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Design fabrication and characterisation of polyaniline and multiwall carbon nanotubes composites-based patch antenna

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Abstract: This study presents the conception, simulation, realisation and characterisation of a patch antenna made of a composite based on polyaniline (PANI) and multiwall carbon nanotubes (MWCNTs). The antenna is designed to operate at the frequency of 4.5 GHz; the dielectric substrate used is Rogers RT/Duroid® 5870 which has a dielectric permittivity of 2.33 and a loss tangent of 0.0012. The conductive polymer (PANI/MWCNTs) fabricated has a conductivity of 4500 S/m. The performance of the proposed antenna is investigated as a function of the thickness of the conductive polymer. The experimental results show an antenna gain of 5.18 dB for a PANI/MWCNTs thickness of 110 µm. The results obtained suggest that this kind of conductive polymers are good candidates for future applications based on organic electronics.

1 Introduction

Printed radiating structures have been studied by many authors in the 1950s [1–3]. In the 1970s the first realisations are registered [4, 5] and the activity dedicated to modelling of such structures begins to become notable. Today for many applications concerning wireless communication and wireless local area network (WLAN) or textile structures [6, 7], the integration of the antenna is necessary. However, for these kinds of applications, there are many constraints especially regarding cost, manufacture method, weight and withstand to weather (metrological) phenomena and corrosion.

The realisation of antennas on flexible substrates that make use of conductive polymers has attracted a strong interest in recent years [6, 8–12]. In particular, studies have shown the advantage of using some composite materials to replace conventional metals [7, 13–15] that are widely employed to realise radiating structures. For example, in [7], silver ink has been used to realise a high conductivity antenna. Besides these traditional composite materials, we find conductive polymers like polyaniline (PANI) [13] and polypyrrole [16–19] which serve for electronic radiation as well as corrosion protective coating [20]. However, the low cost of aniline monomer compared with pyrrole promotes the use of PANI [21]. Nevertheless, a drawback of PANI is its relatively low conductivity. So, a solution proposed to overcome this limitation is the addition of carbon nanotubes in the polymer matrix [22].

Today, carbon nanotubes discovered by Iijima in 1991 [23] can be found in many research fields. In particular, it has been shown that there is an interest for their application to antennas development [24–26]. There are two main types of carbon nanotubes [27] which exist in stable states, single-walled carbon nanotubes (SWCNTs) and multiwall carbon nanotubes (MWCNTs). The carbon nanotubes are a very efficient source of electrons [28], because they are made of one or several coaxial tubes composed of carbon atoms in a sp^2 bonded and hexagonal lattice [29]. So the idea is to make use of this material to increase the conductivity of polymers. The conductivity of MWCNTs is higher than one of the SWCNTs.

It has been demonstrated that improvement of interfacial interaction can be achieved by the surface functionalisation of nanotubes [30]. Actually, the polar groups on the nanotubes surface increase the conductivity of PANI/MWCNTs and consequently offer better response. For fabrication of nanocomposite-based antennas, the interactions between the surface functional groups of MWCNTs and the functional groups of the polymer chains (PANI) can also promote a good distribution of the nanotubes in the composites [31, 32]. Hence, the combination of conductive polymers and MWCNTs is an attractive way to fabricate new antennas to be used in applications where the flexibility property is needed.

The main contribution of this paper is the realisation of an antenna from an original material that is thought to have a great potential. To the best of the author's knowledge, the composite investigated in this study (PANI/MWCNTs) has not been used yet in the field of antennas.

We present in the paper the conception, simulation, realisation and characterisation of patch antennas based on a composite made of PANI doped by MWCNTs (PANI/MWCNTs). The antenna is designed to operate in C-band (around 4.5 GHz). In particular, the effects of the patch thickness on the antenna performance are investigated. The patch antenna is characterised in terms of return losses, radiation patterns and gain to demonstrate the performance of the structure proposed. In Section 2, the preparation of the PANI/MWCNTs composite material is provided. The design of the antenna is presented in Section 3. In Section 4, is given the simulation and the measurement results of the antennas proposed.

2 Preparation of PANI/MWCNTs

This section presents the fabrication steps of the conductive polymer based on PANI and MWCNTs. The general structure of the non-conducting form of PANI emeraldine base (EB) is shown in Fig. 1. In this formula, y is the oxidation degree and for $y = 0.5$, PANI is called EB, a stable form of the non-conducting form of PANI.

To obtain high conductivities, PANI-EB was fully protonated with camphor sulfonic acid (CSA) as described in literature [33]. Dichloroacetic acid (DCAA) was chosen as solvent and as secondary dopant at a concentration of 2.85% w/w. This choice reduces the number of π conjugation defects in the polymer

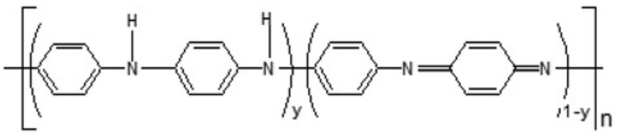

Fig. 1 *Structure of the non-conducting form of PANI: emeraldine base; y = 0.5*

backbone, enhances the cristallinity and increases the conductivity and charge carrier mobility [34]. In this work, by processing PANI-EB with CSA as dopant and DCAA as solvent and co-dopant a high conductivity can be achieved. To increase the mechanical properties of the material for the fabrication of the antenna, the PANI salt was blended with polyurethane (PU) (Bayer, Desmopan 6065A). This polymer is also soluble in DCAA, and therefore, it was possible to blend the two polymers with a precise ratio. In a previous work, it has been shown that this blend had a very low percolation threshold and can reach high conductivity [35]. In this work, we mixed the conducting PANI with PU in the mass fraction of 95% PANI and 5% PU. First PANI-EB and CSA powder were mixed in a mortar in appropriate quantity to get the full protonation of PANI. This powder is added slowly in DCAA under vigorous stirring. After 3 days the initial blue solution turns to green indicating the efficiency of the doping process. A solution of PU and a dispersion of MWCNTs were prepared using the same solvent. After homogenisation and ultrasonication, the solutions were mixed and stirred with a magnetic stirrer (360 rpm) on a hot plate system for 3 days at 60° C. The final solution is now ready to be coated on a Duroid$^{®}$ substrate to realise the antenna with the calculated geometry. The preparation method is shown in Fig. 2.

3 Antenna design

The selection of PANI/MWCNTs as a patch for this kind of antenna was done on the promise that a successful antenna design based on conductive polymers could lead to the development of some interesting integrated circuits and planar structures. The dielectric substrates used is Rogers RT/Duroid$^{®}$ 5870 which has, at 4.5 GHz (operating frequency), a dielectric permittivity of 2.33 and a loss tangent of 0.0012. The carbon nanotubes selected are of multiwall type. The PANI/MWCNTs used has a conductivity of 4500 S/m. The study was carried out for three antennas based on PANI/MWCNTs in order to evaluate the influence of the thickness (50, 70 and 110 µm) on antenna performance. In fact, the thickness of PANI/MWCNTs affects its surface resistivity which influences the antenna performance. In this paper, return loss, radiation pattern and gain are investigated.

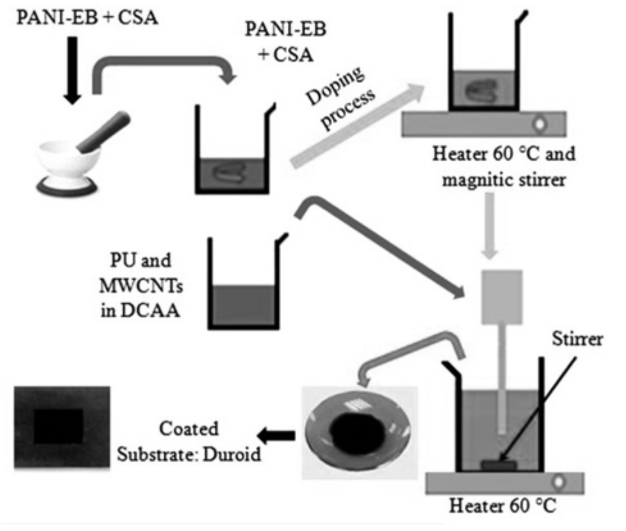

Fig. 2 *Preparation method of PANI/MWCNTs composite based-patch antenna*

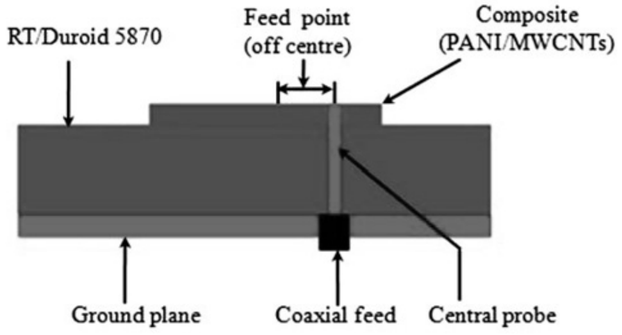

Fig. 3 *Schematic of PANI/MWCNTs based-patch antenna*

In contacting method, the radio-frequency (RF) power is fed directly to the radiating patch using a connected element such as microstrip feed or coaxial feed. The latter one is a very common technique used for feeding the patch antennas. As seen in Fig. 3 the inner conductor of the coaxial connector extends through the dielectric and is soldered to the radiating patch, while the outer conductor is connected to the ground plane. The main advantages of this type of feeding are that it can be placed at any desired location inside the composite (PANI/MWCNTs) patch in order to match with the desired input impedance and it has low spurious radiation. The disadvantage of this method is that the substrate thickness increases. The feeding location of the patch antenna (PANI/MWCNTs) has been optimised to insure a good matching with a standard 50 Ω feeding coaxial. The antenna designs were optimised for good impedance matching. The parameters used to determine the dimension of the PANI/MWCNTs patch antennas (thickness, length and width) are the substrate characteristics (permittivity, loss tangent and thickness) and the PANI/MWCNTs conductivity. The optimised dimensions of the patch antenna proposed are summarised in Table 1.

4 Simulation and measurement results

To achieve a smooth and homogeneous surface after the deposition process of the PANI/MWCNTs, parameters such as the PANI/MWCNTs thickness, PANI concentration, MWCNTs concentration and PU amount are adjusted. The resulting surface topography is investigated by means of a profilometer (Micromeasure^{TM} 2 system, equipped with STIL-DUO). Fig. 4 gives an illustration of the measured profile obtained after optimisation of the parameters. From this figure, an average of ±3 µm of the thickness variation is obtained. The different physical properties including roughness, thickness and adhesion with substrates have been analysed. It has been demonstrated that polymers with a thickness over 120 µm did not adhere well to the substrate. Therefore, we have decided to consider three-patch antennas with a thickness lower than this critical value (50, 70 and 110 µm).

Table 1 Parameters for PANI/MWCNTs patch antennas

Substrate Rogers RT/Duroid 5870			
dimension, mm^2		60×60	
thickness, mm		1.6	
permittivity		2.33	
Tan (δ)		0.0012	
Patch PANI (CSA)0.5/PU/MWCNTs			
Conductivity, S/m		4500	
thickness, µm	110	70	50
length, mm	26.5	27	27.5
width, mm	20.15	20.2	20.25
feed point (off centre, mm)	5.5	6	6.25

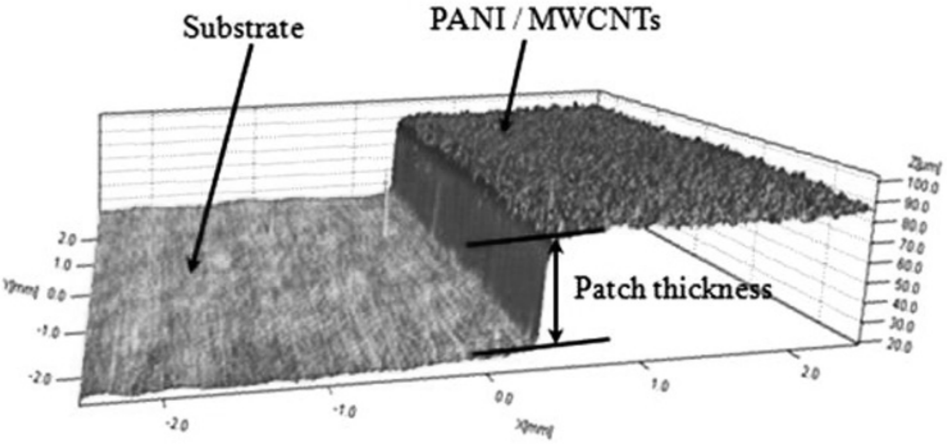

Fig. 4 *Measurement of PANI/MWCNT film thicknesses by using Micromeasure^{TM} 2 system*

The DC conductivity is measured by using Van Der Pauw method. Following the requirements of this method, four probes were placed at the corners of a square-shaped film (13 × 13 mm^2). A current (Keithley 220 current source) is sent through the sample and eight voltages are measured (DMM Keithley 196) following the procedure described in the Keithley application note (no. 7065–901–01) [36]. The conductivity in the range (10^{-6}–10^6 S/m) is extracted using a standard algorithm [37].

A systematic study has been performed to get high conductivity value with good surface roughness and homogeneous distribution of the MWCNTs. A lot of parameters affect the determination of the conductive polymer conductivity (patch thickness, conductivity of the PANI and percentage of MWCNTs and PU etc.). Indeed the maximum value was 4500 S/m.

In the study proposed, the conductive polymer has been modelled as a thin layer of finite impedance with a sheet resistance R_S estimated through the DC conductivity σ

$$R_S = \frac{1}{\sigma e}$$

where e is the thickness of the film.

From this two data (thickness, conductivity) the surface resistivity has been calculated to accurately simulate the PANI/MWCNT-patch antennas. The resulting DC surface resistivity of the PANI/MWCNTs films is shown in Table 2.

To investigate the antenna performance experimentally and to verify the simulation results, three rectangular patch antennas with different thicknesses of PANI/MWCNTs have been realised (Fig. 5). Coaxial line technique is used to excite the antenna at 50 Ω feeding point.

Table 2 Surface resistivity of PANI/MWCNTs films

Patch PANI (CSA)0.5/PU/MWCNTs			
Patch thickness, µm	50	70	110
surface resistivity, Ω/sq	4.44	3.17	2.02

4.1 Return-loss characterisation

On the basis of HFSS™® simulations we have realised three PANI/MWCNTs antennas. After the fabrication of antennas with different thickness of conductive polymer, measurements have been performed using an Agilent PNA–X series N5242A network analyser (10 MHz–26.5 GHz). Measurement results in term of return loss are presented in Fig. 6. For comparison we have also reported the simulation results in the figures.

The experimental results show that S_{11} is always better than −14 dB for the three thicknesses considered. Moreover and for all thicknesses, the measured resonance frequency is slightly shifted to higher frequencies compared with the simulated ones. This could be explained by the change in the input impedance of the antenna, the errors due to the fabrication process and the effect of a non-uniform distribution of the MWCNTs in the PANI matrix that impact the conductivity.

The experiment highlight that the proposed patch antennas achieve a good bandwidth. The bandwidths retrieved from simulation and measurement values are summarised in Table 3.

The numerical results obtained are in good agreement with the experimental data. For a thickness of 110 µm, the patch antenna exhibits a bandwidth between 4.43 and 4.76 GHz (7.33% width). The results demonstrates that for all antennas realised the bandwidth at −10 dB is larger than that calculated from conventional antennas. This result ensures that the conductive polymer is a good candidate to realise antennas compatible with different applications with good performance.

4.2 Radiation properties

It is interesting to measure the radiation patterns of these three antennas and to compare them with the simulation results.

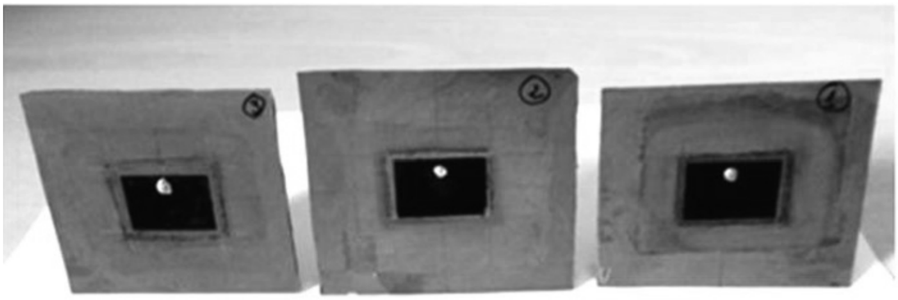

Fig. 5 *Photograph of the realised PANI/MWCNTs-antennas*

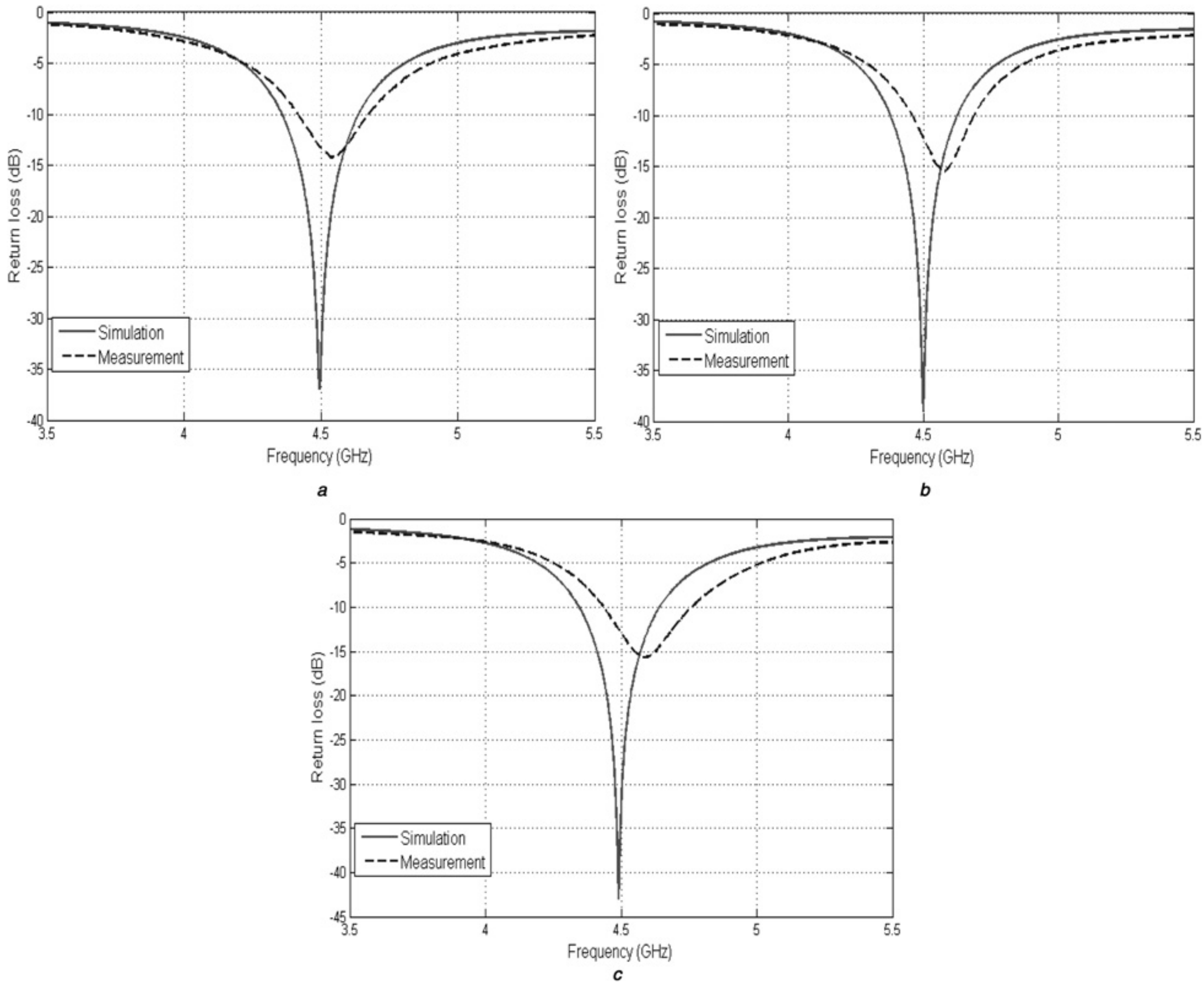

Fig. 6 *Simulated and measured return loss of the patch antennas*

a Thickness = 50 µm
b Thickness = 70 µm
c Thickness = 110 µm

The patch antennas radiation patterns of the principal planes (E and H) were measured in the IEMN anechoic chamber using a standard horn antenna (SAS–200/571) and a vector network analyser (Agilent 8735ES, 30 KHz–6 GHz) at 4.5 GHz. During the measurement, we tried to completely avoid the influence of PANI/MWCNTs antenna position on the radiation properties by totally covering the coaxial cable and SubMiniature version A connector with an absorber (Fig. 7). The measured radiation patterns for both the E and the H planes of the antennas are plotted in Figs. 8–10.

It is observed that the proposed antennas have a good radiation pattern. Actually the patch antennas display a typical patch antenna radiation pattern, with a half-power beam width of 85° to 95° for both the E-plane and the H-plane. Minor perturbations observed in the patterns were likely due to the distribution of the MWCNTs in the conductive polymers matrix, measurement errors and/or residual radiation from the coaxial and SMA connectors. In addition, it can be noted that the PANI/MWCNTs antenna prototypes have almost the same performances in terms of bandwidth and radiation pattern compared with a conventional patch antenna.

Table 3 Bandwidth for PANI/MWCNTs patch antennas

Patch PANI (CSA)0.5/PU/MWCNTs			
Patch thickness, µm	50	70	110
simulated bandwidth (at−10 dB)	280 MHz (6.22%)	245 MHz (5.44%)	300 MHz (6.66%)
measured bandwidth (at−10 dB)	253 MHz (5.62%)	235 MHz (5.22%)	330 MHz (7.330%)

In Fig. 11 the measured gain is compared with simulated values obtained using HFSS™®.

One can note that the gain increases linearly with the conductive polymer thickness in both the cases and that the slopes are in good agreement. For information, the gain simulated by HFSS™® in case of a copper-based patch antenna is 6.2 dB.

All these results together demonstrate the interest of using PANI doped with carbon nanotubes. The high electrical conductivity of MWCNTs makes this kind of patch antennas potentially very attractive.

Fig. 7 *Antenna in the anechoic chamber*

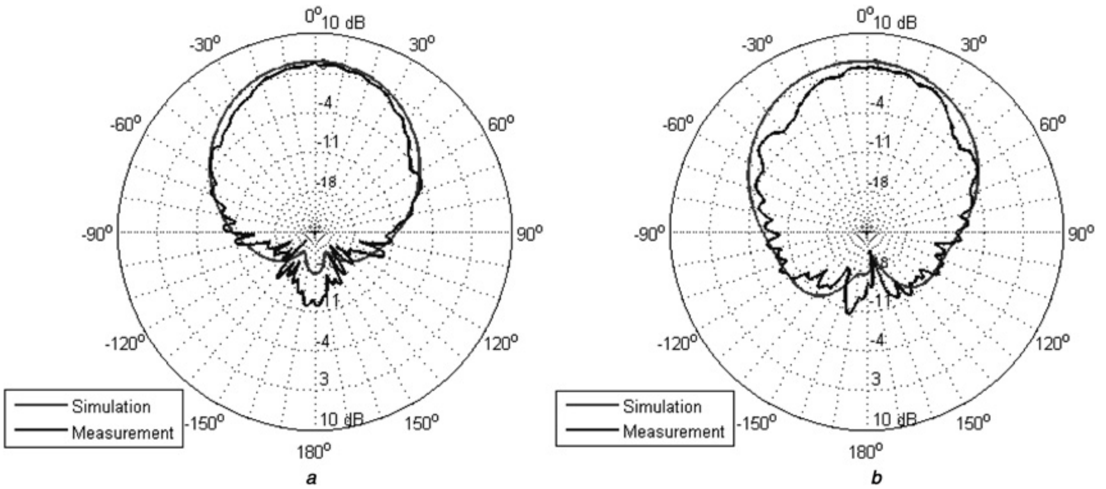

Fig. 8 *Radiation patterns of the patch antenna (thickness = 50 µm)*

a E-plane
b H-plane

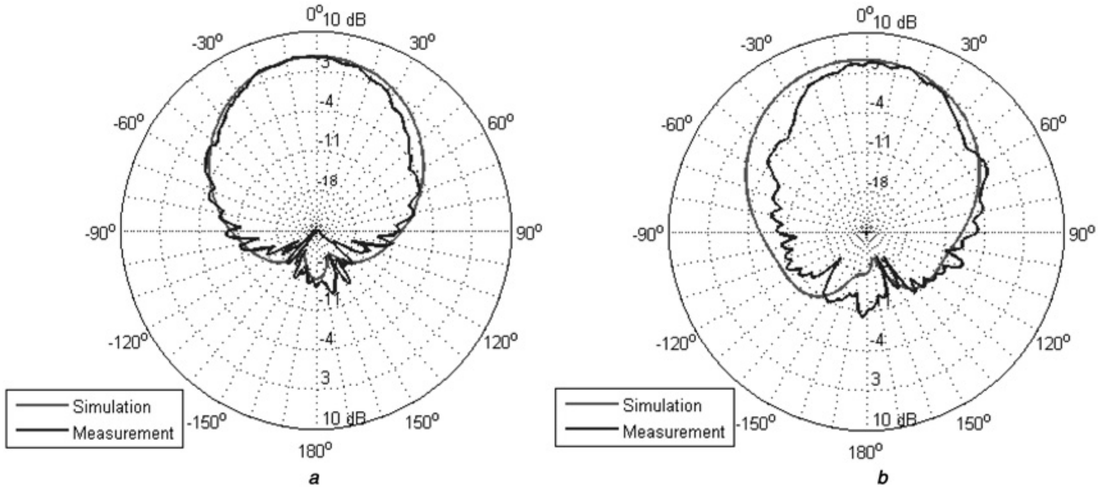

Fig. 9 *Radiation patterns of the patch antenna (thickness = 70 µm)*

a E-plane
b H-plane

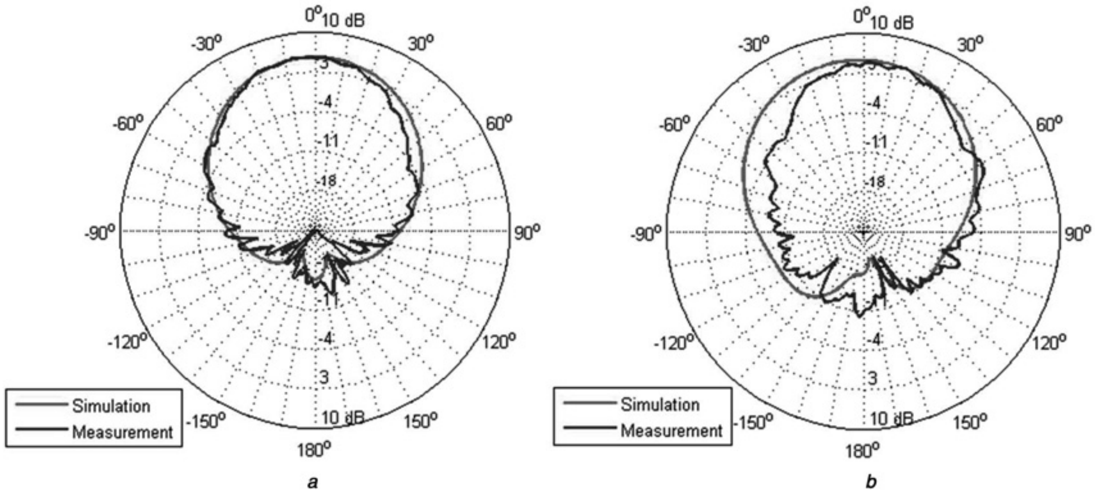

Fig. 10 *Radiation patterns of the patch antenna (thickness = 110 µm)*

a E-plane
b H-plane

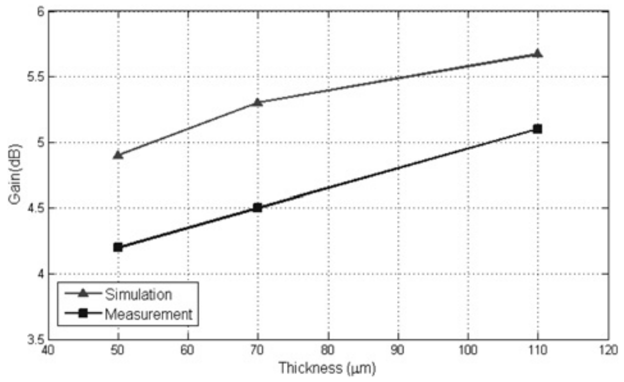

Fig. 11 *Antenna gain as a function of patch thickness ($\sigma = 4500$ S/m)*

5 Conclusion

In this paper, we have demonstrated that the realisation of PANI/MWCNTs patch antennas with good performance is now possible. Actually the comparison of PANI/MWCNTs patch antennas to the conventional ones shows that this kind of antennas are good candidates for applications where features like low cost, flexibility, anti-corrosion properties and light weight are required. Prototypes have been fabricated and measured, and good agreement with the simulation has been observed. The gains measured at 4.5 GHz for three thicknesses 50, 70 and 110 μm are, respectively, 4.3, 4.5 and 5.18 dB. Compared with the antenna structures already published, the addition of MWCNTs in the conductive polymer (PANI) results in performance improvement that makes this kind of antenna a very promising solution.

In fact the mixing of a conductive polymer (PANI) with highly electrically conductive MWCNTs leads to a patch antenna structure comparable with the one fabricated with conventional metals. Thus, these good results indicate that PANI/MWCNTs patch antenna can be commercially viable and also technically suitable for applications such as localisation systems, wireless communications and WLAN.

Through this study of conductive polymer-based antennas, we also foresee the benefits of using this type of materials for future wearable electronics. The integration of this kind of antenna into body-worn RF systems and textile structures presents a strong interest. Future work in this area may involve extending the patch antennas to study different substrates, frequency ranges and shapes for practical applications.

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