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Ionic CNT actuators and arrays – towards cost-efficient manufacturing through scalable dispersion and printing processes

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***Abstract*— As one type of ionic electroactive polymer, carbon nanotube (CNT) actuators have attracted large interest from industry and academia due to their small actuation voltages, relatively large actuation response and their flexible and soft mechanical characteristics.**

Until today, manufacturing is based on labor-intensive laboratory techniques involving costly materials, tedious handling procedures and increased safety precautions. Additionally, their sophisticated chemical compositions, poor reproducibility and sensitivity to environmental influences have so far limited a wider application range. This paper investigates the key findings in optimizing and scaling up manufacturing related processes for CNT actuators and addresses new approaches towards simplified system integration.

It was found that the predominantly used single-walled CNTs can be substituted by much cheaper multi-walled CNTs with having only minor effects on the actuator performance. First tests showed that by employing industrially scalable dispersion techniques and automated printing processes, CNT actuators can be manufactured faster, in larger quantities and with strongly reduced variance in actuator performance. New approaches for safe and simple system integration have been tested and new means of dielectric coating encapsulation were investigated.

I. Introduction

Electroactive polymers (EAP) are smart material systems that exhibit a reversible change in size or shape when stimulated by an electric field. Most attention is currently drawn to dielectric elastomer actuators (DEA), which require very high driving electric fields of up to 100 V μm^{-1} [1]. Taking into consideration the thinnest elastomer layers producible today, actuation still requires driving voltages of several kV [2]. This entails complex voltage conversion circuitry and safety precautions and is widely regarded as a major drawback for applications in any human environment.

Unlike DEA, ionic EAP operate on the principle of electrically controlled ion transport within a bi- or trilayer setup and perform a bimorph bending motion. Some exhibit large deformation and medium force transmission at low operational power (up to 3V), making them highly attractive for many applications where soft, non-hazardous and noise-free actuation is desired. However, most types of ionic actuators have not reached mass-adoption due to complex manufacturing techniques, costly materials or poor performance properties [3,4]. Ionic polymer–metal composite actuators (IPMC) for example require noble metals such as platinum or gold for their chemically plated conducting electrodes, which has an adverse effect on cost efficiency [4]. The same is true for conducting polymer actuators (e.g. polypyrrole), which are fabricated using intricate galvano-static electropolymerization processes [5]. On the contrary, CNT-polymer actuators have been manufactured utilizing standard laboratory equipment and commercially available and affordable base materials [6,7]. For this reason this paper concentrates on CNT actuators, despite their well-known problems regarding actuation performance, creep and temperature stability, which they share with other ionic EAP.

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II. CNT actuators

In 1999, sheets of macroscopic aggregates of single-walled carbon nanotubes (SWCNT) – also called Buckypaper – were reported to show an electromechanical response when laminated together with double-sided Scotch tape and submerged in aqueous electrolyte solution [6]. In 2005, Japanese researchers used this principle to build the first 'Bucky gel' actuator, which could operate in air without external electrolytes. It consists of a polymer-supported ionic liquid (IL) electrolyte layer sandwiched between two polymer-supported Bucky gel electrodes, a gelatinous room temperature ionic liquid containing SWCNTs [7]. Since then, a variety of different active materials have been studied as additives for the actuator electrodes [8,9] and various ILs have been tested for their applicability [10].

A. Working principle

The bimorph bending motion is caused by two superimposing effects, the first being the dimensional changes of SWCNTs in covalently bonded directions caused by the charge injection, which originate from quantum and double-layer electro-static effects [11]. The second effect is due to transport of oppositely charged ions of different sizes to the cathodic and anodic side of the actuator, where they form electrochemical double layers with positively and negatively charged nanotubes [7]. These ion transports result in swelling of the cathode layer and shrinkage of the anode layer, as the cations are larger than the anions (Fig. 1).

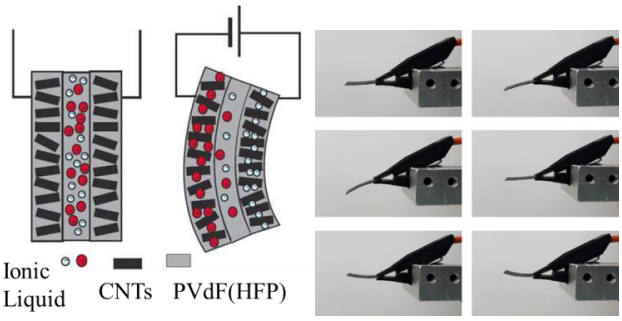

Figure 1. Left: illustration of actuation mechanism of CNT 'Bucky gel' actuators. Right: bending motion of CNT actuator

B. *Performance measurement & characterization*

A fully automated test setup has been implemented for CNT actuator performance measurements. Actuators are clamped into a fixture which provides electric contacts to both electrodes, allowing the actuator to bend in both directions. The displacement is measured via optical laser triangulation (MicroEpsilon OptoNCDT 1302, Fig. 2a) with a range of 20mm and an accuracy of +/- 4µm. A force sensor (KYOWA LTS20GA, 500mN, 0.01mN resolution, Fig. 2b) is mounted on the moving part of a horizontally adjustable servomotor (Nanotec Munich KOWI), providing position resolved blocking force measurements on one side of the actuator specimen. A potentiostat triggers the actuation by providing a predefined tunable voltage level with high accuracy, varying the electric current according to the actuator's impedance currently present. Other parameters obtained are electric current values, motor positions, temperature variations and heat radiation images.

The material strain inside the electrode layers is regarded as a key performance indicator for CNT actuators [7]. For thin actuator strips, the electrode strain ε can be directly calculated from the measured tip displacement δ using (1):

$$\varepsilon = \frac{2d\delta}{L^2+\delta^2} \qquad (1)$$

L is the free length and d is the thickness of the actuator strip. The strain indicates the unidirectional change of length within the electrode layers (Fig. 3a) and therefore relates directly to the induced volumetric swelling or shrinking. Strains of up to 2% have been reported [9].

However, (1) does not take into consideration the width of the tested actuator sample and is only valid for thin strips. The free tip displacement decreases for wider actuator geometries, because the volumetric changes also results in a curvature across the width that interferes with the longitudinal bending motion due to increased compression stress at both edges of the cross section (Fig. 3b).

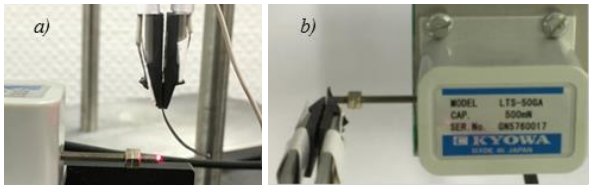

Figure 2. *a)* Laser triangulation and *b)* force sensor for performance evaluation of CNT actuators by measuring force and displacement.

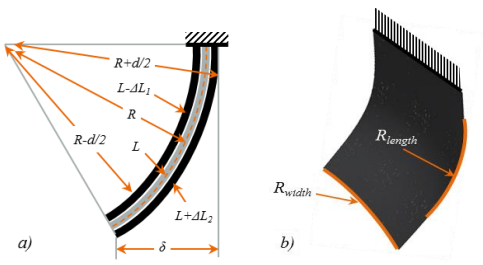

Figure 3. *a)* Schematic drawing for explaining the evaluation of the strain from the displacement $(R-d/2)/(R+d/2) = (L-\Delta L_1)/(L+\Delta L_2)$, where R is the curvature radius and ΔL_1, ΔL_2 are the induced length-change in each electrode. The strain difference ε is given by $\varepsilon = \Delta L_1 + \Delta L_2)/L = d/R$. Since R is given by $(L^2 + \delta^2)/(2\delta)$, we can obtain (1).

III. Scalable Dispersion Techniques

In order to yield large electrochemical effects in the actuator electrodes – and thus high internal material strain for actuator deflection – a thorough separation of CNT agglomerates and an even distribution of all material components in the final dispersion is essential. For actuator dispersions, it was successfully attempted to replace single-walled CNTs with much cheaper multi-walled CNTs. Furthermore, continuous dispersion methods capable of handling large material quantities were tested instead of batch-to-batch dispersion techniques that make extensive use of ultrasonic baths and laboratory stirring equipment.

A. *Contemporary laboratory methods*

The first step in actuator fabrication is to make homogeneous dispersions for both actuator electrode layers and the separator layer. These dispersions contain all material components in matching proportions, which were found through extensive experimental investigations [7,8,9].

Actuators are fabricated using polyvinylideneflouride-cohexafluoropropylene (PVDF(HFP)) as a supporting polymer, SWCNTs as a conductive additive, and ionic liquid EMI-BF_4 as an IL-electrolyte [8]. CNTs were selected due to their high electronic conductivity, good electrochemical stability, mesoporosity, and sufficient specific surface area. To improve actuation performance, CNT electrode additives such as Polyaniline (PANI) were studied [9]. The combined PANI/CNT electrodes showed larger tip deflection compared to pure CNT electrodes.

Organic solvents like 4-methyl-2-pentanone (4M2P) are needed to dissolve the PVDF backbone polymer, which is responsible for the actuator stiffness and serves as a matrix for the ionic liquid and the carbonaceous additives. Besides 4M2P, Dimethylacetamide (DMAc) and N-Methyl-2-pyrrolidone (NMP) have been tested for solving the PVDF and dispersing the carbonaceous particles. The authors have discontinued such experiments due to the high toxicity levels of these solvents and the strict safety precautions required.

After dissolving the PVDF in 4M2P SWCNTs, PANI and EMI-BF_4 is added and stirred with a magnetic stirrer device in a closed flask to prevent the solvent from evaporating. Stirring has been reported to last up to 72h to yield sufficient homogenization. Stirring is followed by high frequency treatment in an ultrasonic bath for up to 24h [8,9]. After casting (Fig. 4b) and drying, a single sheet of electrode material is obtained (Fig 4c).

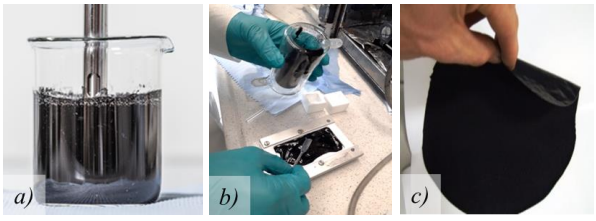

Figure 4. *a)* high energy blade stirring of CNT dispersion. *b)* manual casting of electrode dispersion into PTFE mold. *c)* electrode sheet used for actuator assembly and subsequent performance testing.

B. *Using MWCNT instead of SWCNT*

For cost optimization, multi-walled CNTs (Nanocyl® NC7000) have been tested for actuator manufacturing. NC7000 are produced via the Catalytic Chemical Vapor Deposition (CCVD) process and are commercially available in large quantities. Their primary applications are in electronic packaging, polymer amplification and as power enhancement additives for Li-ion batteries. Compared to most SWCNTs, they are up to 1000 times less expensive, depending on the material source. However, they have different electrochemical and physical properties, so the dispersion and drying processes needed to be adjusted.

As dispersing MWCNTs initially appeared to be easier, the casted electrode layers showed large areas of cracking and crumbling when dried. This happened for all drying processes such as air drying, heat plate drying and vacuum drying (Fig. 5). First, it was observed that one reason for cracking was the excessive use of solvent. After reducing the amount of 4M2P, cracks were still existent to some extent. As de-agglomeration of MWCNTs was easier during dispersing, it was assumed that the dispersion is more likely to re-agglomerate while drying. Dispersing agents were used to keep CNT bundles separated (Fig. 6). Extensive laboratory tests and Design of Experiments (DoE) methods were conducted to develop an optimized dispersion with MWCNTs (Table I).

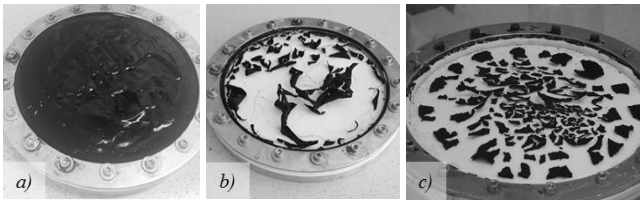

Figure 5. MWCNT dispersion *(a)* before and *(b)* after drying with too much solvent and *(c)* after drying with less solvent. Cracking appeared due to strong re-agglomeration of CNT bundles during the drying process.

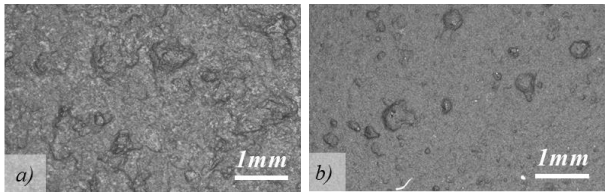

Figure 6. Microscopic images of dried MWCNT dispersions without *(a)* and with *(b)* dispersion agent PE/L 61. Re-agglomeration of large MWCNT bundles during drying was decreased.

TABLE I. COMPOSITION OF OPTIMIZED MWCNT DISPERSION FOR CNT ACTUATOR ELECTRODES

Component	**Function**	**Mass percentage (wt%)**	
		Wet state[a]	***Dry state***
PVDF	Polymer matrix	5.4	30.64
MWCNT[b]	Active material	0.75	4.3
PANI	Conductive additive	3.4	19.2
BMI-BF_4	Ionic liquid	8.0	45.7
PE/L 61	Dispersing agent	0.03	0.16
4M2P	Solvent	82.42	-

a. Dispersion including solvent, b. Nanocyl® NC7000

For the separator layer of the actuator, either pre-fabricated Nafion® membranes or self-constructed layers of PVDF(HFP)/BMI-BF_4 are suitable. Nafion® is a sulfonated tetrafluoroethylene based fluoropolymer-copolymer with special ionic properties. It is mostly used as a proton exchange membrane in fuel cells and has also been tested as an easy-to-use separator membrane for ionic CNT actuators by the authors [12]. However, it is especially well known for its great performance as a separator membrane in ionic polymer–metal composite actuators (IPMC) [4].

The material cost for all electrode and separator components per gram and per electrode (size 2cm x 1cm) are listed in Table II. For a single actuator, two electrodes and one separator layer is needed.

The impact on material costs of utilizing the new MWCNT dispersion instead of well-established SWCNT mixtures is shown in Fig. 7. The most cost-efficient material composition for CNT actuators is a MWCNT electrode paired with a self-made PVDF/IL separator membrane, costing only around 0.60 € per actuator compared to 4.48 € for a SWCNT actuator.

TABLE II. ASSEMBLY OF COMPONENT COSTS

Component	**Function**	**Cost (€)**	
		per gram	***per electrode***[a]
PVDF	Polymer matrix	0.68	0.01
SWCNT[b]	Active material	1070.-	1.93
MWCNT[c]	Active material	0.10	0
PANI	Conductive additive	8.68	0.04
BMI-BF_4	Ionic liquid	1.78	0.02
PE/L 61	Dispersing agent	0.10	0.01
4M2P	Solvent	0.03	0.20
			per separator
Nafion®	Separator membrane (commercial)	-	0.50
PVDF/IL	Separator membrane (self-made)	1.23	0.06

a. Rectangular shape 2cm x 1cm, 0.1mm thickness,
b. CHASM Signis® CG300,
c. Nanocyl® NC7000

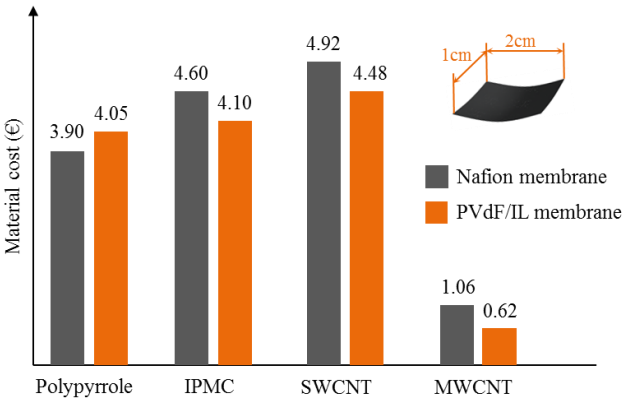

Figure 7. Comparison of material costs between gold-plated PPy and IPMC actuators, SWCNT actuators and MWCNT actuators of the same size, including the choice of separator material.

C. *Industrially scalable dispersion methods*

The before mentioned optimized material compositions were achieved through extensive laboratory dispersion techniques, some of which are transferable to large-scale continuous dispersion processes. The exact fabrication procedure is given in Table III, listing all relevant steps and time durations, respectively.

By applying high-power devices such as an ultrasonic sonotrode it was possible to reduce the overall dispersion time to less than 3 hours, which is a significant improvement compared to conventional techniques taking up to 4 days [8]. The described procedure results in a homogeneous viscous slurry dispersion ideal for subsequent printing applications.

Both continuous high speed blade stirring (dissolver) and inline ultrasonication were applied for large dispersion quantities in order to validate the up-scaling potential of the process (Fig. 8). Viscosity measurements were performed to qualify the dispersions for individual printing techniques. Furthermore, the dispersion quality was verified by measuring the optical and electrical properties of either printed or layer-casted electrode sheets with specified sample thickness.

Air drying was finished after 12 hours, forced heat-plate drying was finished after 3 hours and combined vacuum and heat drying took 1 hour to complete. The dried samples were subsequently assembled to actuator specimen via heat pressing and tested for blocking force and displacement features using the test setup described in section II.

TABLE III. OPTIMIZED FABRICATION PROCESS FOR A MWCNT DISPERSION APPLICABLE FOR CNT ACTUATOR ELECTRODES

Dispersion technique	**Material components involved**	**Duration [min]**
Ultrasonic sonotrode, 40kHz	Dispersing MWCNTs in 4M2P	8.5
Magnetic stirrer 600 rpm (flask immersed in hot water 80°C)	Adding PVDF(HFP) until dissolved	50
	Adding BMI-BF_4	10
	Adding PANI	10
	Adding dispersing agent	10
Dissolver 4000 rpm	All	5
Ultrasonic bath (removing air bubbles)	All	60
Sum		153.5 **(2.56h)**

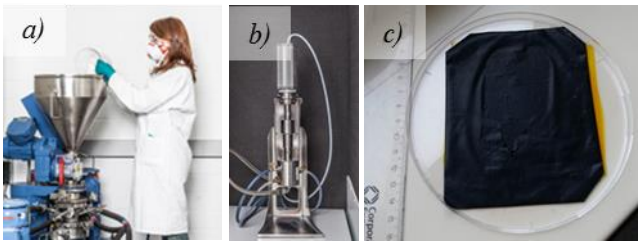

Figure 8. *(a)* Inline disperser Netzsch Micro Ψ-Mix with 2000 rpm rotational speed and 1-2m^3/h throughput potential. *(b)* Industrial ultrasonic homogenization Hielscher UIP500HD with 500W/20kHz cavitational power. *(c)* Large printed sample electrode for optical and electrical characterization.

IV. SCALABLE PRINTING TECHNIQUES

Up to now, the manufacturing process for CNT actuators has been described as a laboratory method of successively casting the electrode and separator dispersions into preformed PTFE casting molds by a layer-by-layer fashion [7]. The subsequent drying process was often performed using vacuum ovens for complete solvent evaporation and has been reported to last up to 72h. Hot-pressing and manual cutting of dried multilayer actuators is followed by gold plating of electric contacts for actuator testing.

It is now demonstrated that printing can be a fast and simple way to obtain various actuator geometries and sizes. First attempts have been performed using a 2D dispensing nozzle as a printing device [13], where complicated actuator shapes were printed in an automated way. However, this fabrication process allowed for printing only one single actuator at a time, so it is not suitable for large scale and cost-efficient industrial manufacturing of CNT actuators.

For this reason, semi-structured and non-structured printing methods have been tested. For an evaluation of feasibility, the discontinuous coating machine Easycoater EC 63 from Coatema Coating Machinery GmbH (Germany) was utilized (Fig. 9). It is equipped with a complete enclosure box and vacuum extraction/scavenging system for explosion protection which facilitates handling of solvent-based inks and pastes. It has a programmable infeed slide carrying an adjustable mounting rack with modular fittings for different printing devices. This permits the operation of non-structured slot-die printing and semi-structured blade and screen printing tools in one single machine.

A. *Non-structured printing*

With slot-die and blade printing, homogeneous layers with exactly controlled thickness were applied onto the printing table (Fig. 10). For screen printing, the viscosity of the dispersion had to be increased to prevent the slurry from delayed screen penetration. With the integrated heating functionality of the printing table, the applied dispersions were able to be dried in a controllable way, until no solvent residues could be detected.

Figure 9. Discontinuous coating machine EC 63 for explosion protected printing of solvent-based inks and dispersions.

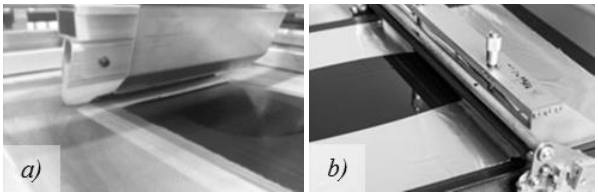

Figure 10. *(a)* Non-structured full-surface screen printing and *(b)* blade printing of MWCNT/PVDF/IL/PANI dispersions.

Large continuous non-structured multilayers were achieved by successive layer-by-layer printing of electrode/separator/electrode dispersions, with short intervals between individual steps for drying and exchanging the dispersion container and feeding tubes. The exact fabrication procedure is given in Table IV, listing all relevant steps and time durations, respectively. It is demonstrated that the entire printing process may last less than 1.5 hours including hot air assisted drying and necessary cleaning activities, which is a significant improvement over manual fabrication processes taking up to three days [8].

Fig. 11 compares performance testing results of MWCNT and SWCNT actuators for both conventional mold casting fabrication and industrial slot-die printing method. It illustrates the maximum strain for both material systems. As expected the general performance of MWCNT actuators is not as good as the performance of SWCNT actuators, however, they yield more than half the strain. Most noticeable is the fact that the printed versions of both actuator species show a considerable smaller statistical spread and are therefore more reproducible. Twelve samples have been tested from each actuator species.

TABLE IV. TIME DURATIONS FOR PRINTING STEPS FOR NON-STRUCTURED MULTILAYERS UTILIZING EASYCOATER EC 63

Comprehensive steps for multilayer generation	**Duration [min]**
Start up printer and set printing program	5
Install dispersion and attach feeding tubes to slot-die	5
Scavenging of printing compartment (safety procedure)	1
Start pump to fill feeding tubes with electrode dispersion	2
Print first electrode layer	0.5
Hot air assisted drying of first electrode layer	10
Switch tubes and printing head, fill with separator slurry	5
Print separator layer	0.5
Hot air assisted drying of separator layer	10
Switch tubes and printing head, fill with electrode slurry	5
Print second electrode layer	0.5
Hot air assisted drying of second electrode layer	10
Retreive multilayer compound	3
Cleaning of tubes, slot-die, compartment and pumps	20
Sum	76 **(1.26h)**

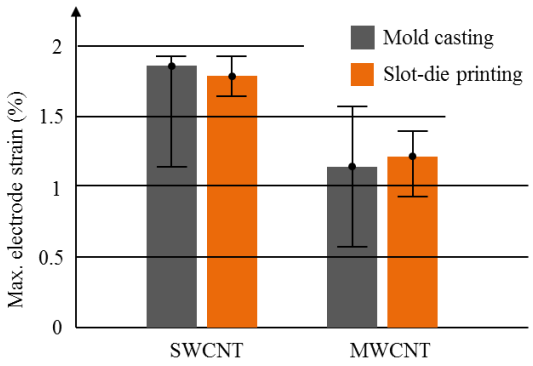

Figure 11. Strain comparison of casted and printed SWCNT and MWCNT actuators (20mm x 5mm) made from non-structured dried multilayer films.

B. *Semi-structured printing*

Semi-structured electrodes for CNT actuators were fabricated with masked blade printing, where rectangular, circular and triangular actuator geometries could be implemented without manual finishing steps such as cutting or stamping (Fig. 12). Different actuator shapes can be applied to generate different kinematic motions for individual application systems. This process is industrially up-scalable and yields unlimited variations of actuator geometries.

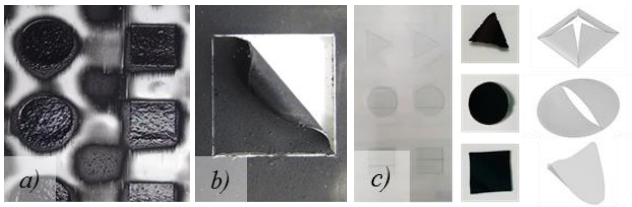

Figure 12. *(a)* Semi-structured blade printing of optimized CNT actuator electrodes using prefabricated PTFE mask. *(b)* dried multilayer actuator (electrode/separator/electrode) after successive layer-by-layer printing. *(c)* Geometric variation of EAP shapes achieved via masked blade printing of CNT-based dispersions. Individual shapes can be used for different motion kinematics.

V. SYSTEM INTEGRATION

To apply CNT actuators in potential real-world applications, fast and simple electric connections are required and the capability of operating in different environments (open air, humidity) must be available. In this chapter we present first feasibility studies of implementing both functionalities already during the manufacturing process.

A. *Electric contacts*

Conventional CNT actuators are not fitted with electric connections. Instead, the bare electrode surfaces are mechanically clamped in between two conductive terminals, mostly for testing purposes. However, future applications require reliable electric contacts for simplified system integration. For testing such electric contacts, different flexible conductors were attached to the PTFE masks for direct integration into the actuator electrodes during the printing process.

Fig. 13 shows the successful application of silver plated conductive threads, copper braids and other conductors into printed actuator electrodes. All methods of electric contacting came along with a significant improvement of the contact resistance between the main conductor and the polymer electrodes. The resulting actuation response and displacement was observed to increase up to 10% and 5%, respectively.

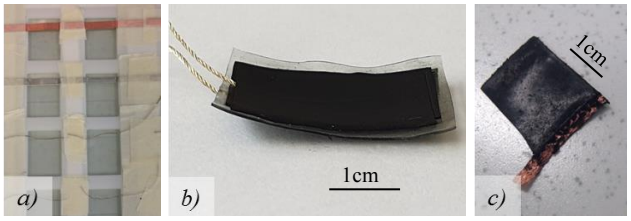

Figure 13. *(a)* mask prepared for printing. *(b)* Fully printed CNT actuator after drying with integrated electric contact material (silver plated conductive thread) for simplified assembly and system integration. *(c)* copper braid connection firmly attached to a CNT polymer electrode after direct printing.

B. Encapsulation

After printing, the actuators are still in their natural form and their conductive electrode surfaces are directly exposed to air and thus capable of interacting with the surrounding environment. Various encapsulation materials like PDMS, PU, nitrocellulose and paraffin-composites were tested [14].

For CNT actuators it was assumed that PVDF could also be beneficial for encapsulation, because it is already used for both electrode and separator layers. It was found that dip-coating of single actuator elements into a solution of PVDF in 4M2P (0.5 wt%) resulted in a very thin layer (10 – 25 µm) of PVDF deposited around the actuator. First tests showed that such encapsulation yields a sufficient dielectric behavior for electrical insulation and prevents penetration of water vapor into the active layers of the actuator.

C. Applications and prototypes

The CNT actuator technology is at an early stage of development and currently there are no real-world applications due to the deficiencies mentioned in section I. However, such soft and flexible actuators with material-intrinsic shape change capabilities are believed to play a small role in civil engineering and architecture [15] where they could have a beneficial impact on the design and functionalities of future lightweight building envelopes. They are planned to be fitted into textile membrane constructions for added functionalities such as switchable breathability, automated ventilation or controllable light shading devices. First functional prototypes were already built to validate their general feasibility (Fig. 14). Future steps will include measures for directly printing multi-actuator arrays onto flexible substrates with subsequently printing the electric contacts and encapsulation layers.

In 2014, the same research team has prototypically employed CNT actuators for driving a micro-actuator pipette which was targeted for medical dosing applications [12].

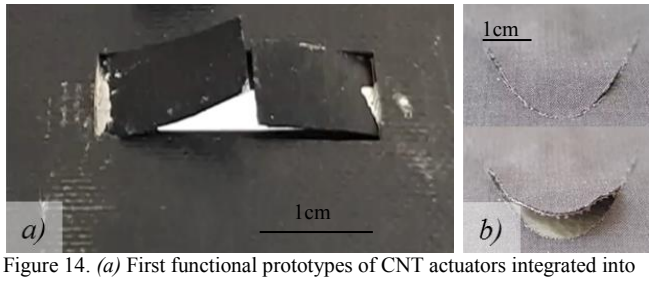

Figure 14. *(a)* First functional prototypes of CNT actuators integrated into paper-based flexible elements for investigating their kinematic behavior *(b)* Printed CNT actuators embedded into textile membranes for generating controllable openings → target application: adaptively breathable building façade.

VI. Conclusion

Dispersion and manufacturing techniques for CNT actuators have been advanced through adopting scalable industrial processes and material costs were reduced by using MWCNTs instead of SWCNTs. Despite the weak forces and displacements obtained, the authors believe that such actuators will find their way first into niche applications in the medical or aerospace sector before reaching maturity for more robust systems like soft actuators for building facades. More research is necessary to reach longer life cycles, better material handling and profound strategies for end-of-life treatment. Simulation and modeling could help design specific applications through focused optimization of kinematic motions and actuator geometries.

Acknowledgment

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