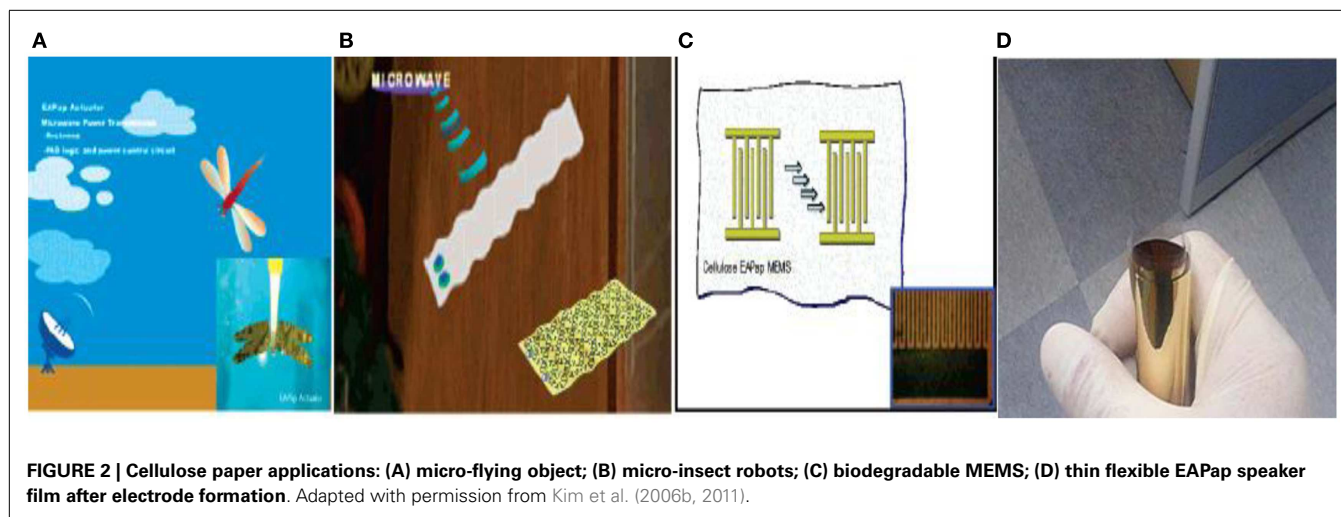
The evaluation of electromechanical coupling is very essential to utilize EAPap for broader applications. To compute piezoelectricity of EAPap, piezoelectric charge constants of EAPap were investigated using quasi-static direct piezoelectricity. Induced charge on the surface of electrode was measured for different specimens when the mechanical stretching was applied to the EAPap. Piezoelectric charge constant of 45° sample has the largest value, which shows the same pattern observed in the electromechanical behavior test. In-plane piezoelectric charge constants of EAPap are very similar to commercial piezoelectric polymer films PVDF (Kim et al., 2008b). XRD patterns showed that samples fabricated by electrospinning and stretching method enhanced crystallinity of cellulose film, which improves piezoelectricity of EAPap (Yun et al., 2008b). Wet drawing method with different stretching conditions during the fabrication process of cellulose film reveals that EAPap is sensitive to drawing ratio and material orientation (Yang et al., 2009). Another effective way to enhance the piezoelectricity of EAPap is using electrically aligned cellulose film. The application of external electric fields induces formation of nanofibers in the cellulose, resulting in an increase in the crystallinity index (CI) values (Yun et al., 2009). Theoretical model for electromechanical actuation behavior of CBC-EAPap shows good correlation with the experiment results, which can be helpful to predict and understand the electromechanical behavior of EAPap devices such as artificial muscle, biomimetic pump, and other potential applications (Jang et al., 2009).

Mechanical properties of EAPap are heavily dependent on the humidity and temperature conditions. The elastic strength and stiffness gradually decreases with the increase of both humidity and temperature. The measured value of Young's modulus of EAPap ranges from 4 to 9 GPa, which is relatively on the higher side as compared to other EAP materials. Layered and oriented cellulose-EAPap is a complex anisotropic material in term of elastic and plastic deformation, creep. It is also dependent on temperature and humidity (Kim et al., 2008a). EAPap creeps at elevated temperature, the elastic behavior varies inversely with temperature and with the bias angle tested. However, the overall viscous behavior of the EAPap tested was relatively complex (Kim et al., 2008c; Lee et al., 2009a). Hydrothermal behavior of EAPap showed that the creep resistance varied by changing the orientation of EAPap samples (Kim et al., 2009).

### APPLICATIONS AND FUTURE FORECAST

Cellulose-EAPap actuation mechanism, which is combination of piezoelectric effect and ion migration effect associated with constituent of cellulose materials and their material properties, demonstrates its potential for many applications. Due to exciting actuation phenomenon, EAPap can be used for sensors and actuating devices. Kim et al. (2006a) studied a micro-patterning technique regarded as micro transfer printing, a gold (Au) electrode pattern for a surface acoustic wave (SAW) MEMS device on cellulose EAPap. Cellulose-based EAPap shows potential as biosensor since it is eco-friendly, sustainable, biocompatible, good mechanical strength and stiffness, and hydrophilic nature. However, some challenges needed to be addressed in terms of consistency, material robustness, and niche application progress (Kim et al., 2008d).

Electro-active paper performance and material characterization shows promise for light weight smart and hybrid actuators, which make use of converse piezoelectric effect (Yun et al., 2008a; Mahadeva et al., 2009b). More recently, a remotely powered and controlled actuator made by using a dipole rectenna array along with modulated microwaves was investigated. Modulated



microwave power transmission is very valuable technology for remotely driven actuators, biomimetic robots, and remote sensing units (Yang et al., 2013). EAPap also shows fair quality results when tested as vibration sensor and for beam vibration control (Lee et al., 2009b; Kim et al., 2010a,b). Acoustic characterization of cellulose EAPap for flexible film type speaker suggests that piezoelectricity of EAPap can be used in acoustic applications (Kim et al., 2011). The reasonable piezoelectric coupling coefficient and ability to withstand high vibration amplitude due to flexible nature can help to develop EAPap energy harvesting wireless sensor nodes. Cellulose film coated with different size metal electrode was excited under different acceleration input to examine its possibility for flexible energy harvesting transducer (Abas et al., 2014a,b). The tests on EAPap demonstrate the feasibility of cellulose EAPap for haptic applications (Yun et al., 2010). Cellulose–silica/gold one-dimensional nano-composite was prepared by sol-gel cross linking process, and was investigated for electronic applications due its optical, electrical, and mechanical properties (Kim et al., 2014). **Figure 2** shows cellulose paper applications.

## CONCLUSION

A review of cellulose EAPap is presented from its discovery to real world technology applications. Cellulose paper has been investigated as a smart material and can be used as biomimetic actuators and sensors. Its smart characteristics and actuation mechanism makes it suitable for many applications including micro robotics, MEMS devices, flexible speaker, micro pumps, remotely controlled actuators, and a possible application as self powered wireless sensor node. There is a lot of potential in cellulose EAPap for a smart material. Nevertheless, some issues exist in terms of consistency, material robustness, environmental effects, and future applications development.

## ACKNOWLEDGMENTS

This research was supported by the Basic Science Research Program, through the National Research Foundation of Korea (NRF), funded by the Ministry of Education, Science and Technology (NRF-2011-0021720).

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**Conflict of Interest Statement:** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Received: 18 July 2014; accepted: 15 September 2014; published online: 30 September 2014.

Citation: Abas Z, Kim HS, Kim J and Kim J-H (2014) Cellulose electro-active paper: from discovery to technology applications. *Front. Mater.* 1:17. doi: 10.3389/fmats.2014.00017

This article was submitted to *Smart Materials*, a section of the journal *Frontiers in Materials*.

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