DOI: 10.1002/ ((please add manuscript number))

**Article type: Full Paper**

An autonomous inchworm-like robot propelled by an actuator akin to a supercapacitor.

Indrek Must\*, Friedrich Kaasik, Inga Põldsalu, Lauri Mihkels, Urmas Johanson, Andres Punning, and Alvo Aabloo

I. Must, F. Kaasik, I. Põldsalu, L. Mihkels, Dr. U. Johanson, Dr. A. Punning, Prof. A. Aabloo A.

Intelligent Materials and Systems Lab (http://www.ims.ut.ee), University of Tartu, Nooruse 1, Tartu, 50411, Estonia

E-mail: indrekm@ut.ee

Keywords: EAP actuators, flexible supercapacitors, in air, ionic liquids, carbon actuators

We report on a centimeter-scale robot, propelled by a single supercapacitor-like ionic electroactive polymer (IEAP) laminate actuator. The cyclic locomotion of the robot is inspired by an inchworm, while the IEAP laminate is used concurrently as an actuator and a structural member. The 827-mg robot is able to crawl on a smooth surface in open air, solely by undulation of its body. The microprocessor-controlled robot has an on-board lithium battery and uses a pulse width-modulated signal to drive the actuator. The robot is able to carry its own power supply and even an extra payload. The constructed biomimetic robot is distinctive for the use of a non-planar actuator that is shape-programmed during its manufacturing process.

The IEAP material with activated carbon-based electrodes stands out by its exclusive combination of high electrically-induced strain and high bending modulus. In addition, it is also characterized by an outstanding capacitance per surface area and a high cyclic efficiency. Due to these properties the laminate is one of the few IEAPs that meet concurrently the requirements of miniature robotics as well as energy storage systems.

# Introduction

The conventional, commercially available supercapacitors are sealed devices, serving for a single purpose – storage of electric energy. Supercapacitors are based on highly porous carbon and highly volatile anhydrous electrolytes, which grant stable operation and high power density, but require complete dehydration and encapsulation before deployment. In recent years, various novel manufacturing methods have been proved to be suitable for their fabrication. Supercapacitors can be printed layer by layer on diverse substrates using the conventional printing technologies, or made in the form of free-standing flexible films, fibers, or textiles.[1-5] The new forms enable their deployment in wearable appliances, often concurrently as a structural member. Unlike the traditional sealed supercapacitors, a flexible supercapacitor possibly operates directly in ambient air. As a consequence, the influence of the environmental factors such as ambient humidity could no longer be ignored. Therefore it should be remembered that the energy density of the hydrous supercapacitors is decreased due to the low electrochemical window of the aqueous electrolytes.

While the working principle of the supercapacitors is based on the movement of ions between the two porous electrodes, there always exists a side-effect – expansion of the electrodes.[6] In the industry of supercapacitors this effect is unacceptable, as it reduces the efficiency of the device and can finally deteriorate the electrodes and the membrane.[7] On one hand, the effect of volume change can be cushioned by selection of the components – rigid current collectors and mutually optimized carbon electrodes and electrolyte. On the other hand, it is possible to optimize the supercapacitor-like structure by maximizing the dilatometric effect. Forfeiting a small proportion of the capacitance, efficiency, and cycle life of the electrochemical system, it is possible to gain a similar laminate of outstanding flexibility and volumetric expansion coefficients. Charging of this tri-layer laminate generates large strain differences between the oppositely charged electrodes, while the amplitudes of the strains are commonly related to the charging level.[8] This in turn causes buckling of the whole membrane. The resulting flexible supercapacitor is in point of fact a specific type of smart materials - ionic electromechanically active polymer (IEAP).

The smart material actuators have been considered being promising for bio-inspired robotic applications almost since the beginning of their development.[9] The optimal operating voltage of the IEAP actuators is in the same range with the contemporary microelectronics – a few volts. In contrast to the conventional electric motors and linear actuators, primarily used in the robotic applications, the actuation character of the soft smart material actuators – changes in the size or shape of the structure itself – is intrinsic to the living nature. The up-to-date IEAP actuators are qualified either by high elastic modulus[10] or high electrically induced strain.[11-13] An actuator suitable for microrobotics must combine these two properties.

An IEAP actuator can easily be miniaturized, while the full potential of the IEAP actuators reveals itself at the lower end of the size scale. IEAP micro-actuators of lateral dimensions of as small as under 50 µm have been developed for the lab-on-chip applications.[14] The few examples of the possible spheres of application of the IEAP actuators are robotic surgery,[15] rescue work in the collapsed environment,[16] surveillance,[17] and manipulation of fragile objects.[18]

Due to the low efficiency of the IEAP actuators, the overwhelming majority of the proposed applications are powered by the off-board power supplies. Hence, the development of fully autonomous robots based on smart materials is a challenging task. Until now, the few examples of the mobile robots, propelled by IEAP actuators and capable to carry their own power supply, have been developed exclusively for underwater operation.[19, 20] The reason is elementary: the swimming robots require only infrequent strokes from an actuator for locomotion, while the buoyancy compensates the weight of battery.

In the current paper we report on the first IEAP robot in the world, capable to run on a smooth surface in open air, carrying its own power supply. For this application we developed an IEAP actuator where high strain and high elastic modulus are combined in a single laminate. Its actuation performance is achieved by appropriately tuned manufacturing process and use of the carbide-derived carbon as an active electrode material, and stable in-air operation is enabled by ionic liquid electrolyte. The characteristic looping gait of the robot and locomotion of its biomimetic prototype – inchworm – are depicted in **Figure 1**. The robot is microprocessor-controlled, can move autonomously on a smooth surface and even carry noteworthy extra payload. Its largest detail is a single IEAP actuator, which, similarly to inchworm, morphs the shape of the whole robot. To the best of our knowledge, weighing only 827 mg with the on-board battery, this is the lightest autonomous electroactive polymer robot developed so far.



Figure 1. a) Locomotion of an inchworm gives inspiration for the design of b) biomimetic IEAP robot.

# Towards biomimetic and autonomous robots

Here we bring some examples and concepts of bioinspired and morphing microrobots, proposed by several research groups in the recent years.

The concept of inchworm-like locomotion is not new in the field of micro-robotics. The hygromorphic bilayer is one of the most used types of actuator for this application. The centimeter-scale robots ‘walking’ on a ratcheted track have been developed using the humidity-sensitive thermally cross-linked poly(allylamine hydrochloride)/poly-(acrylic acid) films[21, 22] and graphene/graphene oxide fibers.[23] This type of actuation does not involve any electrical input or control. Locomotion on a ratcheted surface has also been demonstrated with gel, undergoing spontaneous periodic swelling and de-swelling oscillations.[24] The shortcoming of robots based on hygromorphic or self-oscillating actuators is the limited control over their operation.

Another concept for propelling the morphic micro-robots - the pneumatic actuators - has attracted increasing attention due to their high flexibility, large deformations, and relatively high actuation speed.[18, 25-27] However, these robots are connected to external pumping and commuting units via trailing bundle of tubes. The off-board equipment for such robots is, as a rule, several orders of magnitudes larger and heavier than the robot itself, conflicting the whole concept of autonomous robots.

The working voltage of the dielectric elastomer or piezoelectric actuators is limited by the dielectric breakdown voltage of the dielectric, and is typically between a few hundreds volts and tens of kilovolts [28]. Utilization of dielectric elastomer or polymer piezoelectric actuators in the autonomous robots is challenged due to the complexity of miniaturized high-voltage power supplies.

Ionic polymer-metal composite (IPMC) is supposedly the best known type of IEAP. Therefore IMPC actuators have found widespread deployment in biomimetic robotics. As the traditional IPMCs rely on the presence of water as a solvent, the IPMC actuators are mostly used in the underwater applications. A plethora of IPMC-driven underwater robots, inspired by, for example, rays[29], jellyfish,[30] stalked protozoa,[31] insects,[32] or snakes[33], have been reported in the scientific journals and conferences. IPMCs also show promising results as controlled surfaces of submarines.[34] The IPMC-based in-air operating shape-morphing robots are inspired, for example, by worms[35] or amoeba,[16] or are just some engineered structures without any particular biological prototype [36]. However, the lifetime of these proof-of-concept robots in air is limited, due to evaporation of the solvent. All of them are, as a rule, powered through trailing wires.

# Design of the biomimetic robot

## IEAP actuator

An actuator that can be applied in the construction of a biomimetic microrobot must be able to generate high electrically induced strains, must be able to support the weight of the robot, have the electrodes with exceptionally high electronic conductivity, and should not have considerable creep. An actuator that combines all these properties was manufactured by the combination between casting, spray-painting, and lamination methods.

The actuator is manufactured by first fabricating the membrane containing polyvinylidene fluoride co-hexafluoropropylene (PVdF-HFP) and 1-ethyl-3-methylimidazolium trifluoromethanesulphonate (EMITFS) ionic liquid by casting method. The cast membrane consists of PVdF-HFP that is highly swollen in the EMITFS ionic liquid. The ions of EMITFS ionic liquid can easily move in the fluoropolymer network. The thickness of the cast membrane is approximately 120 µm.

Carbide-derived carbon with an outstanding electrically induced strain[37] is chosen as the active electrode material. A binder is generally needed to form compact layers of carbonaceous electrodes – PVdF-HFP is used for this purpose. The electrodes are formed by painting layer-by-layer the mixture of TiC-CDC, PVdF(HFP), EMITFS, and an appropriate solvent on the membrane. A total of approximately 20 layers of electrode were painted on the actuator to achieve a total thickness of 450 µm. During removal of the volatile solvents used in the painting stage, the individual grains of CDC are tightly wrapped together by the fluoropolymer threads. The outstanding mechanoelectrical properties are achieved by the precise selection of the ratio between the components in the electrode. An exceptionally high electronic conductivity of the electrodes is achieved by gluing three layers of 130-nm gold foils on both sides of the laminate to form current collectors. The scanning electron microscope image of the cross-section of the IEAP is given in **Figure 2**.

As the final step of the manufacturing process, the laminate is shape-programmed. The conventional IEAP actuators are typically made in a planar sheet form. Instead, the constructed IEAP robot utilizes an actuator with a curved initial shape. The initial shape is programmed using heat-treatment during the manufacturing process. The initial shape of the actuator retained well during the performed experiments without any noticeable creep.

An IEAP with non-planar shape is advantageous due to the following consideration: It is known that by applying electric charge between the IEAP actuator, the strain difference is formed between the sizes of opposite electrodes. Consequently, a piece of IEAP with comparable dimensions in length and width turns into a bowl shape. The initially curved shape helps to confine the preferred direction of bending action to the lengthwise rather than to the transverse (width) direction.

In addition to the use of curved initial shape, the key for fabrication of an actuator with a high strain and stress lies in precise selection of the ratio between the components in the electrode and the thickness ratio between the membrane and the electrode.



Figure 2. Scanning electron micrograph of the cross-section of the IEAP.

## Construction of the Robot

The most evident part of the robot is the curved IEAP actuator. The actuator is much larger than the rest of the details; therefore it seems as if the components of the robot were attached to the actuator and not *vice versa*. To implement cyclic motion, the control waveform is generated by an on-board microcontroller, while the driving electric current of the actuator is amplified by an H-bridge integrated circuit. A lithium-polymer (LiPo) rechargeable battery was chosen for the on-board power supply. The constituents of the robot are depicted in **Figure 3**a. The weight proportions of the components are given in Figure 3b. As expected, the heaviest component of the system is the LiPo battery, giving 42% of the total weight of the robot. The weight of the IEAP actuator itself is just over a quarter of the total mass.

Miniature bristles aligned in one direction support both ends of the robot on the ground, while the movement is blocked in the opposite direction. By doing so, the inchworm-like shape-morphing robot can crawl by dragging alternately its front or back end along the surface.

The details about the construction of the robot are given in the **Supporting Information**.



Figure 3. a) Construction elements of the IEAP robot. b) Weight proportions of the components.

# Results

## Locomotion characteristics of the constructed robot

The robot is able to move on the surfaces that have the necessary amount of roughness – cardboard and satin glass provided the best results**. Figure 4**a depicts morphing of the shape of the IEAP robot moving on a satin glass during one working cycle. The robot was driven with a control signal depicted in **Figure 5**a, with the PWM duty cycle fixed at 23.5%. The robot moved 16 mm in one 26-s working cycle.

The robot’s ability of performing useful work is demonstrated in Figure 4b. The robot carries a steel washer as a payload, which weighs 890 mg, on a cardboard surface. The weight of the payload is slightly more than the weight of the robot itself – 830 mg. The payload does not reduce considerably the speed of the robot – it still advanced 14 mm during one working cycle in the driving conditions equal to the previous experiment. In Figure 4c, the robot climbs up a hill with an angle of climb of 11º. The robot advanced 14 mm in one working cycle of equal parameters with the previous experiments. In Figure 4d, the robot, which is hanging on steel pins from one of its ends, lifts itself up. It proves that the rigidity of the IEAP actuator is chosen properly, so that the weight of the robot does not constrain its actuation. More importantly, no creep in the actuator shape was observed even under load. The used 10-mAh lithium-polymer battery can power the robot for approximately 8 minutes.

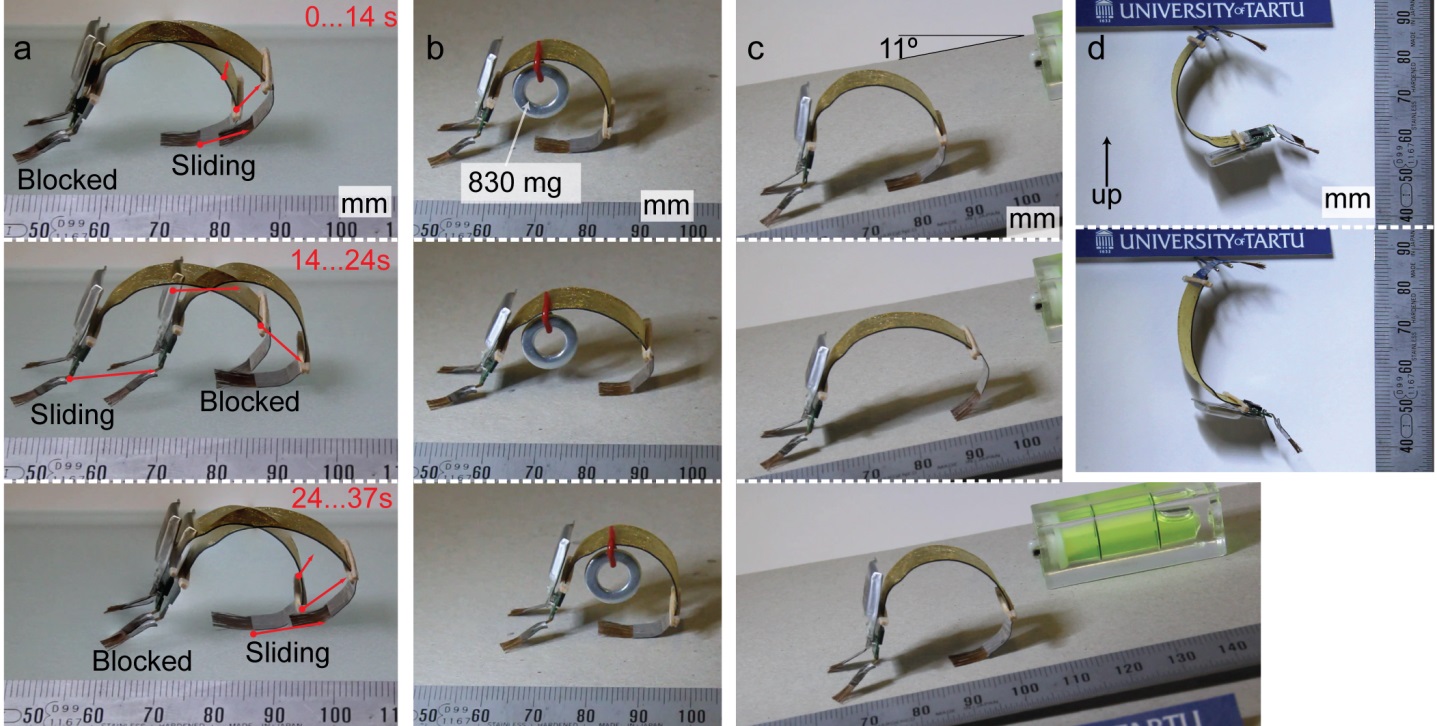


Figure 4. Locomotion of the IEAP robot. a) Composite images of the robot advancing on a satin glass plate. b) The IEAP robot carrying a payload of the weight equal to the robot itself. c) The IEAP robot climbing up on a cardboard inclined at 11°. d) The robot lifting itself up.

A video file containing the clips of all of the experiments described above is available as the **Supporting Information**.

## Pulse width modulation (PWM) control for IEAPs

PWM signals are often used to drive electrical devices of large mechanical inertia. The high electrical capacitance of the IEAP, being the counterpart of the mechanical inertia, makes PWM driving signals favorable for driving the IEAPs. The IEAP actuator is driven using a bipolar signal, consisting of the longer charging periods of alternating polarity using PWM, followed by shorter short-circuiting periods. The two outputs of the driver IC are switched between three states – either connected to the positive or negative LiPo terminal, or disconnected, having a high-impedance output. The two outputs from the driver are directly connected to the IEAP electrodes. The timing for one full working cycle is depicted in Figure 5a. The pulse-width-modulated (PWM) signal consists of voltage pulses of width Ta after every period TPWM. The duty cycle of the PWM signal is expressed as Ta/TPWM. During each charging period, the IEAP was driven using a PWM signal of a constant period and frequency.

In 10 s, the open-circuit voltage between the IEAP electrodes rose only to 1 V. An open-circuit voltage as low as 1 V was found to induce already a peak-to-peak bending deflection of 45 m-1 and 2.0 gf of blocking force between the opposite ends of a 40x10-mm actuator. (Two millimeters of the IEAP was rigidly clamped between the terminals; therefore, the free length was 38 mm.) The corresponding transient courses of free-bending deflection and blocking force are given in Figure 5b. The blocking force was measured at the tip of the actuator in perpendicular to its surface. Such an amount of deflection and force already sufficed to successfully drive the IEAP robot; therefore, the voltage was not increased further. An additional increase in the terminal voltage would have caused higher deflection and force at the cost of increased time per one working cycle. In Figure 5c it can be observed that the voltage on the IEAP electrodes alternates between the level of open-circuit voltage and the LiPo terminal voltage at the frequency of 32 kHz. The used input signal resulted in a stable operation of the actuator – no noticeable decrease in its performance was observed during hundreds of performed working cycles.



Figure 5. a) Control waveform for one working cycle of the robot. b) The transient courses for free-bending curvature and blocking force in one working cycle. c) Voltages at the opposite ends of the IEAP actuator during PWM input signal. d) Surface resistance of the IEAP with gold foil as current collectors.

## Current collectors

Certain types of IEAPs, such as IPMCs with chemically plated platinum electrodes, have surface resistance as high as the signal propagation along the electrode needs to be described as a finite transmission line.[38] The surface resistance is dependent on curvature, which can give feedback on its state,[39] but in turn complicates the control of this type of actuator.

A compact gold foil current collector has been proved to provide the lowest surface resistance without compromising the flexibility of the laminate. During fabrication, both sides of the IEAP actuator were covered with the current collector composed of three layers of gold foil. According to the recipe suggested by Akle et al [Akle, DOI: 10.1007/s10853-006-0632-4 ] the gold foils were glued, layer by layer, on top of the electrode using the Ion Power LQ-1115 15% Nafion solution in a mixture of water and alcohols. Nafion adhesive also acts as an elastic layer, precluding cracking of the gold layers.

It is essential, that the gold foil of nanoscale thickness is a continuous layer of metal; therefore its electric conductivity is excellent. Figure 5d demonstrates that three layers of 130-nm gold foils provide surface resistance that is only two times higher than the calculated theoretical resistance value for the gold foils. Due to its low thickness, the gold current collector was compliant and its resistance did not change considerably when bent.

Compliance of the electrodes is of vital importance in the field of the dielectric elastomer actuators as well.[40] As dielectric elastomers are driven with low current at high voltage, even the surface resistance in the MΩ range is already sufficient. In the case of the IEAP actuators, the surface resistance is more critical due to higher electric currents. In supercapacitors, thick solid aluminum current collector is commonly used.[41] This provides excellent conductivity, but is not compliant.

The gold layer is compliant and flexible due to its pre-buckled surface, which is achieved during the manufacturing process. Please see **Supporting Information** for details on the pre-buckling process. The newly-constructed IEAP actuator always has a uniform voltage across one electrode, despite of the fact that the terminal voltage is switched at frequency as high as 32 kHz. It is remarkable that the voltage difference is established immediately over the whole IEAP surface area, with no evidence of the transmission-line effect.

## Electromechanical performance of the material

Electromechanical performance of the constructed IEAP actuator was investigated using a sinusoidal input signal. In the free-bending mode, the curvature of the IEAP changed uniformly across the whole length of the laminate; therefore, it is possible to determine the IEAP curvature by fitting an arc over the image of the IEAP cross-section. **Figure 6**a depicts the cross-section of the IEAP actuator at different input voltages. The blocking force increased proportionally with the amplitude of sinusoidal input voltage, as given in Figure 6b, Instead, the free-bending curvature increased exponentially with the increased input voltage amplitude; also shown in Figure 6b.

The measurements show that the performance of the IEAP actuators is limited by the amount of charging current averaged in time, not considerably affected by the type of driving signal. Limiting of the time-averaged charging current is important for improving the IEAP lifetime, because the thin gold current collector layer could heat up as a result of high charging currents. Consequently, the speed and deflection rate of an IEAP actuator is limited by the amount of current passing through the material, eventually heating up the actuator. Figure 6c shows a more pronounced decrease between the bending curvature and blocking force at charging currents above 90 mA for a 4-cm2 IEAP. Thus, a time-averaged charging current of up to 25 mA cm-2 is optimal for driving this type of actuator.

Figure 6d gives the values for the calculated bending modulus in relation to the amount of electrical charge consumed by the IEAP in one working cycle. The bending modulus is calculated from the deflection and blocking force amplitude as given by Equation (1), where B is bending modulus, σ is stress, ε is strain, φK is the phase of curvature, and φQ is the phase of charge. φQ is calculated as , where φI is the phase of electric current.

|  |  |  |
| --- | --- | --- |
|  |  | (1) |

The calculation is explained in more detail in **Supporting Information**. It can be observed that the bending modulus B decreases linearly with increasing amount of consumed charge. Extrapolation gives the value of 334 MPa for bending modulus without applying electrical input. The difference in the bending modulus at an increased working voltage and current is, however, notable and should be considered in the design of applications based on the IEAP actuators.



Figure 6. The performance of the IEAP actuator in response to sinusoidal input signal. a) The composite image of the cross-sections of IEAP actuator at various input signal amplitudes. b) The strain difference and blocking force at different input voltage amplitudes. c) The decrease in the blocking force at high charging currents. d) The apparent bending modulus at different levels of consumed charge per one cycle.

## Energy-storage efficiency

The newly-constructed IEAP laminate is optimized for actuation; however, it still has a remarkable energy-storage property in air. Cyclic efficiencies for the supercapacitors are typically not given in the literature, because they are, by default, expected to operate completely without the presence of water. The absorbed water content is often considered as a taboo subject in the field of supercapacitors, as it can be detrimental to the cell performance especially at high voltages. However, the cronocoulonometry experiment results presented in **Figure 7** demonstrate a cyclic energy-storage efficiency of 93.5% in ambient air. (The corresponding ambient relative humidity was 10±4% during the experiments.) Therefore, the constructed IEAP material can potentially also used as an energy-storage medium. What is more, its areal capacitance, calculated from the cronocoulonometry data, is remarkably high – more than 151 mF cm-2. This is comparable to the highest capacitance-per-area values reported for micro-supercapacitors today.[42] The gravimetric capacitance of the 55-mg-cm-2 IEAP is 2.7 F g-1 and the volumetric capacitance of the 450-µm laminate is 3.4 F cm-3. Nevertheless, as it is also emphasized by Beidaghi *et al.* in their recent review paper,[42] the primary consideration for the development of micro-scale supercapacitors is their footprint per surface area, not their gravimetric capacitance.



Figure 7. Energy-storage efficiency of the IEAP actuator.

# Conclusion

This work presents the first IEAP robot that can autonomously operate in air. The constructed robot is very light (total weight 830 mg) and measures less than 4 cm. The results demonstrate the advantage of the IEAP technology especially in small-scale devices – the size of the robot was determined especially by the size of available power supplies, whereas the IEAP actuator itself could easily be used to make considerably smaller robots.

The IEAP actuator technology conforms well to the state-of-the-art microelectronics. The actuator and the driving electronics are both powered directly from a one-cell lithium-polymer battery. A cheap and widely-available PWM-based driver integrated circuit is successfully used in the control board for driving the actuator. In the constructed robot, the weight of electronic components is inconsiderable compared to its other construction elements, which opens possibilities for further miniaturization.

The robot is driven by a single IEAP actuator. The construction of the actuator is optimized for having simultaneously high bending modulus and high strain. At ±3V input voltage, a 3.8-cm piece of laminate generated blocking force as much as 36 mN (3.6 gf). In the free-bending mode, the laminate has an electrically-induced strain difference of 1.8% at 3 V. These levels of strain and force suffice for use in microrobotics. In fact, voltage as low as 1 V is already sufficient to successfully drive the robot.

The biomimetic design suggests the use of IEAP actuator in a non-planar configuration. In the manufacturing process the actuator was programmed having a U-shape. The programmed shape remained unchanged in the course of actuation.

The constructed IEAP actuator stands out by its remarkable efficiency. Although the IEAP actuators are very similar in construction to the supercapacitors, their energy-storage efficiency is usually low. The efficiency of the actuator was improved by the use of multiple layers of thin gold sheet as current collectors. The compliant gold current collector provided the necessary level of electronic conduction without compromising the flexibility of the laminate. An energy-storage efficiency of 93.5% was achieved.

The constructed inchworm-like robot has a top speed of 36 mm min-1, which makes it demonstrably slower than its biological example. However, it could rival a garden snail, which has a top speed of only 1 m h-1,[43] in a sprint contest.

# Experimental Section

*Fabrication of the actuator:* The separator was prepared from polyvinylidene fluoride co-hexafluoropropylene (PVdF-HFP) (Sigma–Aldrich Co.) and 1-ethyl-3-methylimidazolium trifluoromethanesulphonate (EMITFS) (≥99.0%, Fluka) by casting method. The ratio between PVdF-HFP:EMITFS was 1:1 in weight. The electrodes contained TiC–CDC powder (Skeleton Technologies OÜ), EMITFS, and PVdF-HFP in a ratio of 1:1:1. The electrodes were spray-painted on the membrane. Three layers of 130-nm gold sheet (Gold-Hammer) were glued on both sides of the laminate to form current collectors. 15 wt% Nafion solution (LIQUION® LQ-1115 1100EW, Ion Power, Inc.) was used as an adhesive for the gold foils. The non-planar initial shape was programmed by heating the appropriately fixed laminate up to 100ºC. The detailed manufacturing process is described in the **Supporting Information.**

*Electrical and electromechanical measurements:* The current consumption of the robot was measured by applying a 4-V input voltage from an external laboratory power supply. The total current consumed by both actuator and control board was measured. The terminal voltages were registered using National Instruments PCI-6036E and 6XXX DAQ devices. Tektronix TDS 2024B oscilloscope was used in the measurements in Figure 5c.

Blocking force was measured using an ADInstruments MLT0202 load cell connected to the National Instruments PCI-6036E DAQ device. The actuation was recorded by a DMK 22BUC03 USB camera equipped with a long-focus lens C5028-M. The curvature amplitude was determined by fitting the recorded image of the IEAP with a circle of variable radius using National Instruments LabView software package. The amplitudes and phases corresponding to a sinusoidal input were determined by fitting the measured data using a differential evolution algorithm implemented in LabView. The sheet resistance was measured using an Agilent 34420A micro-ohm meter and a custom-made bending rig described previously by Must et al.[44]

The chronocoulonometry experiment was performed on a PARSTAT 2273 potentiostat in two-electrode mode.

The scanning electron micrographs were obtained using a Hitachi TM3000 microscope.

**Supporting Information**

Supporting Information is available from the Wiley Online Library.

* Details on the manufacturing process for the IEAP
* Construction details for the constructed micro-robot
* Calculation for the apparent bending stiffness
* A video demonstration for the microrobot

Acknowledgements

This work was supported by the Estonian Science Foundation grant no. 8553, by targeted financing SF0180008s08 from the Estonian Ministry of Education, by Institutional Research Funding project IUT20-24 from Estonian Research Council, and by development project no. SLOTI12156T by Archimedes Foundation. The authors acknowledge support from national scholarship program Kristjan Jaak, which is funded and managed by Archimedes Foundation in collaboration with the Ministry of Education and Research; the Estonian Doctoral School in Information and Communication Technology, the graduate school ‘Functional materials and technologies’, and the Tiger University Program of the Information Technology Foundation for Education.

Received: ((will be filled in by the editorial staff))  
Revised: ((will be filled in by the editorial staff))  
Published online: ((will be filled in by the editorial staff))

References

[1] Y. Li, R. Torah, S. Beeby, J. Tudor, "An all-inkjet printed flexible capacitor for wearable applications", presented at Design, Test, Integration and Packaging of MEMS/MOEMS (DTIP), 2012 Symposium on2012.

[2] D. Wei, T. W. Ng, *, Electrochemistry Communications.* **2009***, 11*(10), 1996-1999.

[3] J. Bae, M. K. Song, Y. J. Park, J. M. Kim, M. Liu, Z. L. Wang, *, Angewandte Chemie International Edition.* **2011***, 50*(7), 1683-1687.

[4] H. Gwon, H. S. Kim, K. U. Lee, D. H. Seo, Y. C. Park, Y. S. Lee, B. T. Ahn, K. Kang, *, Energy & Environmental Science.* **2011***, 4*(4), 1277-1283.

[5] L. Nyholm, G. Nyström, A. Mihranyan, M. Strømme, *, Adv Mater.* **2011***, 23*(33), 3751-3769.

[6] F. Kaasik, T. Tamm, M. Hantel, E. Perre, A. Aabloo, E. Lust, M. Bazant, V. Presser, *, Electrochemistry Communications.* **2013**.

[7] R. Kötz, P. Ruch, D. Cericola, *, J. Power Sources.* **2010***, 195*(3), 923-928.

[8] B. J. Akle, M. D. Bennett, D. J. Leo, *, Sensors and Actuators A: Physical.* **2006***, 126*(1), 173-181.

[9] K. J. Kim, S. Tadokoro, *Electroactive Polymers for Robotic Applications: Artificial Muscles and Sensors*, Springer**2007**.

[10] P. Cottinet, C. Souders, S. Tsai, R. Liang, B. Wang, C. Zhang, *, Physics Letters A.* **2012***, 376*(12), 1132-1136.

[11] M. Ghaffari, W. Kinsman, Y. Zhou, S. Murali, Q. Burlingame, M. Lin, R. Ruoff, Q. Zhang, *, Adv Mater.* **2013**.

[12] R. Temmer, A. Maziz, C. Plesse, A. Aabloo, F. Vidal, T. Tamm, *, Smart Mater. Struct.* **2013***, 22*(10), 104006.

[13] J. Torop, T. Sugino, K. Asaka, A. Jänes, E. Lust, A. Aabloo, *, Sensors Actuators B: Chem.* **2012***, 161*(1), 629-634.

[14] E. W. Jager, E. Smela, O. Inganäs, *, Science.* **2000***, 290*(5496), 1540-1545.

[15] K. Aw, L. Fu, A. McDaid, *, International Journal of Smart and Nano Materials.* **2013**(ahead-of-print), 1-11.

[16] J. Hou, M. Luo, T. Mei, "The design and control of amoeba-like robot", presented at Computer Application and System Modeling (ICCASM), 2010 International Conference on2010.

[17] A. A. Villanueva, K. J. Marut, T. Michael, S. Priya, *, Bioinspiration & biomimetics.* **2013***, 8*(4), 046005.

[18] S. W. Kwok, S. A. Morin, B. Mosadegh, J. So, R. F. Shepherd, R. V. Martinez, B. Smith, F. C. Simeone, A. A. Stokes, G. M. Whitesides, *, Advanced Functional Materials.* **2013**.

[19] S. McGovern, G. Alici, V. Truong, G. Spinks, *, Smart Mater. Struct.* **2009***, 18*(9), 095009.

[20] X. Tan, D. Kim, N. Usher, D. Laboy, J. Jackson, A. Kapetanovic, J. Rapai, B. Sabadus, X. Zhou, "An autonomous robotic fish for mobile sensing", presented at Intelligent Robots and Systems, 2006 IEEE/RSJ International Conference on2006.

[21] Y. Ma, Y. Zhang, B. Wu, W. Sun, Z. Li, J. Sun, *, Angewandte Chemie.* **2011***, 123*(28), 6378-6381.

[22] S. Lee, J. H. Prosser, P. K. Purohit, D. Lee, *, ACS Macro Letters.* **2013***, 2*, 960-965.

[23] H. Cheng, J. Liu, Y. Zhao, C. Hu, Z. Zhang, N. Chen, L. Jiang, L. Qu, *, Angewandte Chemie International Edition.* **2013***, 52*(40), 10482-10486.

[24] S. Maeda, Y. Hara, T. Sakai, R. Yoshida, S. Hashimoto, *, Adv Mater.* **2007***, 19*(21), 3480-3484.

[25] F. Ilievski, A. D. Mazzeo, R. F. Shepherd, X. Chen, G. M. Whitesides, *, Angewandte Chemie.* **2011***, 123*(8), 1930-1935.

[26] S. A. Morin, R. F. Shepherd, S. W. Kwok, A. A. Stokes, A. Nemiroski, G. M. Whitesides, *, Science.* **2012***, 337*(6096), 828-832.

[27] B. Mosadegh, P. Polygerinos, C. Keplinger, S. Wennstedt, R. F. Shepherd, U. Gupta, J. Shim, K. Bertoldi, C. J. Walsh, G. M. Whitesides, *, Advanced Functional Materials.* **2013**.

[28] P. Brochu, Q. Pei, *, Macromolecular rapid communications.* **2010***, 31*(1), 10-36.

[29] A. Punning, M. Anton, M. Kruusmaa, A. Aabloo, "A biologically inspired ray-like underwater robot with electroactive polymer pectoral fins", presented at International IEEE Conference on Mechatronics and Robotics2004.

[30] J. Najem, S. A. Sarles, B. Akle, D. J. Leo, *, Smart Mater. Struct.* **2012***, 21*(9), 094026.

[31] S. Sareh, J. Rossiter, *, Smart Mater. Struct.* **2013***, 22*(1), 014004.

[32] S. Guo, L. Shi, K. Asaka, "IPMC actuator-sensor based a biomimetic underwater microrobot with 8 Legs", presented at Automation and Logistics, 2008. ICAL 2008. IEEE International Conference on2008.

[33] N. Kamamichi, M. Yamakita, K. Asaka, Z. Luo, "A snake-like swimming robot using IPMC actuator/sensor", presented at Robotics and Automation, 2006. ICRA 2006. Proceedings 2006 IEEE International Conference on2006.

[34] V. Palmre, J. J. Hubbard, M. Fleming, D. Pugal, S. Kim, K. J. Kim, K. K. Leang, *, Smart Mater. Struct.* **2013***, 22*(1), 014003.

[35] P. Arena, C. Bonomo, L. Fortuna, M. Frasca, S. Graziani, *, Systems, Man, and Cybernetics, Part B: Cybernetics, IEEE Transactions on.* **2006***, 36*(5), 1044-1052.

[36] A. Firouzeh, M. Ozmaeian, A. Alasty, *, Smart Mater. Struct.* **2012***, 21*(6), 065011.

[37] U. Kosidlo, M. Omastová, M. Micusík, G. Ćirić-Marjanović, H. Randriamahazaka, T. Wallmersperger, A. Aabloo, I. Kolaric, T. Bauernhansl, *, Smart Mater. Struct.* **2013***, 22*(10), 104022.

[38] A. Punning, M. Kruusmaa, A. Aabloo, *, Sensors and Actuators A: Physical.* **2007***, 133*(1), 200-209.

[39] K. Kruusamäe, P. Brunetto, A. Punning, M. Kodu, R. Jaaniso, S. Graziani, L. Fortuna, A. Aabloo, *, Smart Mater. Struct.* **2011***, 20*(12), 124001.

[40] S. Rosset, H. R. Shea, *, Applied Physics A.* **2013***, 110*(2), 281-307.

[41] P. Aza'is, *, Supercapacitors: Materials, Systems and Applications.* **2013**.

[42] M. Beidaghi, Y. Gogotsi, *, Energy & Environmental Science.* **2013**.

[43] Anonymous *Study of 450 garden snails, Cornu aspersum, in a typical rough lawn habitat*, University of Exeter**2013**.

[44] I. Must, F. Kaasik, I. Põldsalu, U. Johanson, A. Punning, A. Aabloo, *, Carbon.* **2012***, 50*(2), 535-541.

[45] B. J. Akle, M. D. Bennett, D. J. Leo, K. B. Wiles, J. E. McGrath, *, J. Mater. Sci.* **2007***, 42*(16), 7031-7041.

[46] V. Palmre, E. Lust, A. Janes, M. Koel, A. Peikolainen, J. Torop, U. Johanson, A. Aabloo, *, J.Mater.Chem.* **2011**.

**The first autonomous micro-robot that advances using a shape-morphing flexible supercapacitor** is demonstrated. The flexible supercapacitor with carbide-derived carbon electrodes and ionic liquid electrolyte is operable in ambient air and morphs the whole shape of the sub-gram scale inchworm-like robot when being charged. The capacitive actuator stands out for its exclusive combination of high electrically-induced strain and high load-bearing capacity.

**Actuators, Biomimetics, Composite materials, Polymeric materials, Stimuli-responsive materials**

Indrek Must\*, Friedrich Kaasik, Inga Põldsalu, Lauri Mihkels, Urmas Johanson, Andres Punning, and Alvo Aabloo

An autonomous inchworm-like robot propelled by an actuator akin to a supercapacitor



Copyright WILEY-VCH Verlag GmbH & Co. KGaA, 69469 Weinheim, Germany, 2013.

Supporting Information

An autonomous inchworm-like robot propelled by an actuator akin to a supercapacitor

Indrek Must\*, Friedrich Kaasik, Inga Põldsalu, Lauri Mihkels, Urmas Johanson, Andres Punning, and Alvo Aabloo

Table of Contents for the Supporting Information

S1. Manufacture of the IEAP

S2. Design of the robot

S2.1 Control board

S2.2 Power supply

S2.3 Bristles

S2.4 Clamping and terminals

S3. Calculation of apparent bending stiffness

S4. Video demonstration of the performance of the micro-robot.

## Manufacture of the IEAP

The fabrication process of the IEAP laminate involved the following steps:

* Casting of the membrane
* Spray-painting of the electrodes on the previously cast membrane
* Gluing of the gold foils on top of the electrodes
* Shape-programming

The membrane was manufactured by solvent-casting method. First, a mixture of 1-ethyl-3-methylimidazolium trifluoromethanesulphonate (EMITFS) (Fluka, ≥99.0 %), poly(vinylidene fluoride-co-hexafluoropropene) (PVdF-HFP), 4-methyl-2-pentanone (MP) (Sigma-Aldrich), and propylene carbonate (PC) (Sigma-Aldrich) was prepared. The weight ratio for EMITFS:PVdF was 50:50% and for MP:PC it was 24/76%. The solution was cast into a PTFE mold (Figure S 1a). The solution gelated in about 5 minutes and it was further dried in ambient air for 12 h.

The electrode dispersion was prepared by mixing 30 wt% of TiC-CDC powder (Skeleton Technologies), 35 wt% of PVDF-HFP, and 35 wt% of EMITFS. The PVdF-HFP pellets were dissolved in N,N-dimethylacetamide (DMAc) (Fluka). An ultrasonic probe (UP200S, Hielscher) was used to promote homogenization.

The dispersion was painted layer-by-layer directly on both sides of the previously cast PVdF-IL membrane using an airbrush (Figure S 1b). Spray-application of the electrodes, also known as the ‘direct assembly process’ (DAP), has previously been used by Akle et al.[45] and Palmre et al.[46] Approximately 20 electrode layers were applied on both sides of the membrane. The volatile solvents were evaporated after spraying of each successive layer using a flow of warm air.

The laminate was subsequently fixed on the outer surface of a cylindrical tube with a diameter of 10 mm. Three layers of ~130 nm gold foil (Gold-Hammer) were placed on top of the outer electrode (Figure S 1c). A drop of 15 wt% Nafion solution in ethanol and water (LIQUION® LQ-1115 1100EW; Ion Power, Inc) was used as an adhesive. Then the laminate was turned around on the surface. By doing so, the gold sheets were pre-buckled due to compressive force. Then Another 3 layers of gold were subsequently attached on top of the opposite (stretched out) electrode (Figure S 1d).

The shape-programming was performed as follows. The IEAP was fixed on the outer surface of a cylindrical tube with an appropriate diameter. It is preferred that the shape-programming tube is larger in diameter than the one used in gold foil attachment. A tube 30 mm in diameter was selected for this purpose. The tube with the laminate on its surface was heated using a hot air gun to just below the melting point of the polymeric membrane of the IEAP (approximately 100°C) (Figure S 1e). Then the IEAP was removed from the surface of the tube. As the final step of fabrication, the laminate was held overnight under vacuum to remove any solvents remaining.

After manufacture, the IEAP retained its newly-programmed initial curvature radius of 15 mm during all performed experiments. The thickness of the finished IEAP laminate was 450 µm.



Figure S 1. Manufacturing process for the IEAP. a) Casting of the membrane; b) spray-application of the electrodes; c) gluing of the current-collector to one side of the IEAP; d) attachment of the current collector to the opposite side of the; and e) IEAP shape-programming.

## Design of the robot

### Control board

An ATTINY13A microprocessor by Atmel Corporation in a 10-pin 3×3×1.0 mm MLF package (weight: 24 mg) was selected as the control unit. The two PWM outputs of the microcontroller were directly connected to a DRV8837 H-bridge by Texas Instruments in a 2×2×0.8-mm WSON-8 package (weight: 9 mg) for current amplification. A 10-F ceramic multilayer capacitor in a 1×0.5×0.5-mm 0402 package (weight: 3 mg) was used for levelling of the supply voltage. The outputs of the H-bridge were connected directly to the gold terminals for connecting to the IEAP. The 0.4-mm double-sided printed circuit board (8×12 mm; 97 mg) was manufactured by ITEAD Intelligent Systems Co.Ltd.

The program code used in the microprocessor and the circuit board layout is available on request from the Authors.

### Power supply

A 10 mAh 10C LiPo battery cell (microflierradio.com) was chosen as a power supply. Small cylindrical neodymium magnets (0.5×1 mm; 3 mg each) were attached to the battery and the control board, so that the battery could be easily attached and detached. The magnets also prevented from connecting the battery with the opposite polarity. The magnets were first glued on the soldering pad of the circuit board and the LiPo terminals using a drop of silver-based conductive glue (Electon 40AC; Amepox Microelectronics) between the magnet and the substrate. Then a drop of non-conductive epoxy adhesive was used to support the magnets from their sides.

### Bristles

Directional locomotion was achieved by the use of miniature bristles placed under the robot and directed to an appropriate direction. The robot touches the ground only via the bristles at all times. By placing the robot on a surface with an appropriate roughness, the bristles act as a brake, blocking the sliding motion in one direction. The miniature bristles were prepared by lamination of suitably-chosen fibers between polyethylene terephthalate-aluminum laminate foil.

### Clamping and terminals

The corrosive nature of the ionic liquid electrolyte demands for terminal materials of high electrochemical stability. For this reason, the terminals were made of solid gold sheet. Appropriate contact pressure was achieved by clamps made of balsa wood.

## Calculation of apparent bending stiffness



Figure S 2. The transient courses for a) electric current and b) blocking force and bending curvature at a sinusoidal input voltage at the frequency of 16 mHz. c) The phase shifts between electric charge, curvature, and blocking force result in the change in the apparent bending modulus.

Figure S 2a gives a typical transient course for electric current in response to sinusoidal input voltage. At voltage amplitude of 3 V, the electric current increased up to 130 mA. Figure S 2b gives the transient courses of blocking force and free-bending curvature measured in the equal driving conditions. A phase lag of 30º between curvature and force can be observed. In a linear-elastic system, such phase shift is not expected. If the IEAP is constrained from bending during the measurement of blocking force, the laminate can be looked as a spring with its initial curvature changed by an electrical input signal.

Figure S 2c gives phase differences for curvature and force in relation to input charge. As the electric charge is an integral of the input current, the phase of charge, φQ, was determined by shifting the phase of electric current by 90º. At 3 V input voltage, the phase of force leads charge (expressed as φF-φQ) by 42º, while the phase of blocking force (expressed as φK-φQ) leads only by 12º.

It is expected that the neutral plane (a plane with zero strain/stress in bending) is located in the middle of the IEAP. Therefore, the maximum linear strain, ε, can be given as

|  |  |  |
| --- | --- | --- |
|  |  | (2) |

where t is thickness of the IEAP laminate and k is curvature. The values for linear strain are given in Figure 6a. The bending moment can be calculated as

|  |  |  |
| --- | --- | --- |
|  |  | (3) |

where M is bending moment, l is length of the actuator, and F is blocking force. Therefore, in case of a linear-elastic material, it is possible to calculate the stress, σ, as

|  |  |  |
| --- | --- | --- |
|  |  | (4) |

where I is area moment of inertia for a beam of rectangular cross-section. I can be calculated for a beam of length l as

|  |  |  |
| --- | --- | --- |
|  |  | (5) |

Knowing the stress and strain, it is possible to calculate the bending modulus, B, of the laminate. However, the phase difference between curvature (*i.e.* strain) and blocking force (*i.e.* stress) – φK-φF – must be taken into account as follows:

|  |  |  |
| --- | --- | --- |
|  |  | (6) |

Therefore, from the formulae above, B can be calculated for each input signal as

|  |  |  |
| --- | --- | --- |
|  |  | (7) |

The calculated values for B are given in Figure S 2c.