



Invited viewpoint: biodegradable Mg batteries

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Received: 18 July 2023

Accepted: 1 August 2023

Published online:

16 August 2023

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ABSTRACT

The development of biodegradable electronics, or transient electronics, excludes the need for second surgeries for device removal, reduces the potential infection risks, and also opens up opportunities in the development of zero-waste and green electronics which would be degraded after a certain period of operation. Batteries are a vital component of transient electronics since they serve as biodegradable power sources. Recently, magnesium-based biodegradable batteries have gained considerable attention owing to their advantages of high specific capacity, high energy density, long shelf-life, desirable biodegradability, high physiological tolerance, low cost, and high safety. The main drawback of Mg, as a widely explored anode material in biodegradable batteries, is its high degradation rate due to a low corrosion resistance, especially in aqueous environments. Therefore, controlling the degradation rate of Mg anodes is crucial, and this is usually achieved by several strategies such as coating, adding alloying elements, and thermomechanical processing techniques. Moreover, the development of biodegradable battery cathode materials and electrolytes is also crucial for preparing fully biodegradable batteries. The current study is designed to give an overview of the up-to-date research progress on the development of anode, cathode, and electrolyte materials for use in biodegradable Mg batteries. In this respect, the strategies for materials selection, the fabrication schemes, battery architectures, and their electrochemical and in vivo performance are summarized. Finally, the future outlook is discussed to help in the development of green biodegradable Mg batteries that are viable in the fields of medicine, flexible wearables, and consumer electronics.

Handling Editor: C. Barry Carter.

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<https://doi.org/10.1007/s10853-023-08828-2>

Introduction

Biodegradable electronics, or transient electronics, is an emerging type of device with a transient function which have gained considerable attention in recent years for biomedical therapeutic/diagnostic processes or green applications such as powering implantable medical devices (IMDs) and sensors. These devices are made of biodegradable components, and their key feature is their degradation capability in physiological or environmental conditions in a controlled manner after functioning [1–5].

The biodegradability of the electronic devices is beneficial from different viewpoints such as: (1) it excludes second surgeries for device removal and, consequently, could reduce potential risks of infection, and also the accompanying hospital costs, (2) it is favorable for human healthcare and the environment and alleviates concerns accompanying the electronic-waste (e-waste), such as landfill space and toxic elements, and also eliminates the costs and risks of associated recycling operations, and (3) it provides a data-secure hardware system which prevents unauthorized access to security data because of its self-destruction [6–8].

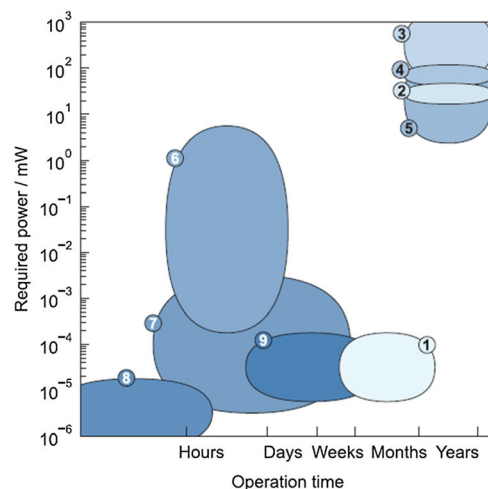
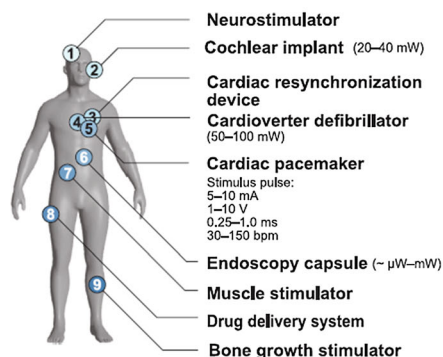
Among biodegradable electronics, transient power sources are the main challenge toward forthcoming medical uses. Currently, wired external power or wireless energy harvesters via near-field coupling are used with the associated limitations of device orientation and position [9]. As a key example, suitable transient energy sources are needed for onboard powering of IMDs which are helpful in treating, monitoring or diagnosing health conditions inside

the body [10]. Figure 1 gives some examples of different implantable medical electronics, their application in the human body, and their estimated required power and expected lifespans. As summarized in this figure, different operating times and degradation rates are favorable depending on the required application.

Batteries, either primary (non-rechargeable) or secondary (rechargeable), remain the prevailing power sources in implantable medical electronic devices, owing to their high energy density and energy conversion efficiency. The operational mechanism of transient batteries is similar to durable batteries, both converting the chemical energy to the electrical energy. The constituents of these batteries are positive (cathode) and negative (anode) electrodes, electrolyte, separator, current collectors, and an external encapsulation or packaging [11, 12]. To provide onboard power for implantable medical devices *in vivo*, a biodegradable battery is a crucial component due to its biocompatibility, biodegradability, long lifetime, compactness, and high achievable energy density. While an appropriate battery design should fulfill the energy and power requirements, it is also necessary to degrade into non-toxic products after approaching the end of its lifetime. It is important to mention that miniaturization is an important design factor for transient batteries that are aimed for biomedical applications [13, 14].

At present, the main challenge in developing biodegradable batteries is the lack of biodegradable battery components leading to excessive materials retention that could be harmful to the human body and the environment, or to battery features including

Figure 1 Some examples of implantable medical devices, their application in the human body, and the estimated power requirements and expected lifespans, reproduced with permission from Ref. [11].

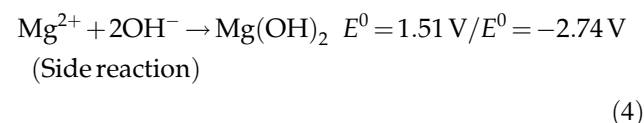
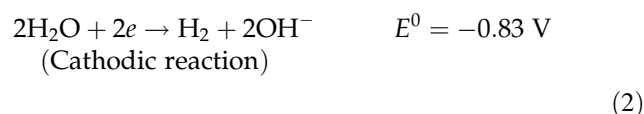


power, voltage, capacity or lifespan that may fall beyond the practical requirements [9]. In the progress of biodegradable batteries, magnesium, Mg, is a widely explored anode material due to its high specific capacity (2200 mAh g⁻¹), high specific energy (6800 Wh kg⁻¹), long shelf-life, desirable biocompatibility, and exceptionally high physiological tolerance (daily allowance of ~ 300 mg day⁻¹) [14, 15].

Most of the research performed on biodegradable Mg batteries has been focused on the advancement of an individual battery constituent, especially the anode, cathode or electrolyte. Apart from review papers on Mg batteries [16–18], biocompatible batteries [11], and transient electronics [4, 19–22], it appears that there are no reviews that comprehensively focus on biodegradable batteries. Accordingly, the focus of the current study is on biodegradable Mg batteries. This study summarizes the up-to-date progress achieved on biodegradable Mg battery components, their fabrication methods, and the battery configurations with a focus on its electrochemical and in vivo performance (if reported). Finally, a summary and perspectives are presented to further advance the biodegradable Mg batteries for health-care and eco-friendly applications.

Anodes for biodegradable Mg batteries

A suitable anode material is one of the most significant influencing factors in the appropriate performance of batteries. Pure Mg, as the most popular anode material in biodegradable Mg-based batteries, possesses attractive electrochemical characteristics. Generally, the degradation mechanism of biodegradable metals is through a corrosion process, in which, by immersing the metal in water/biofluids, electrochemical reactions occur to form metal cations and other products such as hydroxides, oxides, phosphates, and hydrogen gas [22]. Mg is an active metal, and in acid and neutral solutions, it is suggested that oxidation and electrochemical corrosion of Mg occurs at the anode after contacting with H₂O, while the possible reactions at the cathode could be either the reduction of oxygen or the evolution of hydrogen, and also the formation of Mg(OH)₂ on the surface, as follows [23]:



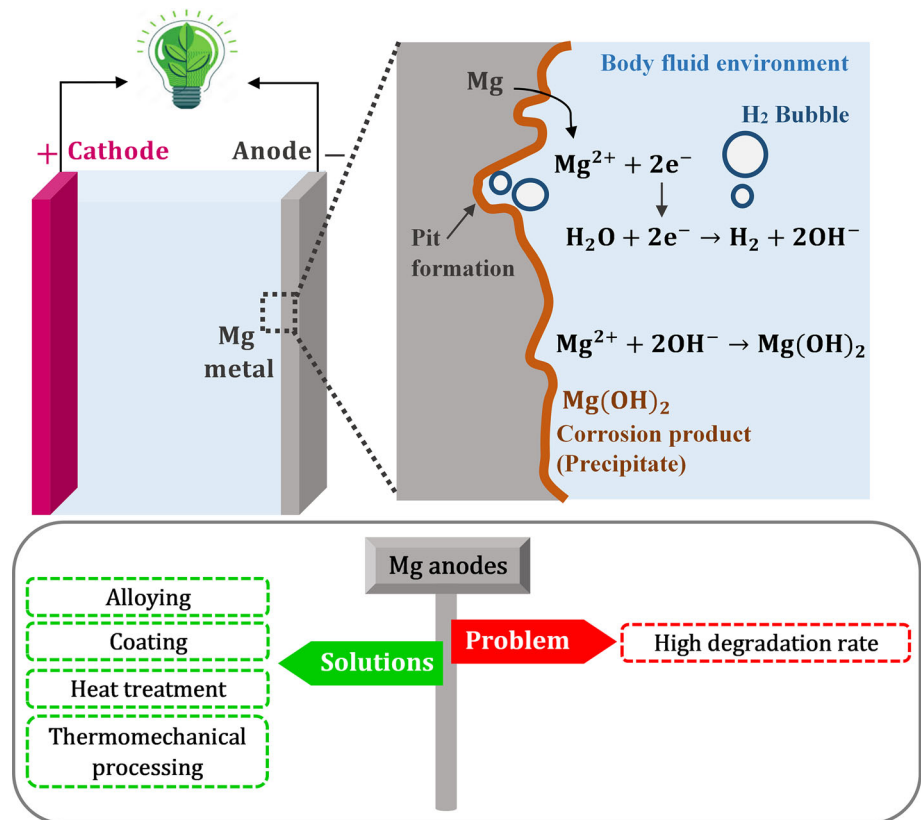
In fact, Mg dissolves as Mg²⁺ ions and then the reaction of these ions with water produces hydrogen bubbles and hydroxyl groups. Through the reaction of dissolved Mg²⁺ ions with hydroxyl groups, precipitates of magnesium hydroxide will form [24]. The schematic illustrations of Mg batteries and the Mg degradation mechanism are shown in Fig. 2.

The types of corrosion that may occur for Mg alloys in aqueous solutions are mostly classified to general corrosion, pitting corrosion, and localized corrosion, while the latter includes intergranular corrosion, filiform corrosion, corrosion fatigue, stress corrosion cracking (SCC), etc. Regarding SCC, implantable devices such as batteries are affected by complicated stresses during placing in the human body, including tensile, compressive, and internal stresses. Therefore, SCC may occur with a high probability under the influence of stresses and the corrosive internal environment, and consequently, it may increase the degradation rate. Various corrosion mechanisms of Mg alloys have been recently reviewed in detail [25].

In order to accurately design and engineer a transient battery, it is crucial to predict the degradation conditions of the battery. Generally, degradation is dependent on a multitude of factors including the environmental conditions (the presence of various ions and compounds, pH, temperature, etc.) and the nature of the battery components (geometry, surface area, morphology, chemical and crystalline structure, etc.) [22]. These variables will be discussed briefly in the following.

Regarding the degradation medium, the body fluid environment is complex, and therefore, the degradation mechanisms of alloys in body fluids are different from those in simulated fluids. Mostly, in vitro

Figure 2 Schematic illustration of Mg batteries and Mg degradation mechanism; the most severe problem associated with Mg anodes and the applicable strategies for solving these problems.



experiments have been conducted to study the effects of body fluid variables on Mg degradation, because the in vivo experiments are expensive and also associated with ethical burdens. A variety of factors in simulated body fluids affect the degradation behavior of Mg alloys, including the presence of various types and concentrations of ionic and organic compounds, pH, temperature, flow field environment, etc. [25].

The existence of a variety of simple ions and complex compounds in human body fluids can lead to unpredicted results in comparison with in vitro studies. Organic ingredients such as proteins, vitamins, glucose, and amino acids are present in human plasma, but the simulated fluids used in most in vitro investigations do not contain these ingredients, and therefore, their effects on the degradation behavior of materials should be considered. The presence of inorganic salt ions such as Cl⁻, SO₄²⁻, NO₃⁻, Br⁻, Mg²⁺, Ca²⁺ may affect the corrosion behavior of Mg alloys in simulated fluids. Other important influencing factors are the experimental parameters such as pH, temperature, and flow field environment. At low pH or too high pH, the degradation rate usually

increases. Temperature affects the degradation rate by changing the chemical reactivity. Moreover, previous investigations have demonstrated that the degradation rate of Mg alloys in a static state is considerably lower than in the flowing state, because of the presence of shear force and mass transport in the flow field. Moreover, the effect of blood flow velocity, which varies between 0.1 and 1.0 m s⁻¹ (depending on the vessel diameter), on the degradation behavior has been extensively studied [22, 25, 26].

The degradation behavior of Mg alloys is diverse in vitro and in vivo, because of the difference in experimental conditions and corrosion rate assessment methods. Although in vitro degradation experiments are designed to simulate the in vivo environmental conditions, they may not fully duplicate the complexity and variability of the in vivo environment. Furthermore, one of the environmental parameters affecting the in vivo degradation rate is the anatomical location of the implanted device. For example, variations in the flow rates, the water content of the tissue, and the hydrogen diffusion coefficients in different animal models have been detected in different sites of the body. Nevertheless, further

investigations have suggested that the order of change in the average corrosion rate is negligible [27].

Another worthy feature is that the degradation could be controlled via external stimuli. For this purpose, the chemistry of the battery constituents (cathode, anode, electrolyte, separator, current collector, and packaging) could be engineered in such a way that the degradation process does not initiate until the introduction of definite triggers such as water, pH change, exposure of light, or temperature. Since tailoring the transiency may affect the electrochemical performance of the battery, therefore the desirable balance between transiency and the electrochemical performance should be considered [22].

Regarding the effect of the battery component properties on the degradation kinetics, the geometry of the battery constituents is an example. Various shapes/forms of Mg possess different crystal orientations, grain sizes, and surface morphologies which dictate various degradation behaviors in the battery. As an example, the corrosion resistance of electroplated Mg ($2.54 \mu\text{m h}^{-1}$) is lower than for commercial Mg foil (9–11 days) in PBS solution (pH 7.4, 37 °C), probably because of the larger grain size and higher surface roughness [22].

To the best of the authors' knowledge, the reported investigations on Mg-based biodegradable batteries are mostly on the effects of chemistry and crystal structure of the battery materials on the degradation and electrochemical behavior of batteries, and the influences of the other mentioned parameters on the battery performance have not yet been studied. The reported literature on biodegradable Mg batteries will be reviewed in the following.

Despite the attractive characteristics of Mg as the anode, Mg-based batteries show low columbic efficiencies and fail to deliver the theoretical capacity because of the occurring corrosion and fast self-discharging, resulting in a low discharge capacity, small specific power, and a short lifetime of the battery. Furthermore, rapid degradation of Mg and/or creation of surface barrier layers would limit the battery lifetime [14, 28–30]. Several ways have been developed and used for controlling the degradation rate of Mg, where the main methods are those based on alloying [31], coating [32, 33], heat treatment [34], and thermomechanical processing [35–38]. By adding different alloying elements such as Zn [35], Ca [39], Sr [40, 41], Y [42], and Ag [43], the degradation rate can be decreased due to special microstructural changes

such as grain refinement [35] and texture modification [44], and also, the effects these elements may have on the stability of the corrosion film [45, 46].

With reference to adding alloying elements, it is important to note that in the case of biomedical uses only a few alloying elements with biocompatibility characteristics could be chosen and this may restrict the range of available elements. Among biocompatible alloying elements, adding Zn, Zr, and Al to Mg has been reported and this can reduce the degradation rate and enhance the discharge behavior of Mg [10, 14]. The high degradation rate, which is the most severe problem associated with Mg anodes, and the applicable strategies for solving this problem are represented in Fig. 2.

Regarding the alloying strategy, an effective biodegradable battery was developed by fabricating a binary $\text{Mg}_{100-x}\text{Zn}_x$ ($0 \leq x \leq 100$ at%) alloy. Six $\text{Mg}_{100-x}\text{Zn}_x$ ($x = 0, 6, 20, 34, 41, 100$) systems were prepared by combinatorial magnetron co-sputtering of pure Zn and Mg. Sputtered Fe and phosphate buffered saline (PBS) served as the cathode and electrolyte, respectively. In practice, PBS is a good electrolyte because of its resemblance to the fluid in the human body. The prepared samples, including pure Zn, pure Mg, and six $\text{Mg}_{100-x}\text{Zn}_x$ alloys denoted as MZ- X ($X = 1-6$, and zinc content increases with X number), were examined via structural and electrochemical properties. Electrochemical measurements in biodegradable Mg batteries usually include two phases of: (1) examining the corrosion characteristics of the obtained samples and (2) studying the discharge behavior of the prepared batteries in respective three-electrode-cell and two-electrode-cell setups [14].

Evaluations by XRD revealed that binary solid solutions were formed in MZ-2 and MZ-3 samples, while at higher amounts of Zn addition (MZ-4 and MZ-5) amorphous single-phase structures were detected suggesting a possible better corrosion resistance. Figure 3a demonstrates that by increasing the Zn concentration in the system, E_{corr} is shifted toward nobler values and I_{corr} is decreased. Faster anodic dissolution was observed for the MZ-3, MZ-4, and MZ-5 systems by comparison with pure Mg, but they were passive upon higher anodic potentials because of the formation of $\text{Mg}(\text{OH})_2$ and $\text{Zn}(\text{OH})_2$ [14].

The discharge profiles of the prepared samples are shown in Fig. 3b where the highest output voltage

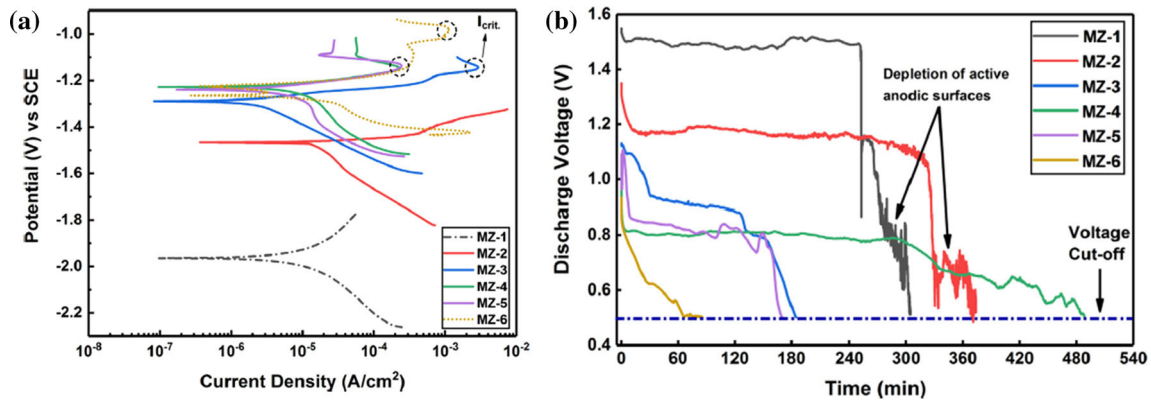


Figure 3 **a** Potentiodynamic polarization and **b** galvanostatic discharge curves at $10 \mu\text{A cm}^{-2}$ (0.5 V was the cutoff potential) of the MZ-X systems, reproduced with permission from Ref. [14].

was observed for pure Mg. Complete degradation before reaching the cutoff potential was evident in the MZ-1 and MZ-2 samples (designated by arrows). Among the binary samples, MZ-4 showed the longest lifetime (optimized sample) which could be ascribed to the synergic effects of amorphous structure and optimum concentration of alloying elements. In conclusion, the main limitations of pure Mg and pure Zn as anode materials are the high electrochemical reactivity of Mg and noble electrochemical potential of Zn, which may be overcome by combinatorial fabrication of the Mg–Zn alloy leading to a modification of corrosion resistance and discharge features of the system [14].

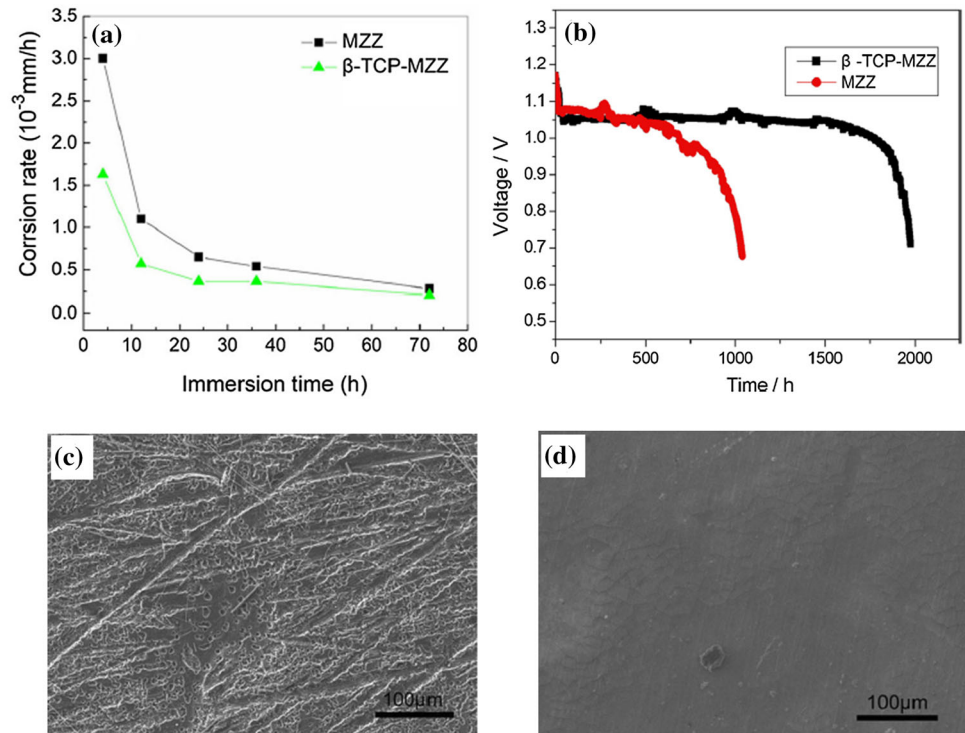
In order to reduce the degradation rate of the anode, AZ31 (Mg-3 wt% Al-1 wt% Zn) was examined in another study [47]. Thus, Fe served as the cathode and water reduction occurred on its surface during the performance of the battery. The alloying elements used and Fe are reasonably economical and biocompatible with the daily recommended intake (in mg) of 8–15, 7–9, and 2–12 for Fe, Al, and Zn, respectively. PBS and seawater were used as the electrolyte. The electrochemical characterizations revealed that using AZ31 as the anode extended the battery life by more than six times by comparison with that of pure Mg anode. The delivered voltage by these two batteries (AZ31/Fe and Mg/Fe) was very similar (nearly 0.27 V at $250 \mu\text{A}$). The capacity obtained, power, energy, and discharge time for the AZ31-based battery were 3303 mAh , $67 \mu\text{W}$, 3.2 J , and 13.2 h , while these values were 468 mAh , $69 \mu\text{W}$, 0.47 J , and 1.9 h for the pure Mg-based battery. The operating voltage and power of the fabricated batteries matched well

with the prerequisites for low-powered electronic devices.

In another recent investigation [23], a transient cell was constructed by using AZ31, anodized CuO, and a chloride-containing media (seawater) as the anode, cathode, and electrolyte, respectively. For comparison, pure Mg was also evaluated as the anode with the same cathode and electrolyte. The findings revealed a discharge life of 8.68 h and a specific capacity of 2.17 mAh cm^{-2} which were twice those of a pure Mg-based cell. Furthermore, the cell with the AZ31 anode delivered a higher specific energy of 1.63 J cm^{-2} by comparison with 1.08 J cm^{-2} of a cell with a pure Mg anode. The improved battery performance was attributed to the confined inherent pitting corrosion of Mg because of using the alloying elements (Al, Zn) in the anodic material which extended the cell life. It should be noted that the formation of an insoluble $\text{Mg}(\text{OH})_2$ passive layer at the surface of the anode in addition to the accumulation of Zn and Al at the boundary of the electrode and electrolyte lowers the nominal battery voltage and also its lifetime due to the polarization effect.

In another study [10], two strategies of alloying and coating were chosen to decrease the degradation rate of Mg anodes. A novel generation of biodegradable MZZ alloys was developed including Mg-3 wt% Zn-0.8 wt% Zr, coated with β -tricalcium phosphate (β -TCP) nanorods which served as the anodic material in Mg biobatteries. Simulated body fluid (SBF) and carbon were selected as the electrolyte and cathode, respectively. The results of polarization measurements revealed that upon coating β -TCP on the MZZ alloy, the corrosion potential increased from -1.72 to -1.39 V , the corrosion current decreased from 2.07

Figure 4 **a** Corrosion rate of MZZ alloy and β -TCP-MZZ alloy in SBF, **b** discharge behavior of the prepared anodes for Mg batteries at $100 \mu\text{A cm}^{-2}$ in SBF, surface morphology of **c** MZZ and **d** β -TCP-MZZ samples after immersion for 10 days in SBF, reproduced with permission from Ref. [10].



to $0.28 \mu\text{A cm}^{-2}$, and the average degradation rate decreased from 0.62 to $0.365 \mu\text{m h}^{-1}$. These findings demonstrate that the β -TCP coating protects the MZZ alloy matrix and enhances its corrosion resistance. The corrosion rate of the two samples is compared in Fig. 4a. It is apparent that the corrosion rate of MZZ is about two times higher than that of the β -TCP-MZZ sample for the first 4 h and thereafter the corrosion rates become closer to each other. The discharge plots of the fabricated Mg batteries are presented in Fig. 4b. The cell with the β -TCP-MZZ anode exhibited a voltage plateau of 1.05 V at a current of $100 \mu\text{A cm}^{-2}$ for almost 75 days, and the discharging continued for 360 h up to 0.7 V, while the delivered voltage of the MZZ alloy (without coating) anode was 1.05 V for only 625 h at the same current. The corrosion characteristics of the samples were also studied by observing their surface morphology via scanning electron microscopy (SEM) after immersion in SBF for 10 days, and the results are shown in Fig. 4c and d. A formation of many corrosion pits on the MZZ alloy surface is observed in Fig. 4c, while structural uniformity was maintained for the β -TCP-MZZ alloy. Evaluating the discharge curves of the Mg battery with the β -TCP-MZZ anode at different currents revealed that increasing the discharge current leads to a decrease in the delivered

voltage and the lifetime, which could be ascribed to the corrosion of β -TCP-MZZ during its working.

Finally, regarding the alloying strategy in Mg anodes, it should be noted that one of the disadvantages of alloying, by comparison with methods which do not change the overall composition of the alloy such as thermomechanical processing or heat treatment, is the probable increase in costs and environmental impacts [48]. Consequently, it would be a promising strategy to enhance the mechanical and degradation characteristics of binary Mg alloys by secondary processes such as heat treatment or thermomechanical processing instead of adding ternary or quaternary alloying elements. However, although such techniques have been successfully used to control the Mg alloy degradation rate for biodegradable implants, these techniques have not been used to enhance the properties of Mg-based anodes for biodegradable batteries.

Cathodes for biodegradable Mg batteries

As a key constituent of batteries, adopting an appropriate cathode material is crucial for a suitable battery performance [49, 50]. Currently, the main challenge in transient batteries is the lack of soluble

biocompatible electrode materials. An appropriate cathode for use in transient batteries should meet the following criteria: (1) suitable electrochemical properties as an energy storage device, (2) fully biodegradation, and (3) a controllable dissolution rate owing to different aims [51]. The conventional cathode materials in primary Mg batteries are either non-degradable, toxic, and/or unsuitable from an ecological viewpoint, and also, long-lasting current collectors are usually involved in their construction. These materials are not suitable for use in biodegradable batteries but by replacing them with biodegradable metals such as copper, iron, zinc, and molybdenum as the cathode material the construction of biodegradable batteries would be facilitated. Although these metallic cathode replacements lead to a decreasing output voltage and current, this may be compensated by increasing the working voltage and/or the current via respective strategies of integration of a series of cells and/or enlarging the surface area of the electrode. Furthermore, the metallic cathodes can also act as the current collector, which thereby eliminate the requirement for using conductive binders so that the overall assembly process becomes simplified [52]. In addition, metal oxides such as copper oxides, iron oxides, and zinc oxides are one of the newest classes of biocompatible materials for use in bio-devices [13].

Fe is the most studied cathode material for transient batteries because of its appropriate electrochemical properties. Degradation of Fe in water/biofluids is very slow and non-uniform because of the formation of insoluble oxide corrosion products and their accumulation on the surface which prevent further corrosion. The degradation rate of Fe foil in PBS solution (pH 7.4, 37 °C) is $0.0034 \mu\text{m h}^{-1}$.

Regarding investigations on cathode materials, fully biodegradable Mg batteries were constructed by using Mg foils as the anodes, Fe, W, or Mo foils as the cathodes, PBS as the electrolyte and polyanhydrides for packaging [52]. The discharge curves of the batteries at a current of 0.1 mA cm^{-2} are presented in Fig. 5a. The output voltages of $\approx 0.75 \text{ V}$, 0.65 V , and 0.45 V for at least 12 h were obtained using Fe, W, and Mo cathodes, respectively. The operating voltages are lower than for conventional Mg/AgCl or Mg/CuCl batteries ($\approx 1.5\text{--}1.6 \text{ V}$). In addition, depletion of the Mg active material limits the battery lifetime. By increasing the discharge current, the operating voltage decreases as observed in Fig. 5b for

the Mg–Mo battery. Figure 5c illustrates the degradation of Mg due to the operational reactions and also corrosion (self-discharging). The type of corrosion is pitting because of the non-uniform degradation of Mg. The appearance of white deposits on the foil surface is due to the formation of $\text{Mg}(\text{OH})_2$. Finally, four Mg–Mo individual cells were stacked in series to increase the output voltage. The stacked battery weight was nearly 3.5 g, containing 0.13 g Mo and 0.14 g Mg. Upon applying a discharge current density of 0.1 mA cm^{-2} , a stable voltage of $\approx 1.5\text{--}1.6 \text{ V}$ was obtained for almost 6 h. The transience of the battery is shown in Fig. 5d.

In another study [13], different combinations of full-cell chemistries were studied by using Mg as the anode, Cu, Fe, and Zn and their anodized forms as the cathode and PBS as the electrolyte. Current–voltage (IV) curves of various electrode combinations are shown in Fig. 6a. In theory, the voltage of the cell is the difference between the standard reduction potentials of the cathode and the anode, in which the more electronegative material would act as the anode. The observed operating voltages of various batteries, for example, $0.70\text{--}0.75 \text{ V}$ for the Mg–Cu battery at a current of 0.50 mA cm^{-2} , are considerably different from the theoretical cell voltage. This difference is attributed to the polarization effects and also the quite high applied discharge rates, particularly at low conductivities of the electrolyte.

Discharge profiles of the different full-cell combinations are shown in Fig. 6b. The fluctuations in the curves can be ascribed to the interactions of the electrode with the PBS solution, and also to the consumption of Mg during the discharge process and the occurrence of pitting corrosion. Furthermore, the formation of hydrogen gas and its adherence to the surface of the cathode and the resultant decreased reaction area is another reason for the observation of fluctuations in the voltage readings. More stable discharge outputs could be observed for the metallic cathodes by comparison with the metal oxide cathodes which is probably because of the pristine surface of the metallic cathodes. The discharge evaluations showed more complex curves for the batteries with metal oxide cathodes. For example, the profile of anodized zinc dropped to 0 V before becoming stable. Another observed behavior was the stripping and elimination of the anodized oxide from the metal surface upon prolonged application of a constant current [13].

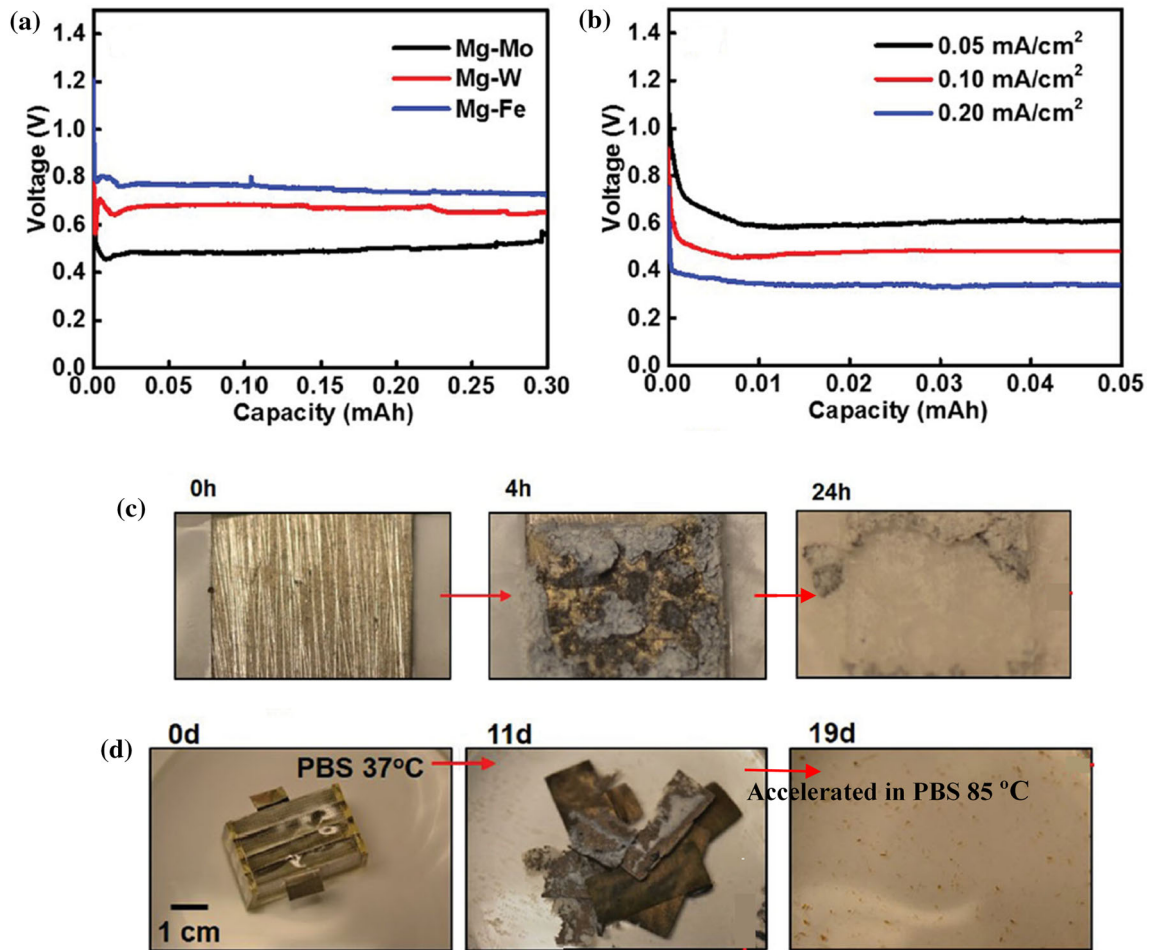


Figure 5 **a** Discharge curves of different battery combinations at 0.1 mA cm^{-2} , **b** discharge curves of the Mg-Mo battery at various currents, optical images of **c** an Mg foil during its degradation in

an Mg-Mo battery, and **d** dissolution of the stacked battery, reproduced with permission from Ref. [52].

Among different battery chemistries, the Mg-anodized Cu cell delivered the highest discharge voltage at a current density of 0.50 mA cm^{-2} . An average voltage of 1.40 V was achieved for the first hour of discharging, and then it dropped and was maintained at about 0.60 V for about 3 h followed by an unsteady voltage in the next hours. Further analysis was done on this full cell by applying a current density of 0.70 mA cm^{-2} (Fig. 6c) which is the current where the maximum power density was delivered based on the data of the I–V curves. Two plateaus could be observed suggesting an interaction of copper oxide with the system, followed by its removal and the exposure of metallic copper to the system. This mechanism was also identified when the applied current was 0.50 mA cm^{-2} and the life of the battery was shorter upon application of higher discharge currents. For this battery (Fig. 6c), the nominal

operation voltage was 1.26 V for the first 30 min and 0.63 V for the next 3.7 h, delivering an average specific capacity of 2.94 mAh cm^{-2} . It was reported that the anodized metal oxides provided further sites for evolution of hydrogen, resulting in an increase in the output voltage [13].

As another example of using metal oxides as the cathode, a high-performance fully biodegradable Mg–MoO₃ battery for an in vivo onboard power supply was developed [9]. The cathode material used in this work was MoO₃ with desirable biocompatibility at a controlled level and a high solubility in aqueous solution ($\approx 1 \text{ g L}^{-1}$). Figure 7a presents the schematic structure of the biodegradable Mg–MoO₃ battery in which Mg foil and sodium alginate (ALG–Na) hydrogel with phosphates served as the anode and the electrolyte, respectively. The cathode consisted of an MoO₃ film, prepared by mixing MoO₃

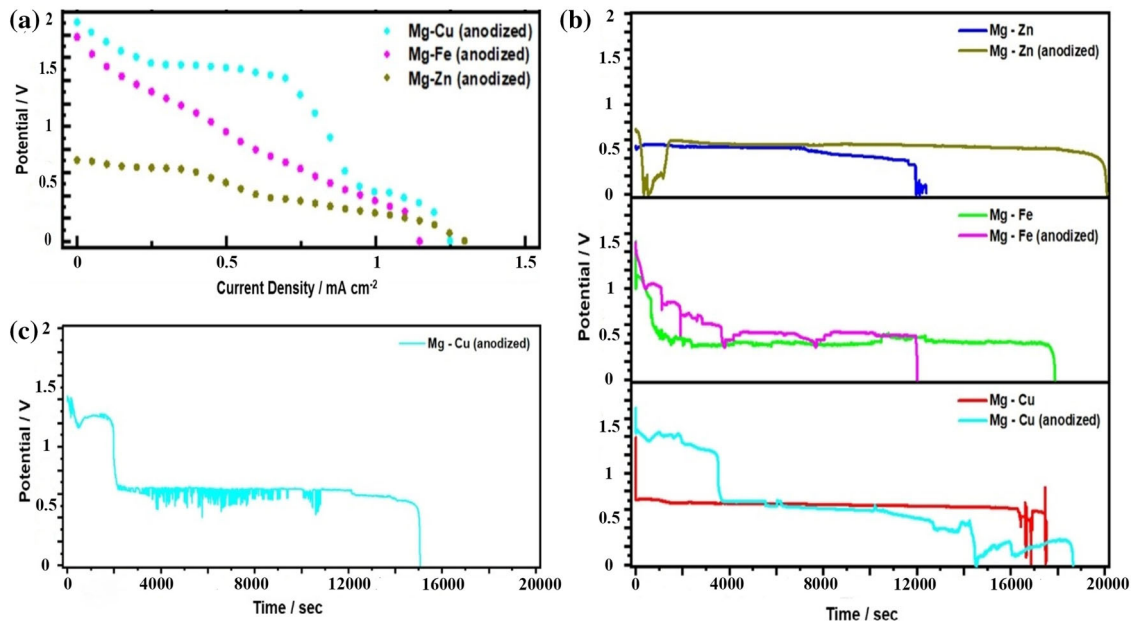


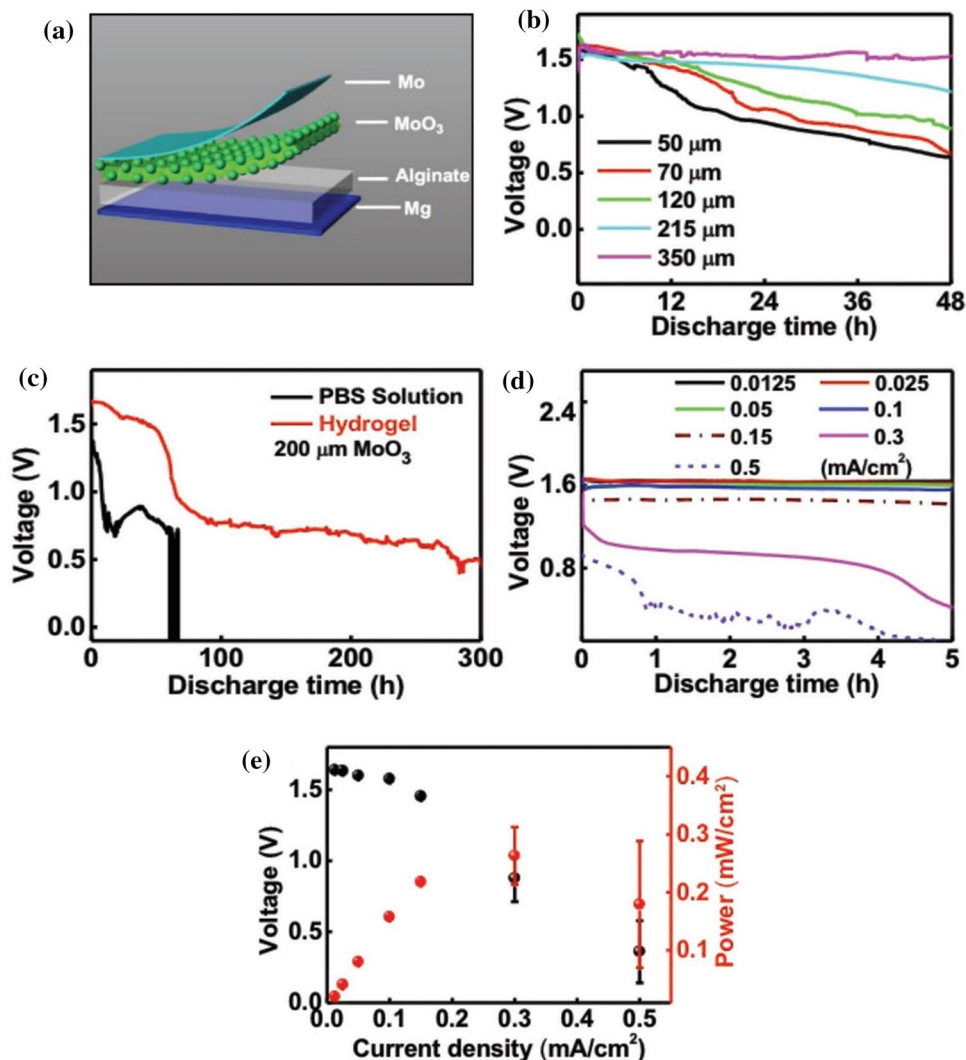
Figure 6 **a** Current–voltage curves, and **b** discharge profiles at 0.50 mA cm^{-2} for different electrode combinations, and **c** discharge curve of Mg-Cu oxide at 0.70 mA cm^{-2} , reproduced with permission from Ref. [13].

powders with biodegradable poly(lactic-co-glycolic acid) (PLGA) as the binder, on top of an Mo foil. The permeability of PLGA to the water in the electrolyte would result in the formation of a 3D porous structure and increasing the effective surface, and therefore, enhancing the battery performance. This study investigated the effect of MoO₃ film thicknesses on the discharge behavior of Mg–MoO₃ cells (Fig. 7b). The results revealed that thinner films would lead to a quicker decrease in the output voltage due to the faster depletion of MoO₃ materials. Figure 7c shows the effect of the electrolyte on the discharge performance of the cells. By using the hydrogel electrolyte, the lifetime of the battery is prolonged to 300 h by comparison with 60 h in the case of the PBS electrolyte. The reason is probably associated with the restricted corrosion of Mg and dissolution of MoO₃ in the hydrogel electrolyte. The influence of different current densities on the discharge behavior of the cells is shown in Fig. 7d. It is apparent that up to 0.15 mA cm^{-2} and a high voltage of $\approx 1.5 \text{ V}$ could be achieved, and by further increasing the current density the voltage dropped considerably. Figure 7e presents the corresponding voltage by averaging the data from the first 5 h and the power. The voltage (up to 1.6 V) and power (0.27 mW cm^{-2}) delivered by the battery will satisfy the requirements of most of the ultralow-power implantable devices which are

typically in the range of $\approx 0.5\text{--}1.6 \text{ V}$ and $\approx 10\text{--}1000 \mu\text{W}$. The potential between MoO₃ and Mg electrodes and the potential of each electrode vs. Ag/AgCl were examined through a galvanodynamic scan. The potential of Mg versus Ag/AgCl remained relatively stable around -1.6 V , while the potential of the MoO₃ electrode decreased upon increasing the current density and displayed a two-stage behavior.

As shown in Fig. 8a, this battery can light up a light-emitting diode (LED) with a threshold voltage of $\approx 1.5 \text{ V}$ and it can maintain its function in PBS for up to 16 h (Fig. 8b), indicating the ability of the battery configuration to be used in aqueous environments. The dissolution stages of the transient battery in PBS are illustrated in Fig. 8c. Most of the battery components including Mg, alginate, polyanhydride, and MoO₃/PLGA completely degrade within 9 days, except for Mo which completely dissolved within another 10 days at an elevated temperature ($85 \text{ }^\circ\text{C}$). Furthermore, the biocompatibility of the cathode was examined, relating to the MoO₃ paste encapsulated by a PLGA film, and the results are shown in Fig. 8c and d. The cell viability of the PLGA samples in the absence and presence of MoO₃ was not significantly different, and both the viability data and the fluorescent images revealed the desirable biocompatibility of the MoO₃ component [9].

Figure 7 **a** The schematics of the biodegradable battery structure, **b** discharge curves for various thicknesses of MoO₃, at the current of 25 μA cm⁻², **c** discharge behavior in PBS and hydrogel electrolytes, at the current of 25 μA cm⁻², **d** discharge performance at various discharge current densities, **e** battery voltage and power versus current density, reproduced with permission from Ref. [9].



Additionally, *in vivo* tests revealed a full degradability of the whole battery. This battery, with dimensions of $\approx 1.2 \times 1.2 \times 0.4$ cm and a shape similar to the cell illustrated in Fig. 8b, was placed in the subdermal region of a rat (Fig. 8e). Traces of battery components could be observed after 2 weeks, while the complete degradation occurred after 4 weeks of implantation. The *in vivo* degradation rate was faster than that of the *in vitro*, possibly because of the difference in the aqueous environments and circulation conditions.

To conclude, the prepared fully biodegradable battery could provide a high stable voltage up to 1.6 V, an appropriate capacity of 6.5 mAh cm⁻² and a lifetime up to 13 days. Application of MoO₃ as the cathode material resulted in increasing the output voltage by a factor of 3 by comparison with that of an Mg–Mo battery, and also, it was higher than most of

the previously reported biobatteries. The cathodic reaction was assumed to be as follows [9]:



In practice, a reduction of Mo(VI) at the cathode led to an increase in the output voltage of the fabricated biobattery.

In a very recent study [12], the MoO₃–MoS₂ composite electrode was synthesized on Mo foil. For this purpose, MoO₃ flakes were grown on an Mo surface via an anodizing process and then the oxidized Mo foil was hydrothermally treated in thiourea solution at 180 °C for 24 h to obtain the MoO₃–MoS₂ composite through the sulfidation reaction. The primary battery was assembled using the Mg foil as the anode, the MoO₃–MoS₂ electrode as the cathode, and alginate–Na film as the electrolyte. For encapsulating,

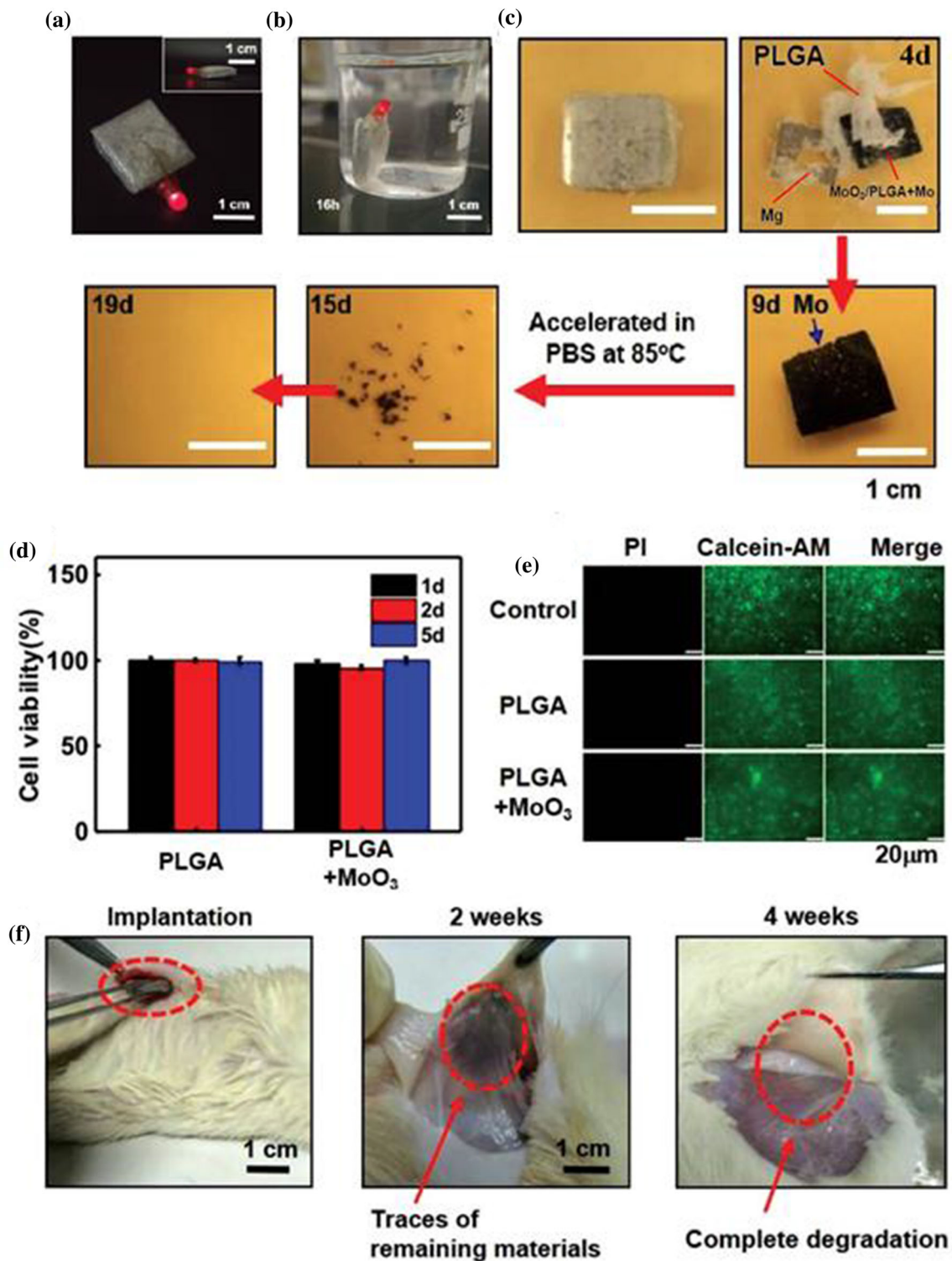


Figure 8 a A red LED powered by the battery and the side view is shown in the inset, b the battery powers the LED in PBS solution for 16 h, c optical images of the dissolution behavior of the biodegradable battery in PBS, d cell viability of MoO₃/PLGA and PLGA films during 1, 2, and 5 days calculated as the fraction of

total living cells, e fluorescent images showing the cell viability with green (calcein-AM)/red (PI) denote live/dead L-929 cells, respectively, f in vivo degradation study of the battery in the subcutaneous area of Sprague–Dawley (SD) rats, reproduced with permission from Ref. [9].

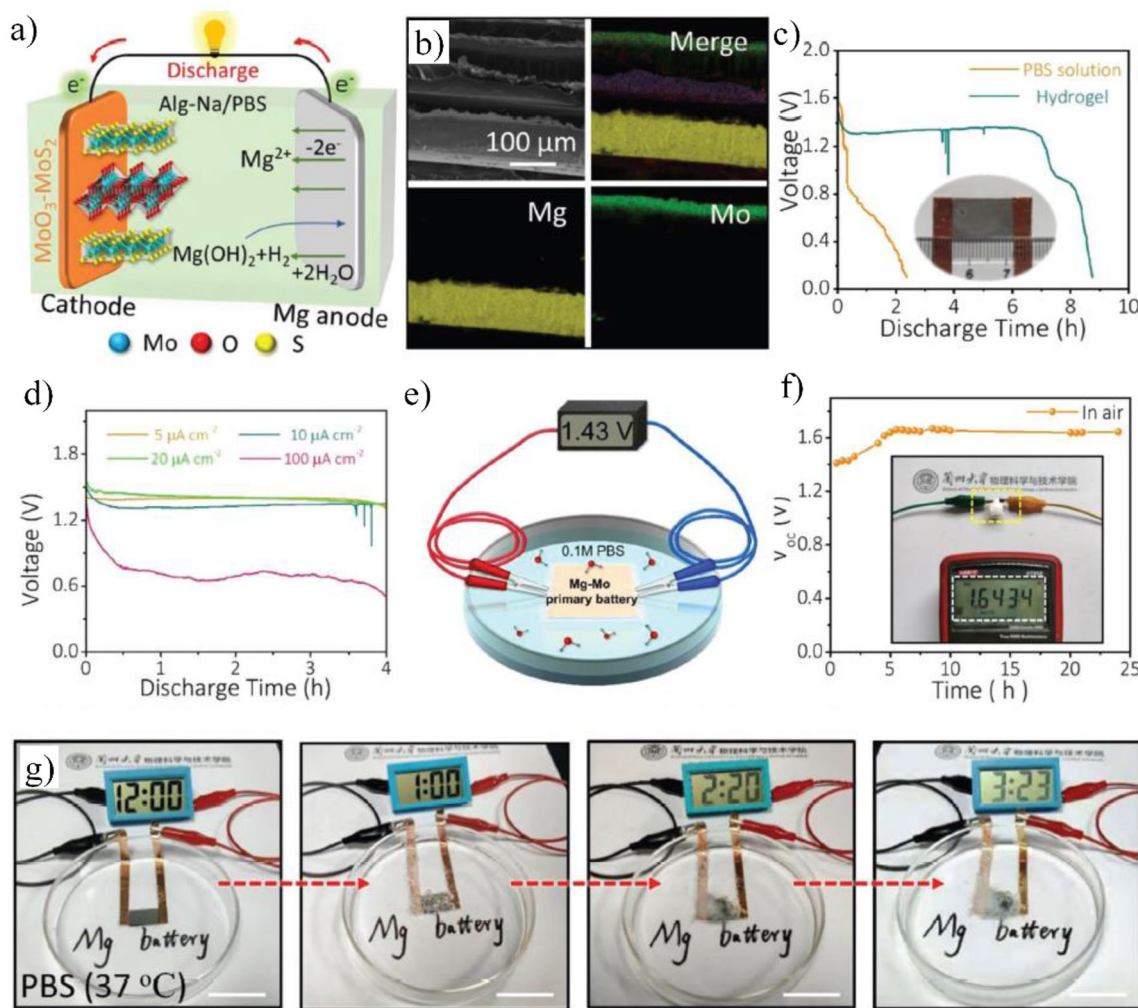


Figure 9 Characteristics of Mg/MoO₃-MoS₂ biodegradable primary battery; **a** the working mechanism, **b** SEM and elemental mapping images of the battery from the cross-section, **c** the discharge behavior of the battery at different electrolytes at 10 μA cm⁻², **d** the discharge performance of the battery at various

discharge currents, **e** illustration of the encapsulated battery in PBS solution, **f** monitoring the OCV in air, **g** optical images showing the operating process of the battery in PBS to power an electric watch (scale bar, cm), reproduced with permission from Ref. [12].

the PLGA was coated on the battery to prevent (1) the electrolyte evaporation in air, (2) the electrolyte ions diffusion to the surrounding solution, and (3) structural failure and dissolution of battery materials to ensure a stable battery operation and prolong its lifetime. The working mechanism of the constructed battery is illustrated in Fig. 9a, and its sandwich structure can be observed in Fig. 9b. The discharge behavior of the assembled battery in different electrolytes is compared in Fig. 9c, and the longer lifetime of the battery in hydrogel electrolyte could be detected, in comparison with the PBS solution. In this figure, a stable discharge plateau above 1.4 V for more than 7 h at a current of 10 μA cm⁻² is observed.

The reason could be ascribed to the limited side reactions such as corrosion and passivation of Mg foil and dissolution of electrode materials. Figure 9d represents the discharge performance at different currents. A flat voltage plateau of 1.4 V was reached after a short time of applying the discharge current. Also, a capacity of 0.45 mAh cm⁻² was delivered at the current of 100 μA cm⁻². The schematic of the encapsulated battery in PLGA is shown in Fig. 9e, and the open-circuit voltage (OCV or V_{oc}) versus time is plotted in Fig. 9f, showing that the OCV of 1.64 V could be delivered for more than 25 h, thereby demonstrating the low self-discharge rate of the battery. Figure 9g shows there is the capability of the

assembled battery to power an electronic clock in PBS solution for 3 h, and after that a failure of the battery structure occurred. Improving the design of the encapsulating material would increase the lifetime of the battery. Finally, the delivered power of the constructed battery is satisfying for biomedical implant applications.

In a recent study on the development of modern electronics [53], a new method was developed for the fabrication of stretchable and biodegradable batteries. These batteries are indispensable for applications in which the electronics are in close interaction with the user or for powering soft robotic devices. The battery architecture was composed of MoO_3 paste as the cathode, Mo foil as the current collector, and Mg foil as the anode. Fabrication of stretchable metal electrodes was performed by kirigami patterning which is a favorable method for engineering reversible elasticity and stretchability in rigid electrodes when combined with an appropriate support material. For example, metallic and metal oxide conductors become stretchable when deposited on a patterned plastic substrate. Calcium alginate electrolyte gel was used as a stretchable separator for the battery. The schematic of the battery design is illustrated in Fig. 10a, a photograph of the prepared battery in Fig. 10b, and the fabrication steps of the stretchable electrodes via kirigami patterning in Fig. 10c [53].

The mechanical characterizations of the fabricated battery demonstrated a reversible elasticity up to 35% for the batteries upon uniaxial stretching and 20% upon extending biaxially. Moreover, the batteries sustained more than 1000 strain cycles with only a slight increase in resistance. The electrochemical measurements revealed the achievement of a peak power output of $196 \mu\text{W cm}^{-2}$ and an energy density of 1.72 mWh cm^{-2} [53].

Figure 10d shows the degradation behavior of the battery in a physiological solution at body temperature (37°C). Among the biometals, the degradation rate of Mg is comparatively fast, while it is much slower for Mo. An Mg foil with a thickness of $50 \mu\text{m}$ ($10 \times 5 \text{ mm}$) requires 10.5 h to become completely dissolved in a PBS solution, whereas 70% degradation of Mo foil in PBS needs 18 days. The degradation rate of the battery can be accelerated by varying the influencing factors such as the presence of salts and enzymes, the pH of the dissolution environment, and its temperature. Concerning the biopolymers, their degradation rate depends on several parameters such

as their chemistry, molecular weight, and terminal groups. In a specific polymer, the degradation rate is further dependent upon the synthesis method and polymerization degree [53].

The dual cells fabricated by this new method were used to power an LED under strong deformations, such as stretching and twisting (Fig. 10e). It was revealed that the applicability of the biodegradable, stretchable, and soft batteries in flexible electronics may be achieved easily. Furthermore, the stretchable batteries were used for powering an on-skin biomedical sensor patch in order to monitor the concentration of sodium in sweat before a workout, as shown in Fig. 10f. The fabrication scheme developed in this study is suitable for achieving biodegradable power sources for soft electronics [53].

In addition to these Mg batteries, the fabrication of a partially biodegradable Mg-air biobattery was reported in another study [54] by using an Mg alloy anode and a silk fibroin–polypyrrole (SF-PPy) film cathode in a PBS electrolyte. The prepared Mg-air battery delivered a discharge capacity of up to 3.79 mAh cm^{-2} at an applied current of $10 \mu\text{A cm}^{-2}$, it offered a specific energy density of 4.70 mWh cm^{-2} , and it degraded after 15 days. This novel battery could be an appropriate choice for the development of biodegradable batteries. It is important to mention that silk materials could be used in the fabrication of various components of biodegradable batteries [55]. For example, a transient zinc-ion battery was constructed by using gelatin–silk fibroin film as the electrolyte [56].

Electrolytes for biodegradable Mg batteries

The electrolyte is a crucial constituent of an energy storage device, and it plays an important role in determining various characteristics of the battery such as lifetime, operating voltage, and specific capacity. Liquid electrolytes have been used extensively in electrochemical devices for decades due to their outstanding conductivities. However, safety problems are always associated with them. In order to eliminate the safety limitation, solid electrolytes are a promising candidate for high energy density energy storage [57]. Solid electrolytes are generally divided into two classifications of inorganic solid electrolytes and polymer electrolytes. Although some inorganic solid electrolytes show a high ionic

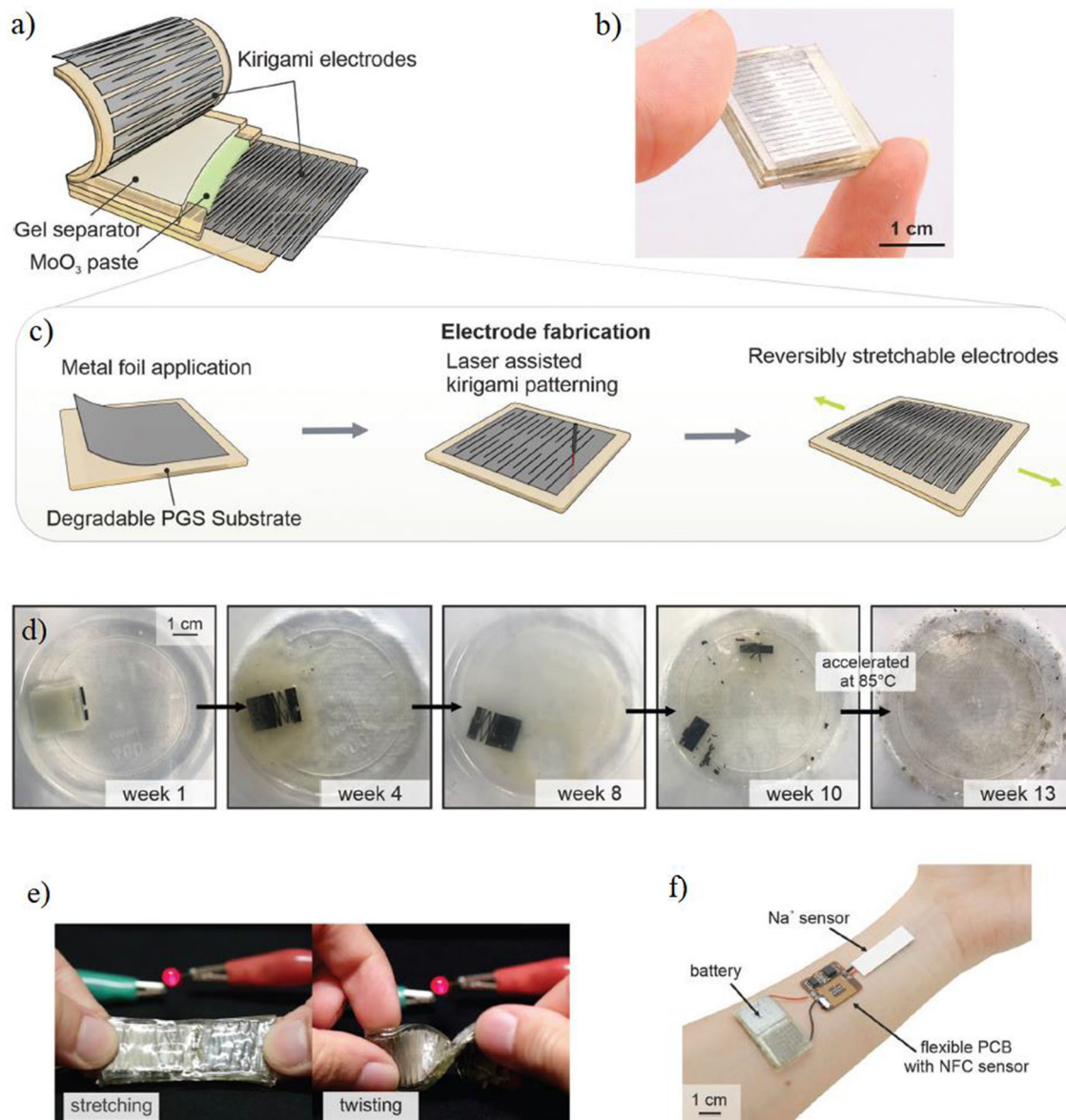


Figure 10 **a** Schematic of a stretchable cell, **b** photograph of a fabricated stretchable biodegradable battery, **c** fabrication process of stretchable electrodes, **d** degradation stages of the battery in PBS, **e** an LED powered by dual-cells while being stretched and

twisted, **f** photograph of using the stretchable battery for powering an on-skin sensor patch to monitor the concentration of sodium in sweat, reproduced with permission from Ref. [53].

conductivity as well as organic liquid electrolytes, their rigid nature would result in several solid–solid interface problems and a low compatibility of electrode–electrolyte interface which is generally the bottleneck for the development of solid-state batteries. The interface issues could be overcome by the use of polymer electrolytes owing to their lightweight, low flammability, flexibility in size, ease of processability, good interfacial contact, and compatibility with electrodes [58, 59].

Currently, polymer electrolytes are of significant interest among different types of electrolytes, and they could be divided into categories of solid polymer electrolytes (SPEs), gel polymer electrolytes (GPEs), and composite polymer electrolytes (CPEs). SPEs are the most studied electrolyte in batteries due to their simple preparation and outstanding chemical, mechanical, and thermal stabilities [60]. SPEs have lower conductivities than liquid polymer electrolytes, but they are preferable for use in

electrochemical devices due to their higher safety and good thermal, mechanical, and electrical stability. Also, the use of SPEs facilitates the integration of battery components to form a thin and compact device that may be easily implanted [61–63]. GPEs are also extensively studied in batteries, and they represent an intermediate state between liquid electrolytes with high flowability and dry SPEs without any liquid [59].

In practice, modern global environmental consciousness is a driving force toward the development of biopolymers. Thus, biopolymers are polymers that could be obtained from living organism and are degradable naturally and they are preferred to synthetic polymers because they are carbon neutral, cheap, environmentally friendly, and easily biodegradable since the presence of functional groups in their backbone provides possibilities for modifying the polymer properties [64, 65]. Some of the solid biopolymer electrolytes used in the fabrication of electrochemical devices are carrageenan, pectin, cellulose, starch, chitosan, and agar-agar. In order to increase the polymer electrolyte conductivity, commonly the polymer is complexed with various salts. Various ions in different salts such as H^+ , Ag^+ , Li^+ , Na^+ , K^+ , Mg^{2+} , F^- , and O^{2-} can be used as mobile species to increase the ionic conductivity of solid electrolytes. Lithium salts are used in the current commercially available batteries although using magnesium in batteries is recommended because it is cheaper, safer, and more abundant with a higher volumetric capacity than lithium and also it is not influenced by anodic dendrite formation [62, 66–68]

Although PBS is the most evaluated electrolyte in biodegradable Mg batteries due to its similarity to the human body fluid, there have been some reports of magnesium batteries constructed from biopolymer electrolytes which will be reviewed here. Regarding the development of biopolymers based on I-carrageenan with magnesium, in a previous study [62] an I-carrageenan membrane with various concentrations of magnesium perchlorate was prepared via a solution casting method. The FTIR analysis revealed the formation of complex between the polymer and the salt. The conductivity of pure I-carrageenan was $5.90 \times 10^{-5} \text{ S cm}^{-1}$ and by adding 0.6 wt% of magnesium perchlorate the conductivity increased to $2.18 \times 10^{-3} \text{ S cm}^{-1}$. The optimum electrolyte was used in the construction of a primary magnesium battery and it was placed between the Mg as the anode and the

cathode pellet which was made up of a mixture of MnO_2 with graphite. The OCV of the battery was stable for 20 days at a value of 1.96 V. In another study [67], an Mg ion-conducting biopolymer electrolyte was prepared by complexing I-carrageenan with different amounts of magnesium nitrate salt via solution casting. The nature of the prepared material was amorphous which is beneficial for ionic transport enhancement. The polymer with 0.4 wt% magnesium nitrate showed the highest ionic conductivity of $6.1 \times 10^{-4} \text{ S cm}^{-1}$ among the prepared samples. The constructed Mg battery had an OCV of 1.92 V for 15 days. In a very recent study [69], kappa carrageenan with various wt% ratios of $Mg(ClO_4)_2$ was prepared by a solution casting method and then used as the biodegradable solid polymer electrolyte film in a primary Mg-ion battery. The highest conductivity of $6.28 \times 10^{-3} \text{ S cm}^{-1}$ was observed for the biopolymer with 0.5 wt% $Mg(ClO_4)_2$. The Mg-ion battery constructed from this electrolyte showed an OCV of 2.36 V.

With reference to the use of pectin, biopolymer electrolytes were developed by fabricating different amounts of pectin with various concentrations of magnesium chloride salt via a solution casting method [70]. A biopolymer containing 30 M wt% pectin: 70 M wt% $MgCl_2$ showed the highest conductivity of $1.14 \times 10^{-3} \text{ S cm}^{-1}$ between the prepared samples. The fabricated Mg battery offered a stable OCV of 2.1 V for 11 days. These prepared biodegradable polymers are effective electrolytes for Mg batteries.

Concerning the use of cellulose, in a very recent study [60] biodegradable electrolyte films were prepared using different amounts of methyl cellulose and magnesium acetate tetrahydrate by a solution casting method. The maximum ionic conductivity at room temperature ($2.61 \times 10^{-5} \text{ S cm}^{-1}$) was observed for the film doped with 25 wt% salt. The electrochemical stability window for this sample was 3.47 V. Furthermore, a primary battery was constructed from this electrolyte, delivering an energy density of 3.405 Wh kg^{-1} , and showing an OCV of 2.01 V which decreased over time, probably due to the reactions occurring between the cell constituents, and reached a constant value of $\sim 1.88 \text{ V}$ after 1500 min. In another study [63], solid biopolymer electrolyte films based on methyl cellulose and $Mg(NO_3)_2 \cdot 6H_2O$ of various amounts were fabricated via solution casting. The highly amorphous film prepared with 25 wt% of

the salt showed the highest ionic conductivity of $1.02 \times 10^{-4} \text{ S cm}^{-1}$ among the prepared samples. The electrochemical stability window of this film was 3.23 V which is appropriate for energy storage applications. In another investigation [71], a solid polymer electrolyte was developed by using cellulose acetate with various concentrations of magnesium nitrate. The results of AC impedance spectroscopy revealed that the electrolyte with 40 wt% of salt showed the highest ionic conductivity of $9.19 \times 10^{-4} \text{ S cm}^{-1}$. Furthermore, biodegradable electrolyte films of carboxymethyl cellulose doped with various wt% of sodium bromide were examined and the electrolyte with 20 wt% of salt showed the maximum ionic conductivity of $9.19 \times 10^{-4} \text{ S cm}^{-1}$ [72].

In a recent study [73], solid biodegradable polymer electrolytes were fabricated from potato starch (PS) mixed with polyvinyl pyrrolidone (PVP) and magnesium sulfate (MgSO_4) with various weight ratios through solution casting. An electrolyte with a PS/PVP/ MgSO_4 weight ratio of 35:35:30 showed the highest ionic conductivity of $4.2 \times 10^{-4} \text{ S cm}^{-1}$ among the prepared materials. For this sample, the electrochemical measurements revealed a stable voltage and also stable redox potentials, suggesting a suitable capability of this biopolymer electrolyte for use in biodegradable magnesium batteries.

Ionic liquids (ILs) are molten salts at room temperature and are currently used as solvents or additives for biopolymers. Ionic liquids are typically inorganic/organic salts consisting of inorganic anions and organic cations with a melting point below 100 °C. Some advantages of ionic liquids are their non-volatile, non-toxic, and non-flammable nature, their high thermal stability, wide electrochemical potential window, high ionic conductivity, very low vapor pressure, and in some cases good biocompatibility [74, 75].

Regarding the application of biocompatible ILs, various amounts of choline nitrate ($[\text{Ch}][\text{NO}_3]$) were introduced into silk fibroin (SF) to fabricate a biodegradable ion-conducting membrane electrolyte. An SF- $[\text{Ch}][\text{NO}_3]$ with a weight ratio of 1:3 was selected as the optimum sample due to its superior ionic conductivity and mechanical properties. An Mg alloy, AZ31, was used as the sacrificial anode in the prepared biodegradable battery. Increasing the thickness of the AZ31 film from 100 to 500 nm led to longer discharge times. The battery with a 500 nm AZ31 film offered a specific capacity of 0.06 mAh

cm^{-2} at a current of $10 \mu\text{A cm}^{-2}$. The biodegradation behavior of the battery with a size of $3.6 \times 2.7 \times 0.017 \text{ cm}$ at 37 °C in buffered protease solution is shown in Fig. 11. It is apparent that the whole battery decomposed after 45 days [74].

Liquid and solid electrolytes are extensively used in batteries. Although they have some disadvantages such as leakage, reactivity, and low safety for the liquid electrolyte and a low ionic conductivity for the solid polymers. Gel polymer electrolytes (GPEs), composed of polymer matrices, inorganic/organic salt, and a plasticizer, are beneficial because of their shape flexibility, interface stability, and fabrication integrity. Aprotic plasticizers with low molecular weight such as propylene carbonate, polyethylene glycol, and diethyl carbonate are the common plasticizers in the fabrications of GPEs. Although the conductivity of these GPEs is sufficiently high for battery applications, their thermal stability and electrochemical performance are not satisfactory. To overcome these limitations, ionic liquids are a suitable option to be used as plasticizers in the fabrication of GPEs [75]. In this respect, different electrolytes were prepared with the use of biodegradable polyvinyl alcohol (PVA), magnesium trifluoromethanesulfonate ($\text{Mg}(\text{Tf})_2$) as the dopant salt, and 1-ethyl-3-methylimidazolium trifluoromethanesulfonate (EMITf) as the ionic liquid, with various concentrations via solution casting [75]. The ionic liquid should be selected according to the principle that both salt and ionic liquid contain the same anions which is favorable for the electrical and electrochemical properties of GPEs. After preparing the GPEs, the highest value of ionic conductivity ($2.10 \times 10^{-4} \text{ S cm}^{-1}$) was observed for an 85PVP:15 $\text{Mg}(\text{Tf})_2$:15-EMITf electrolyte, while in the absence of ionic liquids the electrolyte conductivity was $2.36 \times 10^{-6} \text{ S cm}^{-1}$. Also, the optimum sample showed a wide electrochemical stability window ($\sim 5 \text{ V}$) and appropriate mechanical properties, suggesting that it has a high capability for use in Mg-ion batteries.

Summary and perspectives

As an emerging technology, transient electronics which degrade after their operational task are on track for fast development. Their biological application in the human body excludes the second surgery which is generally required for removing the device,

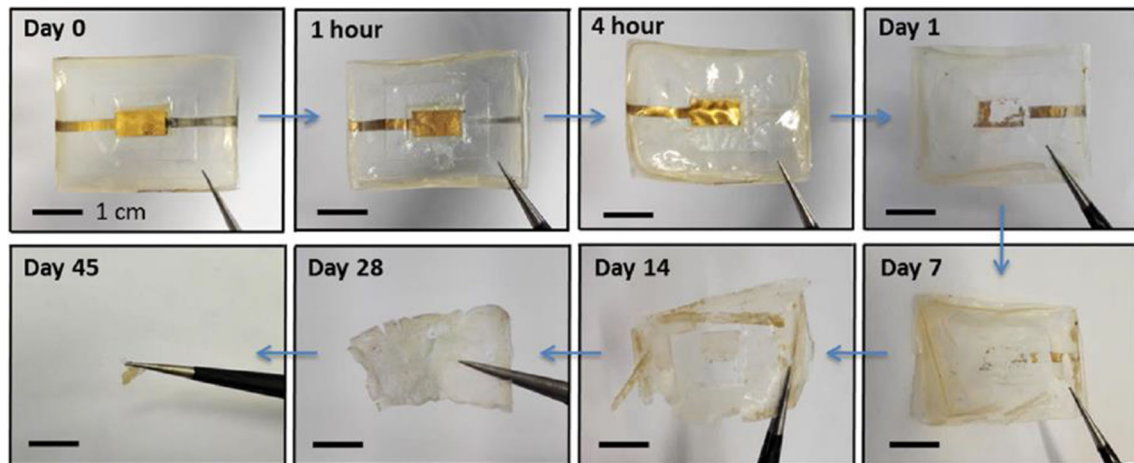


Figure 11 The optical images representing the biodegradation profile of the prepared thin film Mg battery using silk fibroin-ionic liquid electrolyte in buffered protease solution at 37 °C, reproduced with permission from Ref. [74].

and consequently, their use is attractive for preventing probable surgical side infections. Furthermore, the development of biodegradable forms of electronics is a suitable way toward the development of environmentally friendly electronics. This study provides an overview of recent advances in the development of biodegradable electrode materials and electrolytes for biodegradable Mg batteries. Most of the literature in this field has been dedicated in the development of primary (non-rechargeable) batteries. The shortcomings associated with Mg-based biodegradable batteries generally include a relatively low working voltage, power density, stability, and a relatively short lifetime which are mainly attributed to the performance of electrode materials. In this section, the main points of the literature review are summarized and then the discussion centers on the remaining challenges and the potential strategies for addressing these challenges.

1. Magnesium and its alloys are the most common anode material in biodegradable Mg batteries due to their attractive electrochemical, mechanical, and physiological properties. It is suggested that oxidation of Mg is the reaction occurring at the anode and accordingly Mg is consumed during the battery discharging process. Since Mg ions are one of the most abundant cations in the human body, this high physiological tolerance supports the application of Mg as the sacrificial anode material. The Mg anodes suffer from high degradation rates, but there are several strategies to overcome this shortcoming including alloying, coating, heat treatment, and thermomechanical

processing of Mg and its alloys. Regarding alloying strategy, only non-toxic biocompatible elements can be used as alloying components and this restricts the range of available elements. To date, only the Mg–Zn, AZ31 (Mg–Al–Zn), and MZZ (Mg–Zn–Zr) alloys have been studied in transient Mg batteries. Therefore, a wide range of alloying systems should be investigated with the objective of reducing the degradation rate of the Mg metal. Regarding the coating strategy, only the MZZ alloy coated with β -tricalcium phosphate has been studied as the anode in biodegradable Mg anodes. Consequently, other coating systems could be applied on Mg and its alloys for this purpose. Moreover, heat treatment and thermomechanical processing strategies have not yet been studied for controlling the degradation rate of Mg alloys in biodegradable Mg anodes and consequently these are important topics for future research. Furthermore, combinations of these strategies could be adopted to improve the degradation rate of Mg anodes.

An important point which should be considered is that the microstructural changes of the Mg metal associated with the application of these strategies would also affect other properties of Mg, such as their mechanical and biological performance, in addition to the degradation behavior. Accordingly, methods must be chosen which improve the overall performance of the biodegradable batteries and this choice needs further investigations.

2. The main challenge in transient batteries is the lack of soluble biocompatible materials. Generally, the reactions occurring at the cathode surface are either hydrogen evolution or oxygen reduction. The conventional biodegradable metals used as the cathode material in biodegradable Mg batteries include Cu, Fe, Zn, W, and Mo. These metallic cathodes can also act as the current collector which eliminates the need for using conductive binders and thus simplifies the assembly process. Furthermore, metal oxides such as copper oxide, iron oxide, zinc oxide, and especially molybdenum trioxide are one of the cutting-edge classes of biocompatible materials used as the cathode in these batteries. It was revealed that using metal oxides as the cathode material leads to an increase in the battery output voltage. In the case of using molybdenum trioxide, it was suggested that the reduction of Mo(VI) at the cathode surface is the reason for the increase in the output voltage. Further investigations are now needed to more fully understand the precise mechanisms and requirements of a suitable cathode material for developing new promising cathodes.
3. Concerning the electrolyte component, PBS is the most evaluated electrolyte in biodegradable Mg batteries due to its similarity to human body fluid. However, an important affecting parameter on the lifespan and overall size of the biodegradable batteries is the volume of the required liquid electrolyte. Using solid polymer electrolytes facilitates the integration of battery components to form a thin and compact device that may be easily implanted in the body. Biopolymers are carbon neutral, inexpensive, and ecologically friendly with a biodegradation behavior, and therefore, they are preferable to synthetic polymers. The solid biopolymer electrolytes investigated in transient Mg batteries include carrageenan, pectin, cellulose, and starch, complexed with various salts having different concentrations. Since solid electrolytes suffer from low ionic conductivity, complexing is one of the most important strategies to overcome this weakness. Using gel polymer electrolytes is another strategy to overcome the drawback of solid electrolytes although there are to date only a limited number of studies on solid and gel polymer electrolytes.

Moreover, previous studies have rarely reported the performance of the developed biodegradable polymers as the electrolyte in Mg batteries. Therefore, detailed electrochemical measurements are needed to evaluate the performance of the potential polymers as the electrolyte in biodegradable Mg batteries.

Overall, biodegradable batteries are particularly remarkable for *in vivo* applications, but so far only limited experimental evaluations have been reported in this respect. It is important to mention that *in vivo* degradation behavior can be very different from the *in vitro* behavior (even with the same materials), which can be mainly ascribed to the complications in mimicking the complex physiological conditions *in vitro*. The battery constituents interact/ behave in real biological environments differently, and *in vivo* studies are required to know the maximum dosage of materials that is allowed without threatening the human body. For example, the formation of hydrogen gas during the battery degradation may result in unwanted tissue necrosis and blood clotting, while *in vitro* investigations may only confirm the degradation of the battery. Consequently, extensive *in vivo* experiments under biological conditions and studying both the battery performance and its interaction with biotic systems (tissues and organs) are required in order to effectively develop biodegradable Mg batteries for use in healthcare and green electronics.

Author contributions

NH contributed to conceptualization, writing—original draft, and writing—review and editing. TGL was involved in writing—review and editing.

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