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# Hygrothermal behavior of electro-active paper actuator†

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## Abstract

Mechanical properties of the electro-active paper (EAPap) actuator were tested to investigate its hygrothermal behavior. Tensile creep behavior was studied with constant load at 30-70% relative humidity ranges and 25-40℃ temperature. Creep deformation showed typical trend of abrupt strain increase in a short period followed by steady increase of strain, which resulted from the breakdown of cellulose microfibrils. Dependence on the material orientation of EAPap was observed in the creep tests. As changing the orientation of EAPap samples, the creep resistances were varied. Creep strains and creep strain rates were increased as increasing the relative humidity level at 25℃. However, at the elevated temperature of 40℃, the creep strain rate at secondary creep was not significantly raised under increased relative humidity level from 30% to 50%. The hygrothermal effect by increasing the relative humidity level and temperature on the creep rate was reduced due to the saturated moisture at a higher temperature even with lower humidity level. The activation energy levels for creep were around 607-658 kJ/mol for 30% relative humidity level and 623-671 kJ/mol for 50% relative humidity level depending on the material orientation. Understanding of hygrothermal effect in conjunction with the humidity and temperature provides useful information for the potential nano-bio applications of the EAPap actuator.

*Keywords*: Electro-active paper actuator; Hygrothermal behavior; Material orientation; Creep; Temperature; Humidity

## 1. Introduction

The potential of cellulose-based electro-active paper (EAPap) as a smart material was first discovered by Kim [1, 2]. Since then, this material has been investigated as one of the promising electro-active polymer materials due to its intrinsic physical and chemical properties such as biodegradability, light weight, large displacement response, and low power consumption. Cellulose is a cheap and abundant resource for diverse usages, which can be obtained from wood, cotton, algae, etc. [3-5]. Cellulose-based EAPap is a natural composite material made up of multi-layers of cellulose fibers.

Thin metallic electrode deposition on both sides of cellulose paper makes EAPap actuate under an electric field. The actuation principle of this material has been known to be a combination of ion-migration and piezoelectricity [6]. Previous study reported a large bending displacement when an electric field was applied across the thickness direction of EAPap [7]. A maximum tip displacement of 4.3 mm out of 40 mm long was achieved with the 0.25 kV/mm electric field at the resonant frequency of the EAPap actuator [8]. The exciting electric field of EAPap is quite low and 1.1 mN of the maximum force was observed from the EAPap actuator [9, 10]. Such an EAPap also showed strong piezoelectricity and provided great potential for many applications including actuators and sensors [11, 12]. Since the power requirement of the EAPap

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actuator is less than 15 mW/cm$^2$, which is below the safety limit of microwave-driven power, it can be remotely driven by microwave power [13-15]. This idea is useful for applications that require ultra-lightweight multifunctional capabilities such as smart skin, micro insect robots, flapping wings for insect-like flying objects, smart wall paper, MEMS, and so on.

However, due to the intrinsic limitations of this material, influences of environmental factors on the performance of the material need to be investigated. Static mechanical tests were performed in the previous study [16]. However, its tensile creep behavior and hygrothermal effect for the creep test have not been studied. Cellulose-based materials such as paper and paperboard exhibit creep behavior with constant load and also show accelerated creep with varying humidity [17-21]. Cellulose possesses a semi-crystalline structure composed of crystalline and amorphous regions. Due to many hydroxyl groups in cellulose, water molecules can be easily absorbed in the amorphous region [22]. Humidity conditions affect the behavior of most engineering materials including metals, ceramics, composites, and polymers [23-25]. In particular, cellulose materials are easily susceptible to the water and absorbed water changes its structural behavior. The changed structure by absorbed water is strongly related to its material properties [26, 27]. To be used as an actuator, EAPap should withstand loads for a long time regardless of high humidity and temperature conditions. Thus, this paper studies creep behavior of cellulose EAPap in conjunction with hygrothermal effect for the creep test. The hygrothermal effect includes coupled humidity, temperature and load history. Creep tests under different humidity levels with fixed temperature and different temperatures with fixed humidity are expected to provide the hygrothermal effect on the performance of EAPap.

## 2. Experiments

### 2.1 Sample preparation and test setup

Cellulose EAPap is made by dissolving cellulose fiber into a solution and casting it. The cast film is washed with water, resulting in regenerated cellulose. Cellulose films made by Weifang Co., China were used. The size of sample was 50 mm in length, 12 mm in width and 33 μm in thickness. Cellulose films possess material orientation during the stretching and drying process. Since the effect of material orientation is important to characterize the performance of the EAPap actuator, creep samples were prepared parallel to the mechanical direction (0 degree), 45 degrees, 90 degrees from the mechanical direction for present study as shown in Fig. 1.

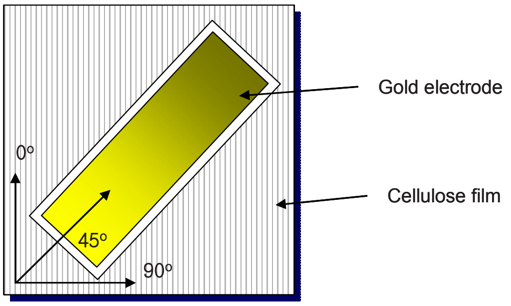


Fig. 1. Orientation of cellulose film and schematic of EAPap.

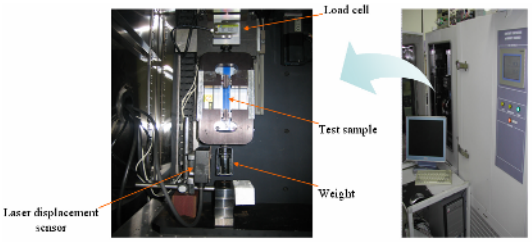
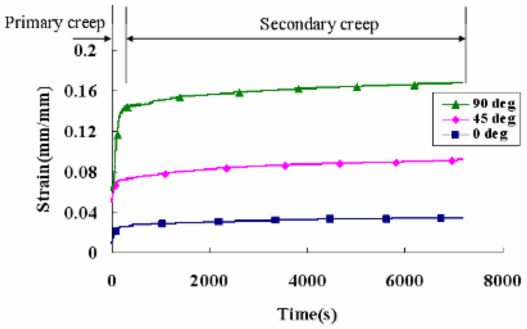


Fig. 2. Creep test setup and constant temperature and humidity chamber.

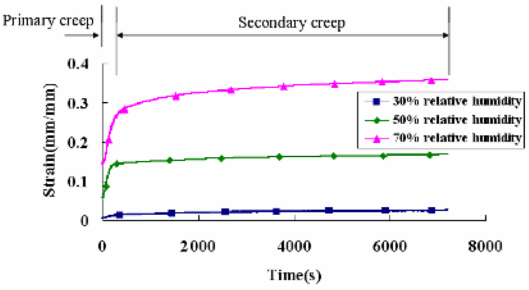
The experimental setup for the creep test was comprised of a dead-weight creep tester and an environmental chamber as shown in Fig. 2. The environmental chamber was manufactured by Labcamp Co. Ltd. Korea (Model No.: CTHC-500P). The creep tester, which was modified from a tensile test machine, was located in an environmental chamber that can control the temperature and humidity. To apply constant load to the sample, a constant weight was attached to the end of samples in the environmental chamber. This setup provided a unique condition to verify the ambient factors of humidity and temperature. Data acquisition was coded by Labview commercial software.

### 2.2 Experimental procedure

In the creep tests, the upper end of the sample was clamped by the fixture of the tensile machine and the constant weight was attached to the lower end as shown in Fig. 2. In this test, the yield strength determined in the previous tensile test was used as a refer



(a) Material orientation (at 50% RH,25℃)



(b) Relative humidity (at 25℃)

Fig. 3. Time dependency of creep in cellulose film.

ence, and 70% load of the pre-determined yield strength was applied to the sample [16]. The creep displacement was measured by a laser displacement sensor (KEYENCE Co. Japan, Model No.: LK-G85). To ensure that creep strain was measured in the saturated conditions, each sample was kept in the environmental chamber at the designated temperature and relative humidity levels during one hour. To investigate the effects of environmental conditions, three different relative humidity levels (RH) and two different temperature conditions were studied: (i) 30, 50 and 70%RH at 25℃, (ii) 30, 50%RH at 40℃. The test scheme was limited due to the sensitivity of the samples to the environmental conditions and limitations of the test apparatus.

## 3. Results and discussions

Creep tests were performed to characterize the hygrothermal effect on the behavior of EAPap. Creep in hydrophilic materials mainly occurs due to the hygrothermal effect of coupled humidity and temperature. The creep strain of hydrophilic materials is commonly increased under high humidity condition. Since EAPap is a cellulose-based hydrophilic material like paper, the material behavior of EAPap is expected to depend on the humidity and temperature variations.

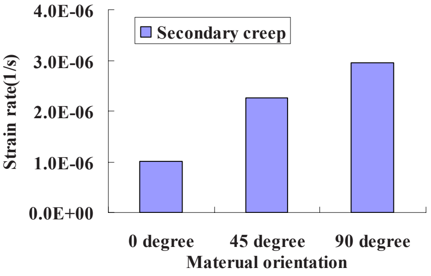


Fig. 4. Creep rate vs. material orientation (at 50% RH,25℃).

Time dependencies of creep in EAPap samples with different material orientation and relative humidity levels are presented in Fig. 3. It is observed that the creep of EAPap consists of two parts, classified as primary and secondary creeps. In primary creep, large displacement was generated just after static deformation. On the other hand, steady increment of displacement occurred in the secondary creep as shown in Fig. 3. Practically, secondary creep is of great interest because it can be a dominant part of whole deformation period. In the EAPap, secondary creep dominated the deformation process in the creep test period. Creep strain of the EAPap was increased by varying the material orientation from 0 to 90 degree at 25℃. Perpendicular to the mechanical direction, termed as 90 degrees, the sample showed the least creep resistance as expected. This indicates that the creep of EAPap is very sensitive to the orientation as other mechanical properties, for example, stiffness of the material. For instance, the 90º case showed about 16% creep strain compared with 4% creep strain of 0º case at 25℃.

It is generally known that creep of polymer strongly depends on the stress, time, and temperature. In EAPap, in addition to the factors aforementioned, humidity plays a key role in the creep deformation process. Thus, the effect of increasing relative humidity level on the creep was studied by using the 90 degree sample at 25℃. Creep was more evident with increasing humidity level (See Fig. 3). This trend is also clearly observed in creep rate. Best fit straight lines along the primary and secondary portions of the creep strain-time curve determine the creep rates at primary and secondary creeps, respectively.

Fig. 4 presents the relation between creep rate and material orientation. As seen, the creep rate increased

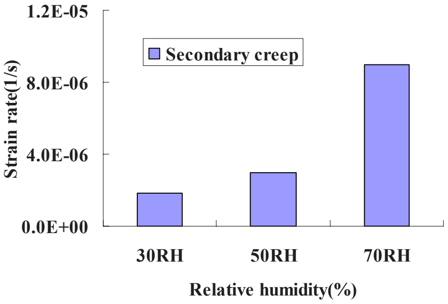


Fig. 5. Creep rate vs. relative humidity at 25℃.

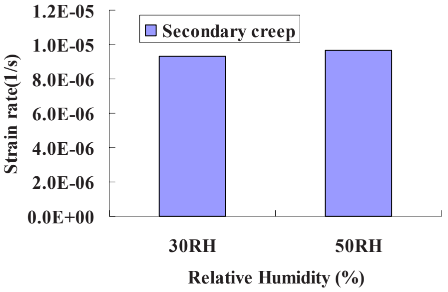


Fig. 6. Creep rate vs. relative humidity at 40℃.

with varying material orientation from 0 to 90 degree at 25℃. The 90 degree sample showed a substantial increase of creep rate (about 300%) compared with the creep rate of mechanical direction (0 degree). The least creep resistive 90 degree sample was selected to see the hygrothermal effect of temperature and relative humidity levels.

Hygrothermal effects by temperature and relative humidity levels on the creep rate are delineated in Fig. 5 and 6. Increasing the relative humidity level with varying temperature showed how the creep in EAPap was affected by two environmental factors when they were combined. In Fig. 5, relative humidity level was increased from 30% to 70% at 25℃. The presence of moisture increased the secondary creep rate by about 63% for 50%RH and 392% for 70%RH compared with that of 30%RH. Creep rate increase with higher humidity might be due to the fact that many hydroxyl groups in amorphous cellulose regions strongly absorbed water molecules (free water). As relative humidity increases, excess free water molecules might penetrate into amorphous cellulose regions, resulting in accelerated creep deformation.

At room temperature, 25℃, there were big differences of the creep rate among 30%, 50%, and 70%RH conditions (see Fig. 5). However, at elevated temperature to 40℃, the creep rate did not increase much as the relative humidity changed from 30 to 50% (see Fig. 6). The moisture level in EAPap can be easily saturated even in lower humidity level at the higher temperature range such that the creep strain rate cannot be significantly raised with increasing the relative humidity level. In other words, the hygrothermal effect by temperature and relative humidity level on the creep rate of EAPap was not strong at high temperature ranges. This information can be used in designing application devices with EAPap by referring permissible creep strain rates according to the ambient conditions.

Table 1. Activation energies for creep deformation of EAPap.

| Sample | Temperature (K) | Relative humidity (%) | Activation energy, Q (kJ/mol) |
|---|---|---|---|
| 0 degree | 298 – 313 | 30 | 658 |
| | | 50 | 671 |
| 45 degree | 298 – 313 | 30 | 638 |
| | | 50 | 649 |
| 90 degree | 298 – 313 | 30 | 607 |
| | | 50 | 623 |

Activation energies for the creep of EAPap were around 607-658 kJ/mol for 30%RH and 623-671 kJ/mol for 50%RH depending on the material orientation as shown in Table 1. Activation energies for creep were obtained from the slope of semi-logarithmic Arrhenius plots of creep rate versus inverse Kelvin temperature. Fig. 7 shows the material orientation dependence on the creep rate of EAPap at 298-313K with 30%RH. The orientation dependence on the creep rate of EAPap at 298-313K with 50%RH is plotted in Fig. 8. Keeping very slow creep rate is critical to maintain the dimensional consistency of sensor or actuator devices. In this sense, even though EAPap has a dependency on material orientations and environmental conditions, creep rates shown in Fig. 7 and 8 indicate promising aspects of the material for possible device applications.

## 4. Conclusions

The effects of material orientation and hygrothermal behavior of electro-active paper (EAPap) were investigated. It was reported that elastic stiffness and strength of EAPap was strongly dependent on the material orientation, humidity and temperature. Similar behavior was also observed in creep behavior.

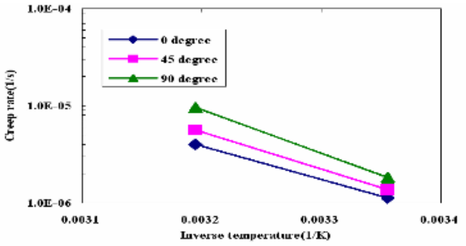


Fig. 7. Creep rate vs. inverse temperature at 30% RH.

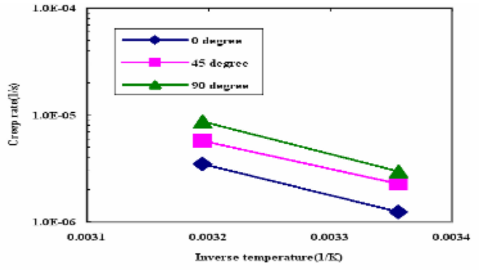


Fig. 8. Creep rate vs. inverse temperature at 50% RH.

Creep resistance was decreased with increasing the material orientation from 0 degree to 90 degree, relative humidity level from 30% to 70%, and temperature from 25℃ to 40℃.

Hygrothermal behavior of EAPap was studied under coupled humidity and temperature effect. At 25℃, the creep strain and creep rates were increased as hygrothermal effect accelerated the creep deformation. However, with elevated temperature, the hygrothermal effect was not severe as expected. The creep rate at 50%RH was not significantly increased from that of 30%RH at the elevated temperature 40℃. This was because moisture in the EAPap was easily saturated at the elevated temperature, and increased humidity did not have much influence on the creep deformation. It is suggested that the hygrothermal effect on the performance of EAPap can be strong at room temperature range but weak at the elevated temperature range.

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