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~~Autonomous robotic inchworm with an open-air supercapacitor artificial muscle~~.

Autonomous robotic inchworm with open-air supercapacitor artificial muscles.

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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 830-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 electrodes, based on activated carbon, stands out by its exclusive combination of high electrically-induced strain and high bending modulus. On the other hand it is characterized by outstanding electric capacitance per surface area and high cyclic efficiency. These properties make the laminate one of the few IEAPs meeting concurrently the requirements of miniature robotics as well as of the energy storage systems.

**Kiri editorile. Praegu veel poolik.**

Dear Editor,

Hereby we submit a manuscript „Autonomous robotic inchworm with open-air supercapacitor artificial muscles.“ for a consideration to be published in the „Advanced Materials“ journal.

In this paper we report on a miniature robot, propelled by an ionic electroactive polymer (IEAP) actuator. Its most important feature is the capability to run in open air, carrying with him its own power supply. To the best of our knowledge this the absolute first autonomous mobile terrestrial propelled by electroactive polymeractuator of any type. The ... is vividly expressed in the video, submitted as the **Supporting Information.**

The high strain and high elastic modulus IEAP actuator was developed purposefully for the robotic applications. The described inchworm robot is just the first example, while the next, more complicated robotic applications are currently being prepared. Observing the parameters of the developed actuator in detail, we found that our IEAP material is of equal value with the best contemporary open-air supercapacitors in terms of the capacitance per area as well as in terms of the cyclic energy storage efficiency. Hence the intriguing title of the paper.

The IEAP actuators have been considered being a promising propulsion system for the miniature robotic applicationsfor about two decades already. The vast majority of the state-of-the-art IEAP-based systems run with external power supply and trailing cables, The few exceptional designs are robotic fishes, where the battery is located in a boat, propelled by IEAP fins. The major reason is the too low development level of the IEAP materials (low elastic modulus, low generated thrust, low efficieny, etc.) Nevertheless many research groups all over the world are attempting to design any mobile apparatus propelled by any electroactive polymer actuator, but with the onboard power supply. We are convinced that with our IEAP artificial muscle we have won this race.

Our newborn device is a giant leap in the development of the IEAP materials, demonstrating that indeed it is possible to .

Stationary

The

# 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 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. Unfortunately the energy density of the hydrous supercapacitors is decreased due to the low electrochemical window of the aqueous electrolytes and the influence of the environmental factors, such as ambient humidity, could no longer be ignored.

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) actuator.

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 similar 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. 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 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 the battery.

In the current paper we report on the world’s first IEAP robot, capable to run in open air, carrying with him its own power supply. For this application we developed an IEAP actuator, where high strain and high elastic modulus are combined with high capacitance within a single laminate. Its actuation performance is achieved by appropriately tuned manufacturing process and use of the carbide-derived carbon as active electrode material, while the 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 a 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 830 mg with the on-board battery, this is the lightest autonomous electroactive polymer-based robot developed up to the present moment.



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

# Towards biomimetic and autonomous robots

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 the 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 the 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 with the whole concept of the autonomy of the robots. A significant shortfall of the dielectric elastomer or piezoelectric actuators is their high working voltage - typically from a few hundreds volts to 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 IEAP type. The IMPC actuators have found widespread deployment in biomimetic robotics. As the conventional 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, evaporation of the solvent limits the lifetime of these proof-of-concept robots in air to a few minutes only.

# Design of the biomimetic robot

The robot is inspired by inchworm and produces a rectilinear motion by manipulating its body – IEAP actuator – in a plane with a bending-relaxation gait with only one bending degree of freedom implemented. The robot’s front end and rear end support to the ground through bristles, providing anisotropic friction and preventing bidirectional locomotion. The net forward motion is produced by alternate bending and relaxation of the IEAP actuator in a periodic step manner. The gait of the robot consists of two phases. During the bending phase the anterior bristles prevent the front end from sliding back, while the bending IEAP actuator drags the skidding back end closer to the front end. Once reached to its maximal bending, the relaxing actuator supports to the posterior bristles and pushes the front end forward. This process is periodically repeated.

## IEAP actuator

The requirements to an actuator for a biomimetic microrobot are rigorous: the ability to generate high electrically induced strains, the ability to support the weight of the robot, the exceptionally high electronic conductivity of the electrodes, and absence of creep. An actuator integrating all of the listed high standards was manufactured by combination of casting, spray-painting, lamination, and thermal pre-shaping techniques. The key factors in the development of this conspicuous actuator are the precise proportion between the electrode constituents, the ratio between the thicknesses of the membrane and the electrodes, and the initially arched shape.

Voltage, applied between the electrodes of a flat IEAP actuator makes the opposite electrodes expand or contract in all directions. The resulting bowl-shaped buckling of the laminate impedes its bending in both lateral directions while the favorable direction is towards the narrower edge. In order to reduce the adverse effects of the unfavorable transverse buckling of the actuator in the constructed IEAP robot, we have taken particular effort to arcuate the actuator.

The choice of materials and manufacturing techniques for the IEAP actuator are explained as follows. The cast membrane consists of PVdF-HFP, highly swollen in the EMITFS ionic liquid. The cations and anions of EMITFS can easily migrate in the fluoropolymer network. The thickness of the cast membrane is approximately 120 µm. The electrodes consist of EMITFS ionic liquid, carbon powder and the PVdF-HFP binder to form compact layers. From the family of the nanoporous carbide-derived carbons[Presser] the one, derived from Titanium Carbide (TiC-CDC) at the temperature of 800ºC [37] showed the most outstanding results.[Vil\_Carbon\_2011]

The electrodes are formed by spray-painting layer-by-layer the mixture of TiC-CDC, PVdF(HFP), EMITFS, and the appropriate solvent directly on the membrane. After spraying of each successive layer, the volatile solvents were evaporated using modest flow of warm air. As a result the individual grains of CDC are tightly wrapped together by the fluoropolymer threads. A total of 20 layers were painted on the both sides of the membrane to attain the the thickness of the electrodes of 160-170 µm. The gained total thickness of the symmetrical laminate is 450 µm. The exceptionally high electronic conductivity of the electrodes is achieved by gluing three layers of 130-nm gold foils on both sides of the laminate. The final step in the manufacturing process is pre-shaping of the actuator to an arched shape by heat-treating at 100 ºC , using a cylindrical tube as a jig.

The scanning electron microscope micrograph of the cross-section of the IEAP is presented in **Figure 2**, while the detailed manufacturing process is given in the **Supporting Information.**



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

## Construction of the robot

The largest part of the robot is the arched 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 the bending-relaxation gait, an appropriate control waveform is generated by an on-board microcontroller, while the driving electric current is boosted by an H-bridge integrated circuit (IC). A lithium-polymer (LiPo) rechargeable battery was chosen as the on-board power supply. The constituents of the robot and the weight proportions of the components are depicted in **Figure 3a** and **Figure 3b** respectively. 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 nearly a quarter of the total mass.

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

## The robot’s locomotion characteristics

In order to ensure the anisotropic friction of the bristles, the surfacing of the ground must be smooth, but of necessary roughness – the 600 Grit sandpaper, cardboard and satin glass provided the best results**. Figure 4**a depicts sequential morphing of the shape of the IEAP robot, moving on a satin glass. The robot was driven with a pulse-width modulated (PWM) voltage signal with the fixed PWM duty cycle of 23.5%. In this regime the used 10-mAh LiPo battery can power the robot for approximately 8 minutes. During a 26 s gait the robot advanced for 16 mm.

The next experiments demonstrate that the weight of the battery and the rest of the components are not at the upper limit of the actuator’s muscular capabilities. As shown in Figure 4b, the IEAP actuator is rigid enough to carry a payload, roughly equal to the weight of the robot itself, including the battery. The 830 mg robot, carrying a 890 mg payload, advanced 14 mm in each gait. In Figure 4c, the robot climbs up with an angle of climb of 11º, advancing 14 mm in each gait.

In Figure 4d, the hanging IEAP actuator is lifting up the LiPo battery and the controller board. This experiment can be used to estimate the efficiency of the actuator. With non-symmetric bending-relaxation cycle it was able to lift the **550** mg weight for **10-12** mm as much as **28** times until total discharge of the battery. Even more important is the fact that no creep of the actuator was observed even under unidirectional load.

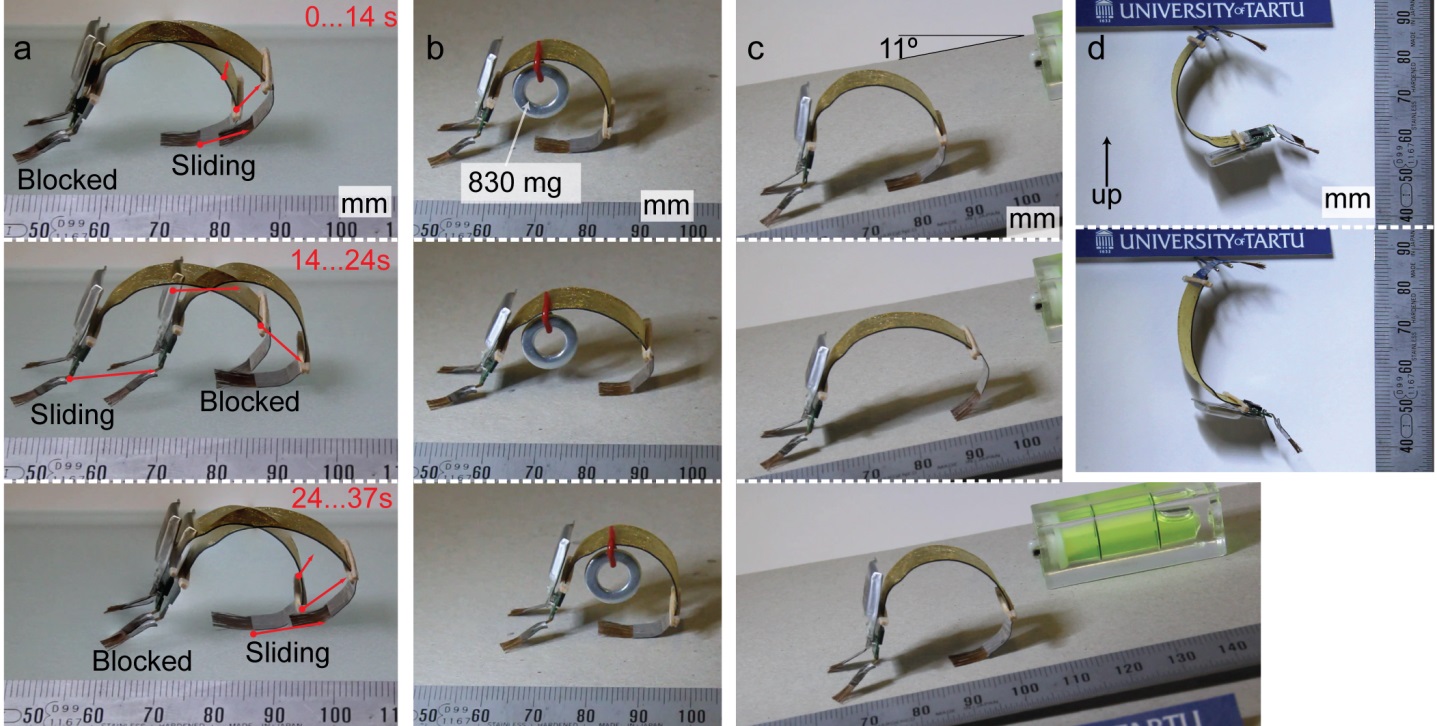


Figure 4. Locomotion of the IEAP robot. a) Overlay images of the robot, advancing on a satin glass plate. b) The robot carrying a payload of the weight equal to the robot itself. c) The robot climbing up on a cardboard inclined at 11°. d) The robot lifting itself up.

All described experiments are more vividly expressed in the video, available at the **Supporting Information** at the Wiley Online Library.

## Pulse-width modulation (PWM) control

PWM signals are often used to drive electrical devices of large mechanical inertia. The high electrical capacitance of the IEAP, being the counterpart for the mechanical inertia, makes PWM driving signals favorable for driving the IEAPs. The IEAP actuator is driven using a bipolar signal, consisting of longer charging periods of alternating polarity using PWM, followed by shorter short-circuiting periods. The two outputs of the driver IC, directly connected to the opposite electrodes of the IEAP actuator, are switched between three states – they are either connected to the positive or negative LiPo terminal, or disconnected, having a high-impedance output. The timing for one full gait 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 constant period and frequency.

In 10 s, the open-circuit voltage between the IEAP electrodes rises to 1 V only. This is enough to induce the 45 m-1 peak-to-peak bending deflection or 2.0 gf blocking force between the opposite ends of a 40x10-mm actuator. (two millimeters of the IEAP strip was rigidly clamped between the terminals; therefore, the free (operational) 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, perpendicular to its surface. This amount of deflection and force is already sufficient to drive the IEAP robot; therefore, the voltage was not increased further. Further increase of the terminal voltage will cause higher deflection and force, yet with the cost of increased time per gait. As depicted in Figure 5c, the voltage on the IEAP terminals is switched at the PWM sampling frequency - 32 kHz - between the the IEAP open-circuit voltage and the positive or negative polarity voltages of the power supply.

The used input signal resulted with stable operation of the actuator – no noticeable decrease of 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

The conductivity and compliance of the electrodes are of vital importance in the fields of IEAPs, supercapacitors, as well as dielectric elastomer actuators.[40] As the dielectric elastomers are driven with a low current at a high voltage, even the surface resistance in the mega-ohm range is already satisfying. In the case of the IEAP actuators, the surface resistance is more critical due to higher electric currents. In supercapacitors, the commonly used thick solid aluminum current collector provides excellent conductivity, but is not compliant.[41] The electrode conductivity of some certain types of IEAPs, e.g. IPMCs with chemically plated platinum electrodes, is so low that the signal propagation along the electrode can be described as a finite transmission line.[38] Moreover, the conductivity of electrode is dependent on its curvature, which in one hand can give feedback on its state,[39] but in other hand definitely complicates the control of this type of actuator. newly constructed

In order to improve the conductivity of the carbonaceous electrodes of the IEAP actuator fabricated for our IEAP robot, additional thin layers of gold were added on top of both sides of the laminate. A compact gold foil 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 130-nm 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. The Nafion adhesive also acts as an elastic softening layer, precluding the gold layers from cracking.

It is essential that a gold foil of nanoscale thickness is a continuous layer of metal with an excellent electric conductivity. **Figure 5d** demonstrates that the resistance, provided by three layers of 130-nm gold foils, is only twice higher than the calculated theoretical resistance value for the uniform and flawless layer of gold of equal thickness. The gold layer is compliant and flexible, and its resistance does not change considerably when bent due to its low thickness and convoluted surface, achieved during the manufacturing process. The convoluting process is described in detail in the **Supporting Information**.

## Electromechanical performance of the IEAP material

The electromechanical performance of the constructed IEAP actuator was investigated using a sinusoidal input signal. In the free-bending mode, the curvature of the IEAP changes 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 solely by the amount of the charging current averaged in time, and is not considerably affected by the form of driving signal. Limitation of the time-averaged charging current is important for another reason - for improving the lifetime of the IEAP. Resistance of the thin gold current collector layer still holds certain real value and can warm up due to the high charging currents, eventually heating up the whole actuator. On the basis of the 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. **Ma ei saa 6c-st aru!**.

Figure 6d gives the relation between the calculated bending modulus and the amount of electrical charge consumed by the IEAP in one working cycle. The bending modulus is calculated according to 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. This calculation is explained in more detail in **Supporting Information**.

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

The bending modulus B decreases linearly with increasing amount of consumed charge. Extrapolation gives the value of 334 MPa for bending modulus with no electrical input applied.



Figure 6. The performance of the IEAP actuator in response to sinusoidal input signal. a) The overlay images 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

Although this novel IEAP laminate is optimized for actuation in the ambient environment, it still has remarkable energy storage properties. The commercially available supercapacitors are sealed devices of anhydrous environment; therefore their cyclic efficiency is close to unity. 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 higher voltages. However, the chronocoulometry results presented in **Figure 7** demonstrate the exemplary cyclic energy storage efficiency (94%) of our IEAP actuator in air. Due to its high energy storage efficiency value, this IEAP material has a great potential to be an energy storage medium. Its capacitance per area, calculated from the chronocoulometry data, is remarkably high – over 150 mF cm-2. In terms of the capacitance per area our IEAP material is on a par with the contemporary micro-supercapacitors.[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 thick laminate is 3.4 F cm-3. Nevertheless, as emphasized by Beidaghi *et al.*, the primary consideration for the development of micro-scale supercapacitors is their footprint per surface area, rather than the gravimetric capacitance. [42]



Figure . Energy-storage efficiency of the IEAP actuator.

# Conclusion

Kokkuvõte, abstract, sissejuhatus, ja kiri editorile tuleb teha ühtses stiilis – sellises nagu me Urmasega just arutasime. Põhirõhk on materjalil, mis juhuslikult kõlbas muuhulgas roboti jaoks.

This work presents the mobile terrestrial IEAP robot, powered by an onboard battery that can autonomously operate in air . The constructed robot is very light (total weight 830 mg) and measures less than 4 cm. The constructed inchworm-like robot has a top speed of 2.2 m h-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 possible occasion of a sprint contest.

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. The robot is driven by a single IEAP actuator. The biomimetic design suggests the use of the IEAP actuator in a non-planar configuration. In the manufacturing process the actuator was programmed having a U-shape.

The constructed IEAP is optimized for having simultaneously high bending modulus and high strain. At ±3 V 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 was already sufficient to successfully drive the robot.

The constructed IEAP actuator stands out by its remarkable efficiency. A compliant gold current collector provided the necessary level of electronic conduction without compromising the flexibility of the laminate. An energy-storage efficiency of 94% as a supercapacitor was achieved.

# 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 current consumption of the sole IEAP was determined by subtracting the current consumption of the system without IEAP attached. 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 lens of long focal length 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 the 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 a two-electrode mode.

The scanning electron micrographs were obtained using a Hitachi TM3000 microscope at 15-kV acceleration voltage with back-scattered electron detector.

**Supporting Information**

Supporting Information is available from the Wiley Online Library.

* S1: Details on the manufacturing process for the IEAP
* S2.1-S2.4: Construction details for the constructed micro-robot
* S3: Calculation for the apparent bending stiffness
* S4: A video demonstration for the microrobot

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**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



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Supporting Information

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## 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 . 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 . 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.