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**Article type: Communication****A Miniaturized Flexible Antenna Printed on High Dielectric Constant Nanopaper Composite***Tetsuji Inui, Hirotaka Koga,\* Masaya Nogi,\* Natsuki Komoda, Katsuaki Suganuma*

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Portable electronic devices such as smartphones and tablets have become widespread in recent years, and this has promoted demand for reducing their size and weight. Making such portable devices flexible is also an aim of next-generation wearable electronics.<sup>[1]</sup> Antenna components allow the transmission and reception of radio waves as information signals, and are essential in wireless communication. The trend towards flexible and wearable electronics has promoted the development of miniaturized and flexible antenna materials. Flexible antennas have been prepared by patterning conductive metal lines on plastic substrates.<sup>[2-4]</sup> However, downsizing antenna materials remains a challenge.

Paper serves various purposes in daily life, including in writing, printing, wiping, wrapping, and packaging applications. Paper materials are sustainable, inexpensive, lightweight, and flexible. Paper has recently been applied in electronic applications, and electronic devices have been prepared on paper substrates,<sup>[5]</sup> such as inorganic transistors,<sup>[6]</sup> complementary metal oxide semiconductor,<sup>[7,8]</sup> and memory.<sup>[9]</sup> We previously developed a paper substrate based on wood-derived cellulose nanofibers (denoted cellulose nanopaper).<sup>[10,11]</sup> It has applications in flexible electronic components<sup>[12]</sup> such as conductive lines,<sup>[13,14]</sup> transparent conductive films,<sup>[15-17]</sup> inorganic transistors,<sup>[18]</sup> organic transistors,<sup>[19]</sup> nonvolatile memory,<sup>[20]</sup> and antennas.<sup>[21]</sup> Traditional paper prepared from micrometer-sized cellulose pulp fibers has

a high porosity and surface roughness. Metal nanoparticle lines on traditional paper therefore exhibit poor electrical conductivity, which hinders their use as antennas. The cellulose nanopaper substrate has a densely packed nanostructure and smooth surface, allowing highly conductive and sensitive antenna lines at high frequency ( $>1$  GHz).<sup>[21]</sup> The nanopaper antennas even maintain their sensitivity after folding. Despite this, downsizing these nanopaper antennas for use in wearable wireless communications remains a challenge.

Shortening the antenna length is one approach for preparing miniature antennas. The length of the antenna element typically needs to match one quarter of the radio-wave wavelength and set as a function of the objective frequency band, to ensure resonance between the radio-wave and element for efficient transmission and reception. The radio-wave frequency is affected by the dielectric environment; *i.e.* the wavelength decreases upon transfer from air into high dielectric constant ( $k$ ) materials.<sup>[22]</sup> Thus, employing high  $k$  substrates can shorten the length of the antenna element, without affecting the radio-wave frequency.

Herein, we fabricated a miniaturized flexible antenna on a high  $k$  nanopaper. Dispersing a small amount of silver nanowires inside the cellulose nanopaper, while keeping them nonparcolative, dramatically increased the  $k$  value of the nanopaper, and retained its flexibility. The resulting silver nanowire/nanopaper was used as an antenna substrate. The silver antenna printed on this support was downsized by about a half, compared with the antenna printed on the original nanopaper or conventional plastic film. The antenna retained its sensitivity to specific radio frequencies.

High  $k$  substrates are essential for the miniaturization of antennas. Traditional paper substrates made from untreated cellulose pulp fiber with micrometer-scale widths exhibited lower  $k$  values ( $k=2.9$  at 1.1 GHz) than typical plastic films such as polyethylene terephthalate (PET,  $k=3.1$ ), polyethylene naphthalate (PEN,  $k=3.4$ ), and polyimide (PI,  $k=3.4$ ). The  $k$  value of cellulose is reportedly 6–8.<sup>[5]</sup> Traditional paper has a density of  $0.72$  g cm<sup>-3</sup>, which is lower than that of cellulose ( $1.59$  g cm<sup>-3</sup>). The low  $k$  value of traditional paper may be because of its

porous microstructure of micrometer-sized cellulose fibers (Figure 1a) and air ( $k \sim 1$ ). In this study, cellulose nanofibers were used to prepare high-density paper. To prepare the cellulose nanofibers, pulp fiber was nanofibrillated using a water-jet system equipped with a ball-collision chamber.<sup>[11]</sup> An aqueous suspension of pulp fiber was repeatedly collided with a ceramic ball under high pressure. The average width of the cellulose fiber decreased with increasing collision number (pass number), from several 10  $\mu\text{m}$  to about 15–40 nm after 10 passes (see Supplementary Information, Figure S1). To prepare the cellulose nanopaper, an aqueous suspension of cellulose nanofibers was dewatered through a commercial membrane filter, followed by hot pressing and peeling from the filter. The density and  $k$  value of the resulting paper were increased with increasing pass number of the fibers (see Supplementary Information, Figure S2). Thus, the  $k$  value increased with increasing paper density (Figure 1c). The cellulose nanopaper contained a densely packed nanostructure (Figure 1b), with a density of up to 1.3  $\text{g cm}^{-3}$  and  $k$  value of up to 5.3. Cellulose nanofibers could therefore be used to increase the papers density and  $k$  value. The nanopaper had a higher  $k$  value than typical PET, PEN, and PI substrates used in flexible electronics. However, the  $k$  value of the nanopaper was lower than that of conventional high  $k$  polymers such as polyvinylidene difluoride ( $k=8.5$ ).<sup>[23]</sup>

The  $k$  values of polymer matrices have previously been increased by incorporating inorganic nanofillers.<sup>[24-26]</sup> Various inorganic nanofillers were dispersed into the current nanopaper ( $k=5.3$ ), by mixing the nanofiller with an aqueous suspension of cellulose nanofibers. Barium titanate ( $\text{BaTiO}_3$ ) has a high  $k$  of 5000,<sup>[27]</sup> and  $\text{BaTiO}_3$  nanoparticles of diameter  $\sim 100$  nm were dispersed in the nanopaper (see Supporting Information Figure S3a). Figure 2a shows that the  $k$  value of the  $\text{BaTiO}_3$  nanoparticle/nanopaper composite gradually increased with increasing  $\text{BaTiO}_3$  content. However, the  $k$  value of the  $\text{BaTiO}_3$  nanoparticle/nanopaper composite was 8.1 at 20.9 vol.% filler content, so high  $k$  nanofillers evidently had little effect on improving the  $k$  value.

The  $k$  value of polymer matrices can also reportedly be increased by dispersing conductive metal nanofillers in the target matrix, while keeping the nanofiller nonpercolative.<sup>[28-32]</sup> Highly conductive silver nanoparticles of diameter  $\sim 100$  nm were dispersed in the nanopaper to further increase its  $k$  value (see Supporting Information Figure S3b). The  $k$  value of the silver nanoparticle/nanopaper composite was 24.15, at 13.15 vol.% filler content (Figure 2a). Thus, the conductive silver nanoparticles more effectively increased the  $k$  value than the BaTiO<sub>3</sub> nanoparticles. However, such high filler contents lead to a decrease in flexibility of the resulting polymer composites.<sup>[33]</sup> Alternative nanofillers that can significantly increase the  $k$  value at low concentration are required for use in flexible and wearable electronics.

The  $k$  value of metal-polymer composites reportedly sharply increases near the percolation threshold of the conductive metal nanofiller.<sup>[34,35]</sup> Silver nanowires with high aspect ratios (see Supporting Information Figure S3c<sup>[36]</sup>) were then dispersed in the nanopaper. Figure 2b shows that the silver nanowire/nanopaper composite had a much higher  $k$  value at low filler concentration, compared with the silver nanoparticle/nanopaper composite. The  $k$  value was 726.5 at a filler content of 2.48 vol.%. A higher filler content resulted in the formation of percolation networks, and a loss in dielectric activity. According to previous reports,<sup>[35,37,38]</sup> this dramatic increase in  $k$  was possibly due to the formation of microcapacitor networks by neighbouring the conductive silver nanowires and the insulating cellulose nanofibers in between. In addition, the entrapment of free charges at the interface of the cellulose nanofibers and the silver nanowires would also provide the improvement of the  $k$  value. The silver nanoparticles and nanowires were both well dispersed in the nanopaper (Figure 2c and d). However, nanowires with aspect ratios of  $\sim 100$  (average diameter  $\sim 100$  nm, average length  $\sim 10$   $\mu\text{m}$ ) have a lower percolation threshold than those with aspect ratios of  $\sim 1$ . Therefore, the silver nanowire/nanopaper composite achieved an extremely high  $k$  value (726.5 at 1.1 GHz) from a small amount of nanowires, which was about 140 times higher than that of the original nanopaper. This was much higher than the  $k$  values of reported high  $k$

polymer composites such as  $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$  nanoparticle/epoxy composite<sup>[39]</sup> ( $k=24$  at 2 GHz),  $\text{Fe@SiO}_2$  nanoparticle/poly(dimethyl siloxane) composite<sup>[40]</sup> ( $k=4.3$  at 1 GHz), and  $\text{Fe@Ag}$  nanoparticle/epoxy composite<sup>[41]</sup> ( $k=22.3$  at 1–3 GHz). The silver nanowire/nanopaper composite had paper-like qualities, because of its low silver nanowire content. It could be cut to shape with scissors, and folded into specific patterns (see Supporting Information Figure S4). The high- $k$  nanopaper composite demonstrated sufficient reliability enough to maintain its  $k$  value after storing in the air for at least 1 month and after thermal treatment at 60 °C for at least 1 day. For further improvement of the stability, the moisture resistance of the cellulose paper can be dramatically enhanced by using various techniques, as previously reported.<sup>[42-45]</sup> In addition, conventional sealing processes would be also effective.

The nanopaper composite was used as a substrate for miniature antennas. Dipole antenna elements were mask printed on the original nanopaper and 2.48 vol.% (14.38 wt.%) silver nanowire/nanopaper composite, by using silver nanoparticle ink (Figure 3a). Their resonance frequency and return loss were compared, as shown in Figure 3b. The resonance frequency is that which receives the radio-wave signal most effectively, and the S11 parameter of return loss indicates the loss of signal power during signal reflection in the antenna element. Figure 3b shows that the 17-mm-long dipole antenna element on the silver nanowire/nanopaper composite had the lowest return loss, at a resonance frequency of 2.60 GHz. This return loss was similar to that of a 30-mm-long dipole antenna printed on the original nanopaper. The silver nanowire/nanopaper substrate decreased the required antenna element length by about a half, compared with the original nanopaper and conventional PI and PEN substrates (see also Supporting Information Figure S5). The target radio-wave frequency was not affected. The wavelength of the radio waves decreased upon entering the high  $k$  substrate.<sup>[46]</sup> Considering the resonance frequency of 2.60 GHz, the wavelength of the propagation signal was about 115 mm. On the other hand, the 17-mm-long dipole antenna received the radio wave with wavelength of 68 mm. Thus, the high  $k$  silver nanowire/nanopaper composite would shorten

the wavelength of the radio waves from 115 mm to 68 mm (see also Supporting Information Figure S6), leading to the reduction of the required antenna element length. Shortening the element reduced the size and weight of the nanopaper antenna and substrate by about a half (Figure 3a). There was no significant difference in return loss of these antennas, indicating that antenna sensitivity was maintained by the silver nanowire/nanopaper substrate. This was because the dielectric loss of the nanopaper was not significantly affected by its mixing with silver nanowires (see Supporting Information Figure S7). Figures 3c and d show the flexibility of this miniaturized nanopaper antenna. In our previous studies, the nanopaper-based devices such as transistors<sup>[19]</sup> and nonvolatile memory<sup>[20]</sup> offered sufficient flexibility enough to maintain their performance during and after bending. The miniaturized nanopaper antenna also retained its sensitivity after the 1000-cycles bending test, indicating the high flexibility performance.

To conclude, the high  $k$  silver nanowire/nanopaper substrate resulted in a downsizing of the nanopaper antenna by about a half, while retaining its sensitivity and flexibility. The high  $k$  nanopaper composite can be applied in various flexible electronics applications. The miniaturized flexible antenna on the high  $k$  nanopaper demonstrated the lowest return loss around 2.60 GHz, indicating the applicability for Wi-Fi communication (IEEE 802.11n, frequency band: 2.45 GHz). Thus, this small flexible nanopaper antenna has potential as a component in wearable wireless communication devices such as smartphones and tablets.

### *Experimental*

*Materials:* Cellulose nanofibers were prepared as previously reported.<sup>[11]</sup> Silver nanowires of diameter  $\sim 100$  nm and length  $\sim 10$   $\mu\text{m}$  were synthesized by the reduction of silver nitrate (Wako Pure Chemical Industries, Ltd., Japan) in the presence of poly(vinylpyrrolidone) (Wako Pure Chemical Industries, Ltd., Japan) and iron chloride ( $\text{FeCl}_3$ ) (Wako Pure Chemical Industries, Ltd., Japan) in ethylene glycol (Wako Pure Chemical Industries, Ltd., Japan), as

described previously.<sup>[36,47]</sup> Silver nanoparticles of diameter ~100 nm were also synthesized without the addition of FeCl<sub>3</sub>. BaTiO<sub>3</sub> nanoparticles of diameter ~100 nm (Wako Pure Chemical Industries, Ltd., Japan) were purchased and used for comparison.

*Preparation of cellulose nanofiber paper and silver-containing cellulose nanofiber paper:*

Cellulose nanofiber paper, denoted cellulose nanopaper in this study, was prepared as follows. An aqueous suspension of cellulose nanofibers (0.7 wt.%, 35.7 mL) was degassed using a vacuum mixer (ARV-310; Thinky Co., Ltd., Japan) at 2000 rpm for 9 min. The resulting suspension was dewatered by suction filtration through a mixed cellulose ester membrane filter (A010A090C; Advantech Co., Ltd., Japan). The wet paper on the membrane was sandwiched between a hydrophobic glass plate and paper towel, and dried by hot pressing at 110 °C for 20 min (1.1 MPa). The paper was then peeled away from the membrane. Traditional cellulose paper was also prepared from pulp fibers that had not been nanofibrillated. For preparing the silver-containing cellulose nanopaper, an aqueous dispersion of silver nanowires or silver nanoparticles was mixed with an aqueous suspension of cellulose nanofibers. The resulting suspension was fabricated into the paper, in a similar manner to the cellulose nanopaper. BaTiO<sub>3</sub> nanoparticle-containing nanopaper was prepared similarly. The vol.% values of the fillers in the nanopaper composites were determined by the addition amount of the fillers and cellulose nanofibers, because almost all the fillers were retained in the nanopaper.

*Printing of dipole antennas on various substrates:* Four types of substrates with thicknesses of ~50 μm were used: cellulose nanopaper, silver nanowire/nanopaper (silver nanowire content: 2.48 vol.%), PEN film (Teonex Q51; Teijin DuPont Films Japan Limited, Japan), and PI film (Kapton 300H; Teijin DuPont Films Japan Limited, Japan). V-shaped dipole antenna patterns of length 30 or 17 mm and width 5 mm were mask printed with silver nanoparticle paste



(GOP-010; InkTec Co., Ltd., Korea) on these substrates. The printed substrates were then treated at 100 °C for 60 min in air.

*Analyses:* Samples were observed by field-emission scanning electron microscopy (JSM-6700F; JEOL Ltd., Japan), after coating with gold by ion sputtering (E1045; Hitachi High-Technologies Co., Ltd., Japan). The dielectric properties were measured using an impedance analyzer (E5071C; Agilent Technologies, Inc., USA) with a split post dielectric resonator (QWED, Poland) at 1.1 GHz. Prior to measurement, samples were stored in a humidity chamber (SH-641; ESPEC Co., Ltd., Japan) at 23 °C and 50% relative humidity overnight. The return losses of the V-shaped dipole antennas were evaluated inside an electromagnetic shielding bag at 0.5–3.5 GHz, using a network analyzer (E5061A; Agilent Technologies, Inc., USA). For flexibility testing, the silver antenna on the silver nanowire-containing nanopaper was subjected to the 1000-cycles bending test (pulling and compressing rates: 100 mm min<sup>-1</sup>, span length: 12.5 mm), using a Shimadzu EZ-SX test instrument.

### Supporting Information

Supporting Information is available online from the Wiley Online Library or from the author.

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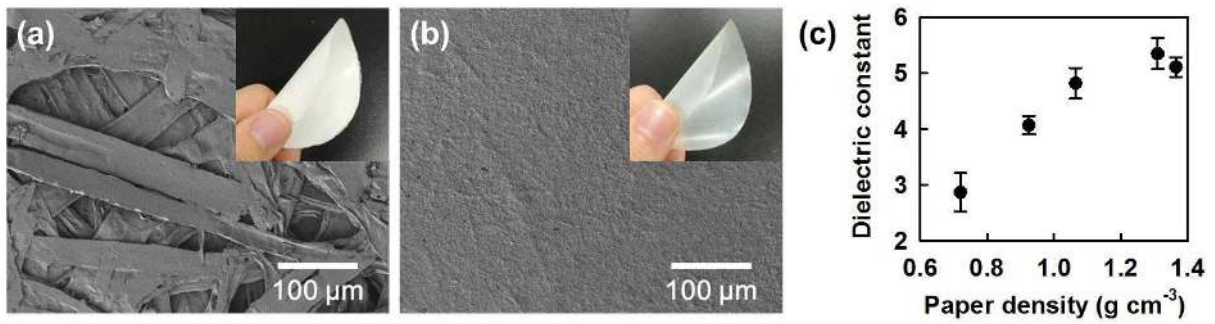
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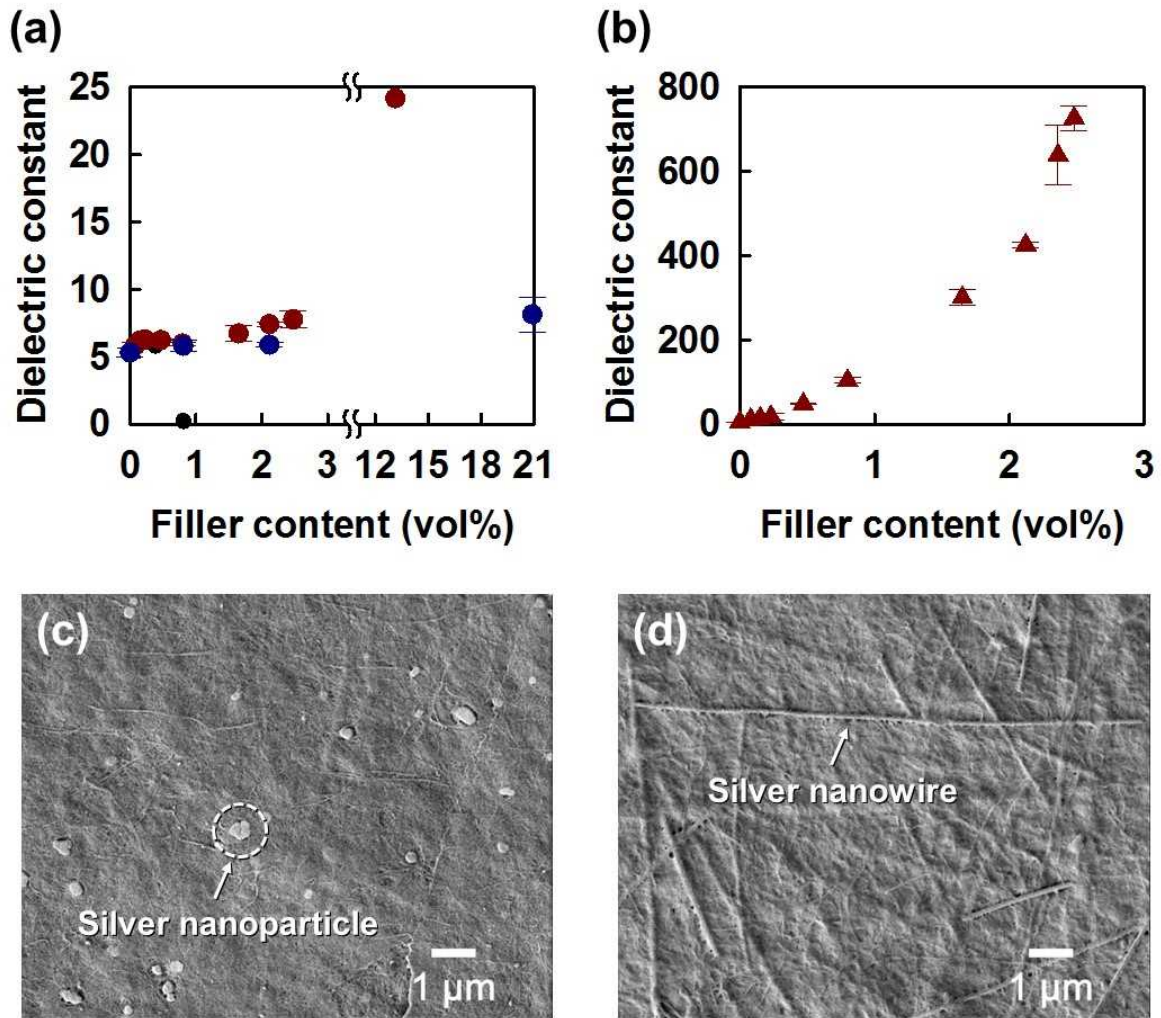
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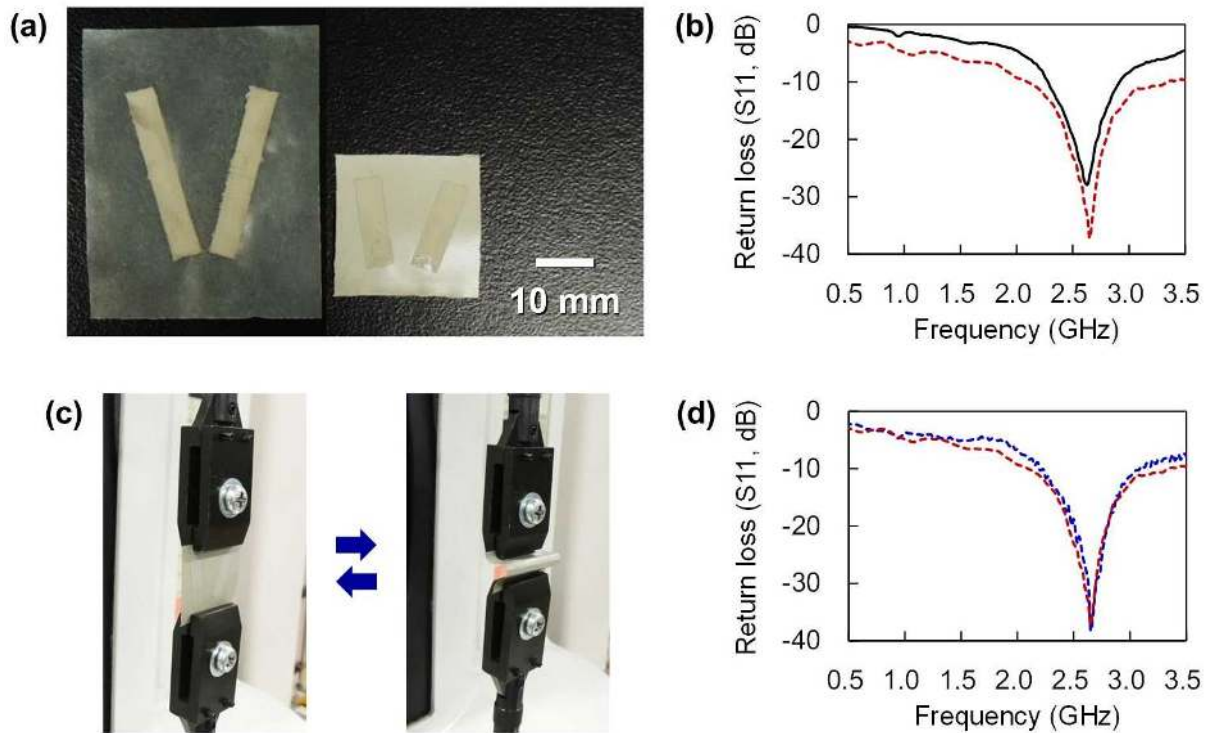
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**Figure 1.** Optical (inset) and field-emission scanning electron microscopy images of papers prepared from a) cellulose pulp fibers (0 passes) and b) cellulose nanofibers (10 passes). c) Dielectric constant of the nanopaper at 1.1 GHz, as a function of paper density.



**Figure 2.** a) Dielectric constant of the nanopaper composite at 1.1 GHz, as a function of BaTiO<sub>3</sub> (blue) and silver (red) nanoparticle filler content. b) Dielectric constant of the nanopaper composite at 1.1 GHz, as a function of silver nanowire filler content. Field-emission scanning electron microscopy images of the c) silver nanoparticle/nanopaper composite and d) silver nanowire/nanopaper composite at 2.48 vol.% filler content.



**Figure 3.** a) Optical images of the 30-mm-long dipole antenna on the 50- $\mu\text{m}$ -thick nanopaper (left), 17-mm-long dipole antenna on the 50- $\mu\text{m}$ -thick 2.48 vol.% (14.38 wt.%) silver nanowire/nanopaper composite (right). b) Return losses of the 30-mm-long dipole antenna on the nanopaper (black solid line), and 17-mm-long antenna on the 2.48 vol.% (14.38 wt.%) silver nanowire/nanopaper composite (red dotted line). The sizes of the nanopaper and silver nanowire/nanopaper substrates were 50.0 $\times$ 37.5 and 25.0 $\times$ 25.0 mm, respectively. c) Optical images during bending of the miniaturized nanopaper antenna, and d) its return loss before (red dotted line) and after (blue dotted line) the 1000-cycles bending test.