Artificial Muscles Powered by Glucose

Fariba Mashayekhi Mazar, Jose G. Martinez, Manav Tyagi, Mahdi Alijanianzadeh, Anthony P. F. Turner, and Edwin W. H. Jager*

Untethered actuation is important for robotic devices to achieve autonomous motion, which is typically enabled by using batteries. Using enzymes to provide the required electrical charge is particularly interesting as it will enable direct harvesting of fuel components from a surrounding fluid. Here, a soft artificial muscle is presented, which uses the biofuel glucose in the presence of oxygen. Glucose oxidase and laccase enzymes integrated in the actuator catalytically convert glucose and oxygen into electrical power that in turn is converted into movement by the electroactive polymer polypyrrole causing the actuator to bend. The integrated bioelectrode pair shows a maximum open-circuit voltage of 0.70 ± 0.04 V at room temperature and a maximum power density of 0.27 μW cm⁻² at 0.50 V, sufficient to drive an external polypyrrole-based trilayer artificial muscle. Next, the enzymes are fully integrated into the artificial muscle, resulting in an autonomously powered actuator that can bend reversibly in both directions driven by glucose and O₂ only. This autonomously powered artificial muscle can be of great interest for soft (micro-)robotics and implantable or ingestible medical devices manoeuvring throughout the body, for devices in regenerative medicine, wearables, and environmental monitoring devices operating autonomously in aqueous environments.

Autonomous movement is an essential capability in living organisms. It allows the individual species to grasp and manipulate objects, as well as to move, to escape danger, find food, or meet other individuals. Nature has developed various means that enable movement and locomotion, from protein motors that drive cilia to specialized muscle tissue, all autonomous and self-powered. In mammalian muscles, glucose and oxygen (O₂) are consumed to generate adenosine triphosphate (ATP) by aerobic respiration. The ATP, being converted to adenosine diphosphate due to hydrolysis, in turn drives the nanoscopic conformational changes in the myosin heads with respect to the actin filaments that cause the macroscopic muscle contraction and elongation.[1] In contrast, human-made contraptions such as industrial or humanoid robots are typically driven by classical actuators that use electrical, pneumatic, or thermal means and are often tethered or require large batteries. Driving actuators, or artificial muscles, directly using biofuels such as glucose is highly anticipated. It would enable (soft) robotic devices to be fueled by green biofuels such as glucose, methane, or alcohol and open up possibilities for driving implants or injectable vehicles in the body without the need for batteries or external charging sources such as by radiofrequency coils.

Analogous to the mammalian muscles, we demonstrate here artificial, polymer muscles driven by glucose and O₂. Soft, artificial muscles have been reported based on dielectric elastomers,[2] conducting polymers,[14] carbon nanotubes,[5,6] and even fishing lines[7] and textiles,[8] but they still all are driven by electrical or thermal means. Conducting polymer (CP) artificial muscles are interesting as they are operated at low voltages and behave similarly to mammalian muscle. During oxidation and reduction, conducting polymers change their volume—as well as other material properties such as color (electrochromism),[9] wettability,[10] and adhered protein conformation[11]—which is used to construct actuators. The volume change is based on the dimensional changes that occur as a result of electrochemically induced ion insertion and desertion, along with associated solvating species,[12,13] accompanied by conformational changes in polymeric chains.[14] Artificial muscles based on conducting polymers come in many configurations, one of which is the so-called trilayer or bimorph configuration (Figure 1A), which we use here to demonstrate the principle. The conducting polymer, e.g., polypyrrole (PPy) or poly(3,4-ethylenedioxythiophene), is sandwiched on two sides of a flexible membrane.[15,16] A potential difference is applied between the two conducting polymer layers, electrochemically oxidizing the conducting polymer on one side and reducing the conducting polymer on the other side. This causes one layer to shrink and the other to swell, thus resulting in a bending...
motion. However, these artificial muscles are still driven by an external, often tethered, electrical power source. Direct conversion of chemical energy into electrical energy to drive these biomimetic machines would be the most elegant solution and would capitalize on the efficiencies already achieved by nature. Catalysts such as enzymes can achieve such direct conversion and are, for instance, integrated in bioelectrodes. Coupled as a pair such bioelectrodes can form a biofuel cell that can generate electrical power from biofuels.[17] The power output of biofuel cells can drive microdevices that need relatively low power.[18] Bioelectrodes that convert glucose and O₂, respectively, are particularly interesting as they enable autonomous power generation in physiological fluids because glucose is present in all organs, thus eliminating the need to pump fuel. Also, O₂ is present in all organs, thus removing the need for air supply through a membrane at the cathode compartment.[19] Several research groups have used glucose as a biofuel. For example, Mano and Heller reported biological motion in the presence of glucose and oxygen to move a carbon fiber using glucose oxidase (GOx) and bilirubin oxidase to create a H⁺ flux that drove the fiber.[20] Movement of carbon nanotubes was achieved by using GOx and catalase to generate

Figure 1. Working principles of a trilayer artificial muscle and bioelectrodes. A) Schematic description of the functioning of a “trilayer” conducting polymer actuator or artificial muscle. B) Schematic description of the bioelectrode pair producing electricity to power, e.g., artificial muscles.
hydrogen peroxide that in turn was catalyzed into O$_2$ bubbles that propelled the nanotubes.\cite{21} Miyake et al. report a biofuel cell that used fructose and O$_2$ as fuels.\cite{22} It was prepared by modifying fructose dehydrogenase and bilirubin oxidase on carbon nanotube–decorated carbon fiber fabrics and could drive light-emitting diodes. A pacemaker powered by a glucose/O$_2$ biofuel cell was presented by MacVittie et al.\cite{23} Strack et al. used enzymes in solution to construct a biochemical logic circuit to control a chemical actuator.\cite{24} However, despite these advances, artificial muscles are still being driven using external means.

Here, we present a polymer artificial muscle powered by glucose and O$_2$ biofuels in which enzymatic electrodes are fused with the polymer artificial muscle to create an integrated bionic system. The artificial muscle was based on a PPy trilayer actuator using dodecylbenzenesulfonate (DBS) as the dopant. A tetraphiafulvalene 7,7,8,8-tetracyanoquinodimethane (TTF-TCNQ)/Aspergillus niger glucose oxidase enzyme–modified Au electrode and a 2,2’-azinobis(3-ethylbenzothiazoline-6-sulfonic acid) (ABTS)/Trametes versicolor laccase enzyme–modified Au electrode were used as the bioelectrodes.

First, the individual, external enzyme-based bioelectrodes were investigated. Each step of the surface-modified Au/Si electrode was verified by the cyclic voltammetry (CV) experiments in 0.1 M phosphate buffer solution (PBS, pH 5.0), in the absence and presence of glucose with scan rate of 100 mV s$^{-1}$. Figure S2A, curve a (Supporting Information) shows the CV obtained with the bare Au electrode as a control experiment. The TTF-TCNQ-modified Au electrode produced a current of 13.14 μA cm$^{-2}$ at a potential of 0.16 V and this peak was ascribed to the redox processes of TTF-TCNQ adsorbed on the Au electrode (Figure S2A, curve b, Supporting Information).\cite{25} As shown in Figure S2A, curve c (Supporting Information), after casting GOx enzyme on the electrode forming TTF-TCNQ/GOx, the current increased to 88.8 μA cm$^{-2}$ at a potential of 0.32 V, and along with the increase in anodic current, there was a decrease in the cathodic peak at 0 V. After adding the glucose, the electrocatalytic current increased to 262 μA cm$^{-2}$ for 0.1 M glucose at a potential of 0.32 V, showing that the bioanode exhibited a good bioelectrocatalytic activity for the oxidation of glucose (Figure S2A, curve d, Supporting Information).

Figure S2B (Supporting Information) shows the CV of the biocathode under the same conditions as for the bioanode experiment, i.e., in 0.1 M PBS (pH 5.0), and in the absence and presence of O$_2$ at a scan rate of 100 mV s$^{-1}$. The bare Au electrode was investigated as a control (Figure S2B, curve a, Supporting Information). When the bare Au electrode was modified with ABTS, the current of the ABTS/Au electrode increased to 16.84 μA cm$^{-2}$ at a potential of 0.64 V (Figure S2B, curve b, Supporting Information). Thereafter, lactase was chemically bound to the ABTS/Au electrode, and the current increased to 27.15 μA cm$^{-2}$ at a potential of 0.62 V (Figure S2B, curve c, Supporting Information). Finally, operation in the air-saturated 0.1 M PBS (pH 5.0) showed an increase of the peak current to 33.31 μA cm$^{-2}$ at a potential of 0.58 V, which was close to the redox potential of lactase and ABTS\cite{26} (Figure S2B, curve d, Supporting Information), with well-defined cathodic waves attributed to the reduction of O$_2$ by the lactase. The results indicate that lactase electrochemically catalyzed the reduction of O$_2$ through the mediated electron transfer reaction between the active center of lactase and the electrode.

Next, the bioelectrode pair was inserted (without a separating membrane) in a 0.1 M glucose and 30 min air-saturated 0.1 M PBS (pH 5.0) at room temperature. Figure 2A shows the polarization curves of TTF-TCNQ/GOx-modified Au electrode (curve a) and the lactase/ABTS-modified Au electrode (curve b). As expected, the recorded open-circuit potential (OCP) of 0.70 ± 0.04 V corresponds to the difference between the individually measured OCPs of the bioanode (Figure 2A, curve a) and the biocathode (Figure 2A, curve b). Figure 2B presents the OCP of the cell when the cell was operated continuously for 7 days. The cell OCP decreased by 20% per day and reached 0.15 V at day 7. This drop could be due to the consumption of glucose and O$_2$. The variation of the power density as a function of the cell voltage presents the typical bell-shaped curve observed by others,\cite{27,28} with a maximum of 0.27 μW cm$^{-2}$ at 0.50 V in 0.1 M PBS (pH 5.0) (Figure 2C). The cell voltage under an external load of 1 MΩ resistance was recorded for 27 h, operating in the 0.1 M PBS (pH 5.0). When the cell was operated continuously under this load, it lost 25% of its original power in the first 12 h and the power output dropped by 50% after a 27 h continuous work. The cell voltage decreased slowly to 0.10 V (Figure 2D). It was observed that the bioelectrode pair retained 60% of its initial power density up to 3 days when it was operated continuously. The decline in power density output after 3 days might be caused by a loss of mediators and enzymes, since they were not covalently attached to the electrodes.

Before connecting the artificial muscle to the external enzyme-based bioelectrodes, the muscle’s performance was checked using an external potentiostat in two different electrolytes, 0.1 M NaDBS and 0.1 M PBS (pH 5.0), using both CV and chronocoulometry (Figure S1B–E, Supporting Information). As can be seen in Figure S3 (Supporting Information), the actuator showed the normal reversible movement as previously seen for such trilayer actuators, indicating that the actuator was functioning correctly. The movement was reduced in the physiological PBS electrolyte as compared to simple NaDBS salt solution, which is consistent with previous results using cerebral physiological solutions.\cite{29} Operating at a slightly lower pH should not influence its performance.\cite{30}

Having established that the bioelectrode pair can deliver sufficient potential difference (~0.70 V) and power to drive a trilayer actuator\cite{31} and that the actuator operated normally, we connected the PPy/Au/poly(vinylidene fluoride) (PVDF)/Au/PPy trilayer artificial muscle to the external bioelectrodes. As can be seen in Figure 3, the movement of artificial muscle driven by the external bioelectrodes is clear. At $T_0$, the initial position, the artificial muscle was connected to the bioelectrodes and the muscle bent toward the anodic side as would be expected.\cite{32} On the anodic side, PPy(DBS) was oxidized and shrank and on the cathodic side PPy(DBS) was reduced and swelled, thus causing a bending to the left. The position of the artificial muscle after 4 h is indicated by point $T_2$ showing a 5 mm displacement.

Next, we integrated the bioelectrodes on the PPy/Au/PVDF/Au/PPy trilayer artificial muscle. The separate bioanode was integrated at the top on one side of the actuator and likewise the separate biocathode was integrated at the top on the opposing
side. In the integrated device, we initially found some leakage and mixing of the bioanode and biocathode materials through the porous PVDF membrane, and to ensure better retention, an additional reduced graphene oxide (RGO) layer was introduced as a barrier between the bioanode and biocathode. This resulted in the required performance. When inserted into the glucose/O$_2$-containing electrolyte, the actuator started to move directly. Figure 4A shows the movement upon insertion into the glucose/O$_2$-containing electrolyte. As with the external bioelectrodes, the muscle bent 2 mm toward the anode side demonstrating that the biocatalytic electrodes can be integrated with the artificial muscle. Thereafter, we fully integrated the enzymes onto the entire artificial muscle. The movement of the now self-powered PPy/Au/PVDF/Au/PPy artificial muscle is shown in Figure 4B. Again, the displacement occurred toward the anode side, moving clearly from its starting position $T_0$ to the final position $T_1$ after 3 h with a total displacement of 3 mm. Figure 4C shows the displacement over time of the fully integrated artificial muscle. It shows the typical displacement profile as seen for CP actuators that are diffusion limited, although slower than when driven by an external power supply.

To check whether the actuator is truly driven by faradaic reactions at the catalytic bioelectrodes, three control experiments were performed. First, the actuator without any bioelectrodes was inserted into the biofuel-containing electrolyte (Figure S4A,
Supporting Information). Next, the actuator with the fully integrated bioelectrodes was inserted in deionized (DI) water (Figure S4B, Supporting Information) and in a N2-saturated 0.1 M PBS (pH 5.0) (Figure S4C, Supporting Information), respectively, that is, two solutions without any biofuel. In neither of the three cases, any movement could be observed, indicating that indeed the movement was caused by the catalytic conversion of O2 and glucose.

A single movement toward one side only has limited applications, for instance, as a single-use valve. To be useful as self-powered artificial muscle to drive (soft robotic) devices, reversible movement of the actuator is required. To achieve this, we integrated an anode and cathode on both the top and bottom of each side of PPy/Au/PVDF/Au/PPy trilayer artificial muscle, but in a mirrored configuration. We inserted the integrated muscle into the solution while keeping the top electrode pair above the water level (i.e., inactive). As seen previously, the muscle bent toward the anode side that is to the left, displacing 2.5 mm in 3 h (from \( T_0 \) to \( T_1 \) in Figure 5A). Next, we rotated the muscle so that the inserted electrode pair went above the water level and noninserted electrode pair came under the water level, thus de facto changing the polarity of the inserted bioelectrodes and thus reversing the electrochemical reactions in the PPy layers. The left side, which was now the cathode side, was reduced and the PPy(DBS) layer swelled and the right side, now the anode, was oxidized and shrunk. This resulted in a bending motion to the right, returning to the original position (from \( T_2 \) to \( T_3 \) in 3 h) as shown in Figure 5B, i.e., a reversible self-powered artificial muscle.

We have demonstrated that enzyme-based bioelectrodes that use glucose and O2 could power artificial muscles. The immobilization of the enzymes with redox mediators resulted in a potential difference over the bioelectrodes of more than 0.50 V, which was sufficient to drive an externally connected actuator. In glucose and air-saturated 0.1 M PBS (pH 5.0), a maximum power density of 0.27 \( \mu W \) cm\(^{-2} \) was measured at room temperature. The OCP of catalytic bioelectrode pair was 0.70 \( \pm \) 0.04 V. The stability of the bioelectrodes was tested for around 27 h with a load of 1 M\( \Omega \). We then demonstrated an autonomously powered artificial muscle by integrating the catalytic bioelectrodes. These self-powered artificial muscles showed reversible motion in both directions. The bioelectrodes...
manufactured according to this process have good stability and are promising candidates to autonomously drive artificial muscles.

In this work, we demonstrated for the first time the principle of self-powered artificial muscles using the body’s principal biofuel. To enhance the biofuel-powered actuator performance, including speed, power output stability (e.g., by improved immobilization, engineering the enzymes) and the magnitude of the generated potential (as increasing the potential difference will increase the speed of actuation\(^{31}\)) need to be increased, for instance, by better integration of the enzymes and optimization of the ratio of biocathode/anode materials and conducting polymer. We used the trilayer actuator configuration, whose range of motion can be improved by optimizing the actuator design (e.g., using a softer, more flexible membrane, optimizing the ratio between the three actuator layers\(^{33}\)) to exemplify biofuel-powered artificial muscles as a bending motion is clearly visible, but this is only one possible configuration of conducting polymer artificial muscles. The concept can be generalized to other artificial muscle configurations. Linear actuation\(^{8}\) and complex devices comprising multiple actuators\(^{4}\) are especially interesting future targets. The laccase and glucose oxidase reactions might have asymmetric electron transfer, which could result in nonsymmetric movements, which is not visible in trilayer actuators since the bimorph design with two identical PPy layers ensures symmetric deflection. To get a symmetric charge transfer, one would, for instance, need to optimize the ratio between laccase and glucose oxidase enzymes and/or adjust the mediators that assist in the charge transfer from the enzyme to the conducting polymer layer. Here, we used the body’s principal biofuel glucose, but the same concept can be extended to using other in vivo fuels or even other biofuels present in the environment to enable autonomous propulsion of robots that can “swim” through lakes taking samples for environmental monitoring. We envision a future where biofuel-powered artificial muscles offer an efficient solution for much needed silent autonomous propulsion in soft microrobots, ingestible devices, and implantable artificial muscles.

**Experimental Section**

**Chemicals**: Glucose oxidase EC 1.1.3.4, from *Aspergillus niger*, laccase enzyme of *Trametes versicolor*, ABTS, and TTF-TCNQ were purchased from Sigma-Aldrich. Pyrrole (from SAFC, acquired through TCI, Japan) was distilled under vacuum prior to use and stored at \(-20^\circ\)C. Porous PVDF with 0.45 µm pore size and a thickness of 110 µm (Immobilon-P) was purchased from Merck (Germany). \(d\)-Glucose, \(NH_4\)Cl (98%), and \(H_2O_2\) (30%) were purchased from VWR. Potassium phosphate dibasic anhydrous (\(K_2HPO_4\)), potassium dihydrogen phosphate (\(KH_2PO_4\)), sodium acetate, and glacial acetic acid were obtained from Merck (Germany). All solutions were prepared using 18 MΩ DI water (Milli-Q).

**Bioelectrode Fabrication and Characterization**: The bioelectrodes acting as working electrodes and containing enzymes and mediators were prepared by immobilizing the constituent materials onto a 0.5 cm × 0.5 cm Au/Si electrode. First, the bare Au electrode surfaces were cleaned by immersion in \(NH_4\), \(H_2O_2\), and DI water (1:1:5) at 85 °C for 10 min and thereafter washed gently with DI water. For the bioanode\(^{34}\) preparation, 10 µL of TTF-TCNQ was drop cast on the Au electrode and dried at room temperature for 15 min. Then, 10 µL GOx enzyme solution (5 mg mL\(^{-1}\)) was deposited onto the TTF-TCNQ/Au surface and gently dried under airflow in a laminar flow hood for 15 min. The biocathode\(^{34}\) was prepared by pipetting 10 µL of a 0.002 M ABTS solution and drying at room temperature for 15 min. After that, 10 µL of laccase solution (5 mg mL\(^{-1}\)) was cast on the ABTS/Au support and gently dried under airflow in a laminar flow hood for 15 min. For the bioelectrodes integrated on the artificial muscle, 10 µL of RGO was drop cast as a barrier on both sides of each bioelectrode before adding mediators and enzymes. The RGO was synthesized from graphite through a modified Hummers’ method\(^{35}\).

Electrochemical measurements were performed at room temperature using an ivium compactstat or IviumStat potentiostat/galvanostat (Eindhoven, The Netherlands) in a three-electrode electrochemical cell, which included a modified Au/Si electrode as the working electrode, a piece of Au/Si 3 cm × 3 cm as the counter electrode, and a BASi MF-2052 Ag/AgCl (3 M KCl) reference electrode. All potentials in this work are referred to this electrode. Cyclic voltammograms were performed in a cell containing 5 mL of 0.1 M PBS (pH 5.0) at 100 mV s\(^{-1}\), over potential ranges from –0.50 to 0.50 V for the bioanode and 0–1 V for the biocathode. The electrochemical properties of the anodic and cathodic electrodes were obtained in the absence and presence of 0.1 M glucose (dissolved in 0.1 M PBS) and air saturation (after 30 min bubbling air), respectively.
To assess possible power generation, both bioelectrodes were submerged in a beaker filled with 20 mL of 0.1 M PBS (pH 5.0) with 0.1 M glucose and saturated with air (30 min). The electrodes consisted of 1 cm × 1 cm pieces of a Au-coated silicon wafer. The TTF-TCNQ/GOx served as the bioanode for the oxidation of glucose and mediated electron transfer. The ABTS/laccase served as the biocathode for reduction of O$_2$ by mediated electron transfer. The bioanode and biocathode were combined, thus de facto forming a classical membraneless biofuel cell, and Figure 1B illustrates the mechanism. The complete cell was allowed to equilibrate at OCP before measurements were performed.

The OCP of the bioelectrode pair was investigated over a period of 7 days. The electrodes were stored at room temperature and the OCP checked regularly using a potentiostat/galvanostat (Ivium compactstat) with a very high input impedance (>1000 GΩ). For the stability test, an external load of 1 MΩ was connected between the two electrodes. The power output of the cell in the stability tests was calculated from the potential and resistance values according to Ohm’s law.

**Artificial Muscle Preparation:** For the preparation of the artificial muscle, a thin Au film was sputtered on both sides of a piece of PVDF membrane using a Vacutec Plasma Systems Sputter at a pressure of 4.6 mTorr, 36.6 cm$^3$ min$^{-1}$ Ar flow, and a sputter rate of 5 nm min$^{-1}$. Next, PPy was synthesized electrochemically on both sides of Au/PVDF/Au membrane at 0.6 V versus Ag/AgCl by immersing the membrane in a solution containing 0.1 M pyrrole and 0.1 M NaDBS. For the electrosynthesis, each side of Au/PVDF/Au was connected to the working electrode separately, a stainless steel mesh (2 cm × 7 cm) was used as the counter electrode, and the same Ag/AgCl reference electrode as the previous one was used. A typical chronoamperogram of the PPy synthesis on one side of Au/PVDF/Au for 3000 s is shown in Figure S1A (Supporting Information). After PPy electropolymerization on both sides, strips of 0.5 cm × 2 cm were cut, resulting in PPy/Au/PVDF/Au/Ppy trilayer artificial muscles. The artificial muscles were actuated in 0.1 M NaDBS solution and 0.1 M PBS (pH 5.0) at room temperatures using CV and chronoamperometry (Figure S1B–E, Supporting Information). CVs of the PPy/Au/PVDF/Au/Ppy artificial muscle were taken over the potential ranges −1.7 to 0.50 V (in 0.1 M NaDBS) and −1.2 to 0.2 V (in 0.1 M PBS, pH 5.0), in both cases with a scan rate of 20 mV s$^{-1}$. The chronoamperometric activation of the PPy/Au/PVDF/Au/Ppy artificial muscle was performed at constant potentials of −0.45 V (in 0.1 M NaDBS) and −0.50 V (in 0.1 M PBS, pH 5.0).

**Enzyme-Driven Artificial Muscles:** The artificial muscle was fabricated integrating the enzymes in four different configurations to investigate different properties, and with increasing complexity and increasing integration: i) the enzymes as bioelectrodes externally connected to the artificial muscle; ii) the enzymes integrated on artificial muscle as a single, separated bioelectrode directly connected to each side of the PPy/Au/PVDF/Au/Ppy actuator (i.e., bioanode on one side and biocathode on the other side); iii) fully integrated enzymes on each side of the actuator; and iv) the enzymes as a bioelectrode pair on each side, but mirrored (i.e., anode-cathode pair on one side and cathode-anode pair on the other side). The fabrication of the artificial muscle with the integrated enzyme-based bioelectrodes combined the fabrication steps as described in the previous two sections. Sketches of the configurations are included in each figure.

**Artificial Muscle Driven by External Potentiostat:** To investigate the function of artificial muscle before connecting to the external bioelectrodes, the motion of PPy/Au/PVDF/Au/Ppy trilayer artificial muscle was surveyed using the potentiostat in chronoamperometric mode for two cycles of 0 V for 300 s and −0.70 V for 300 s in two different electrolytes, 0.1 M NaDBS and 0.1 M PBS (pH 5.0), respectively. The potential range of 0 to −0.70 V was chosen to simulate the maximum OCP potential. After showing good performance of the artificial muscle movement using the potentiostat, the PPy/Au/PVDF/Au/Ppy trilayer artificial muscle was connected to the external bioelectrodes and characterized. This movement experiment was run for around 4 h.

All the movements were recorded using a digital microscope (Dino-Lite AM785MZT). All images in Figures 3–5 are merged from two frame grabs as indicated by the dashed line. The original figures are found in the Supporting Information.

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

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**Conflict of Interest**
The authors declare no conflict of interest.

**Keywords**
artificial muscles, electroactive polymers, glucose oxidase, laccase, polypyrrole

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[1] G. J. Tortora, S. R. Grabowski, Principles of Anatomy and Physiology, Wiley, New York 2000.
[2] R. Pelrine, R. Kornbluh, Q. B. Pei, J. Joseph, Science 2000, 287, 836.
[3] E. Smela, O. Inganas, I. Lundstrom, Science 1995, 268, 1735.
[4] E. W. H. Jager, O. Inganas, I. Lundstrom, Science 2000, 288, 2335.
[5] R. H. Baughman, C. X. Cui, A. A. Zakhidov, Z. Iqbal, J. N. Barisci, G. M. Spinks, G. G. Wallace, A. Mazzoldi, D. De Rossi, A. G. Rinzler, O. Jaschinski, S. Roth, M. Kertesz, Science 1999, 284, 1340.
[6] J. Foroughi, G. M. Spinks, G. G. Wallace, J. Oh, M. E. Kozlov, S. Fang, T. Mirfakhrai, J. D. W. Madden, M. K. Shin, S. J. Kim, R. H. Baughman, Science 2011, 334, 494.
[7] C. S. Haines, M. D. Lima, N. Li, G. G. Wallace, J. Foroughi, J. D. W. Madden, S. H. Kim, S. Fang, M. J. de Andrade, F. Goktepe, O. Goktepe, S. M. Mirvakili, S. Naficy, X. Lepro, J. Oh, M. E. Kozlov, S. J. Kim, X. Xu, B. J. Swedlove, G. G. Wallace, R. H. Baughman, Science 2014, 343, 868.
[8] A. Maziz, A. Concias, A. Khalidi, J. Ståhlman, N.-K. Persson, E. W. H. Jager, Sci. Adv. 2017, 3, e1600327.
[9] F. Vidal, C. Plessie, P.-H. Aubert, L. Beouch, F. Tran-Van, G. Palaprat, P. Verge, P. Yannine, J. Citerin, A. Kheddar, L. Sauques, C. Chevrot, D. Teyssie, Polym. Int. 2010, 59, 313.
[10] J. Isaksson, C. Tengstedt, M. Fahlin, N. Robinson, M. Berggren, Adv. Mater. 2004, 16, 316.
[11] K. Svennersten, M. H. Bolin, E. W. H. Jager, M. Berggren, A. Richter-Dahlfors, Biomaterials 2009, 30, 6257.
[12] Q. Pei, O. Inganas, J. Phys. Chem. 1992, 96, 10507.
[13] L. Bay, T. Jacobsen, S. Skaarup, K. West, J. Phys. Chem. B 2001, 105, 8492.
[14] T. F. Otero, J. G. Martinez, Chem. Mater. 2012, 24, 4093.
[15] F. Vidal, C. Plesse, D. Teyssie, C. Chevrot, Synth. Met. 2004, 142, 287.
[16] D. Z. Zhou, G. M. Spinks, G. G. Wallace, C. Tiyapiboonchaiya, D. R. MacFarlane, M. Forsyth, J. Z. Sun, Electrochim. Acta 2003, 48, 2355.
[17] M. Falk, Doctoral Thesis, Faculty of Health and Society, Malmö University, Malmö, Sweden 2014.
[18] A. Pizzariello, M. Stred’ansky, S. Miertus, Bioelectrochemistry 2002, 56, 99.
[19] A. Heller, Phys. Chem. Chem. Phys. 2004, 6, 209.
[20] N. Mano, A. Heller, J. Am. Chem. Soc. 2005, 127, 11574.
[21] D. Pantarotto, W. R. Browne, B. L. Feringa, Chem. Commun. 2008, 1533.
[22] T. Miyake, K. Haneda, S. Yoshino, M. Nishizawa, Biosens. Bioelectron. 2013, 40, 45.
[23] K. MacVittie, J. Halamek, L. Halamkova, M. Southcott, W. D. Jemison, R. Lobeld, E. Katz, Energy Environ. Sci. 2013, 6, 81.
[24] G. Stack, V. Bocharova, M. A. Arugula, M. Pita, J. Halamek, E. Katz, J. Phys. Chem. Lett. 2010, 1, 839.
[25] A. R. Harris, J. Zhang, R. W. Cattrall, A. M. Bond, Anal. Methods 2013, 5, 3840.
[26] R. Bourbonnais, D. Leech, M. G. Paice, Biochim. Biophys. Acta, Gen. Subj. 1998, 1379, 381.
[27] Y. Chen, P. Gai, J. Zhang, J.-J. Zhu, J. Mater. Chem. A 2015, 3, 11511.
[28] A. Zebda, C. Gondran, A. Le Goff, M. Holzinger, P. Cinquin, S. Cosnier, Nat. Commun. 2011, 2, 370.
[29] E. D. Daneshvar, E. Smela, Adv. Healthcare Mater. 2014, 3, 1026.
[30] S. Shimoda, E. Smela, Electrochim. Acta 1998, 44, 219.
[31] T. W. Lewis, G. M. Spinks, G. G. Wallace, A. Mazzoldi, D. De Rossi, Synth. Met. 2001, 122, 379.
[32] Q. Pei, O. Inganas, J. Phys. Chem. 1993, 97, 6034.
[33] M. Benslimane, P. Gravesen, K. West, L. Bay, S. Skaarup, in Proc. 7th Int. Conf. on New Actuators, Messe Bremen GmbH, Bremen, Germany 2000, pp. 635–638.
[34] Note: The definition of the cathode and anode depends on whether the system is regarded as a galvanic cell or electrochemical cell. Since we are combining two cell types into a fused device, this definition can become confusing. We have used the following conventions: The bioanode is the electrode where glucose is oxidized and electrons are transferred into the electrode, and the biocathode is the electrode where O2 is reduced and electrons are transferred from the electrode (following classical biofuel cell definition). The cathode is the electrode that reduces PPy (by donating electrons) and the anode is the electrode that oxidizes PPy (by accepting electrons). Thus, the bioanode is the cathode and the biocathode is the anode with respect to the electrochemical actuator and as used in Figures 3–5.
[35] D. C. Marcano, D. V. Kosynkin, J. M. Berlin, A. Sinitskii, Z. Sun, A. Slesarev, L. B. Alemany, W. Lu, J. M. Tour, ACS Nano 2010, 4, 4806.
[36] R. L. Arechederra, S. D. Minteer, Fuel Cells 2009, 9, 63.
[37] E. W. H. Jager, N. Masurkar, N. F. Nworah, B. Gaihre, G. Alici, G. M. Spinks, Sens. Actuators, B 2013, 183, 283.