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Stabilizing moisture-sensitive thermoelectrics via anodic protection

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MAIN TEXT

The rapidly growing demand for thermal management in advanced fields such as integrated circuits, optical communications, and low-temperature medicine has accelerated the development of efficient, quiet, and compact thermoelectric cooling devices[1-3]. However, conventional Bi_2Te_3-based systems are constrained by the scarcity of tellurium (Te) and limited performance at elevated temperatures due to their narrow band gap and associated bipolar effects, which lead to rapid efficiency degradation above about 400 K[4,5]. In contrast, $Mg_3(Sb, Bi)_2$ exhibits excellent thermoelectric performance over a broad temperature range (300-773 K), along with advantages such as low cost, light weight, and good mechanical robustness[6,7]. These attributes make them strong candidates to replace n-type Bi_2Te_3 alloys in next-generation devices[8]. However, their practical application is severely hindered by a critical challenge: the high chemical reactivity of Mg makes these materials vulnerable to moisture-induced corrosion, leading to rapid performance degradation[9,10]. Existing protection strategies, such as using organic, HfO_2, Al_2O_3, and Mg-Mn alloy coatings, provide only partial mitigation and remain susceptible to failure due to cracking or delamination[11,12]. More importantly, they fail to address corrosion during material storage and processing. Therefore, developing a fundamental solution that ensures intrinsic humidity stability while preserving high thermoelectric performance throughout the entire material lifecycle remains an urgent priority.

In a study published in *Nature Materials*, Sui and co-workers introduced an innovative sacrificial anodic protection method into the design of $Mg_3(Sb, Bi)_2$ materials and devices[13]. Through multi-objective optimization, they screened and constructed *in situ* uniformly distributed multi-scale $Mg_{17}Al_{12}$ anode second phases within the $Mg_3(Sb, Bi)_2$ matrix [Figure 1]. Owing to its lower equilibrium potential, $Mg_{17}Al_{12}$ forms micro-galvanic cells that preferentially corrode, thereby providing thermodynamic cathodic protection to the matrix. Meanwhile, corrosion-induced Mg/Al hydroxides and oxides generate a self-healing hybrid passivation layer, which further

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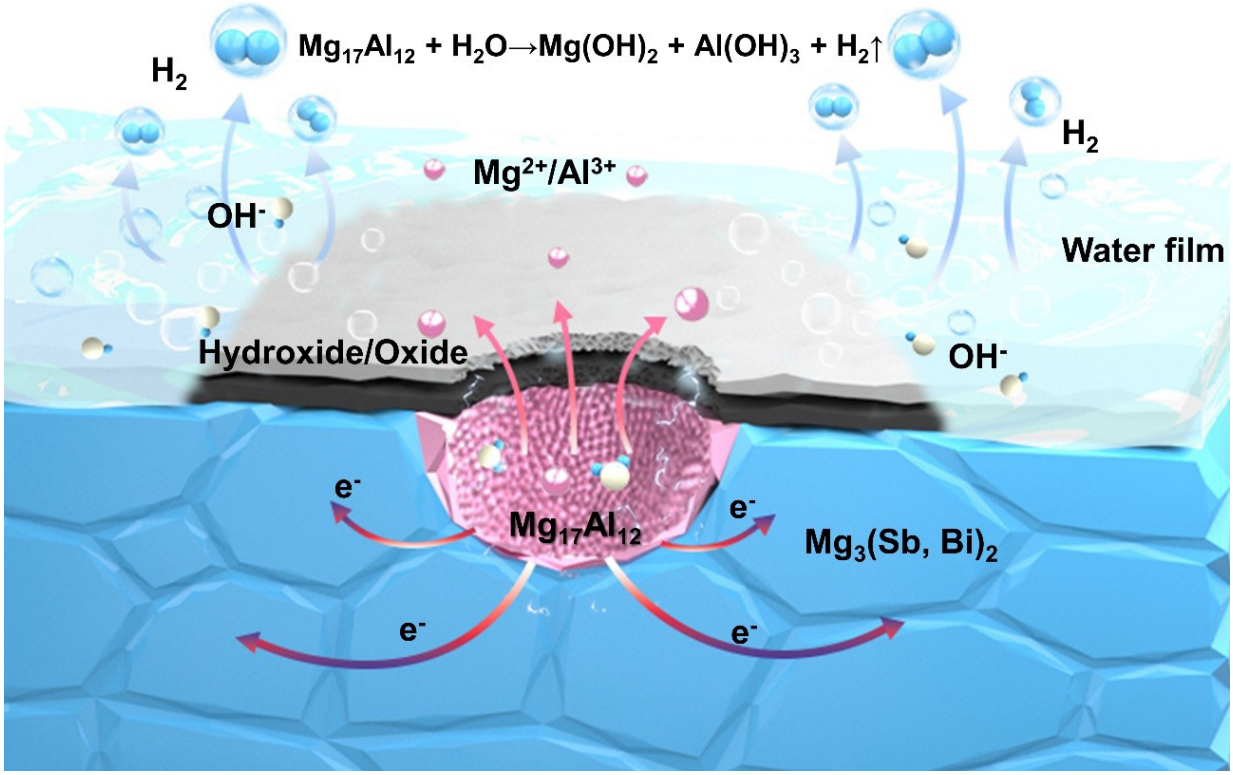

Figure 1. Schematic of the autonomous anodic protection mechanism. The $Mg_3(Sb, Bi)_2$ matrix, $Mg_{17}Al_{12}$ secondary phase, and the condensed water film collectively constitute a self-driven micro-galvanic cell, functioning as the cathode, sacrificial anode, and local electrolyte, respectively. Driven by the electrochemical potential gradient, the anodic $Mg_{17}Al_{12}$ undergoes preferential oxidation, releasing Mg^{2+} and Al^{3+} ions while concurrently transferring electrons to the cathodic matrix. This electron flux facilitates the reduction of water at the $Mg_3(Sb, Bi)_2$ surface into H_2 gas and OH^- ions. The resultant local supersaturation triggers the precipitation of dense Mg/Al-based oxides and hydroxides, which rapidly coalesce into a passivation film that seamlessly shields the $Mg_3(Sb, Bi)_2$ surface against further environmental corrosion.

isolates the material from moisture. The self-healing capability depends on the Pilling-Bedworth (PB) ratio[14]. A PB ratio slightly above unity facilitates self-healing by generating a moderate compressive stress that ensures newly formed oxides seamlessly seal surface voids, preventing further moisture infiltration. This intrinsic material design reduces the average corrosion rate of $Mg_3(Sb, Bi)_2$ by 92% (to 95 $\mu m \cdot year^{-1}$) in air and by 86% (to 0.36 $\mu m \cdot h^{-1}$) in water, effectively solving corrosion issues during material storage and processing that cannot be addressed by conventional coating strategies. Importantly, since the $Mg_{17}Al_{12}$ phase is generated *in situ* by introducing Al, the content of Mg in the matrix will also be concomitantly influenced. Thus, the content of Mg vacancies, as well as the carrier concentration and thermoelectric properties, might change with the addition of Al[15]. For practical applications, we must balance the stability and energy-conversion efficiency. In the *Nature Materials* study[13], the thermoelectric performance is even slightly enhanced by incorporating 20 at% Al. Moreover, the biased carrier concentration in the matrix due to the *in situ* formation of the second phase can be further optimized via tuning the content of other dopants, such as Te and lanthanide elements[16]. Therefore, the second-phase engineering, in conjunction with carrier concentration optimization, can enable an ideal balance between stability and functionality. This strategy is also generalizable, as demonstrated in other moisture-sensitive systems, including $Mg_2(Sn, Ge)$ and $CaMg_2Bi_2$, through incorporating Al to construct anodic secondary phases. Yet, the appropriate dopants can vary from Al, depending on the matrix material, the thermodynamically favorable second phase, and the difference in equilibrium potential between them. The *Nature Materials* article offers an excellent paradigm and principles for screening suitable element candidates.

In addition to material optimization, Sui and colleagues tackled another hidden corrosion source that

limits device-level reliability, namely accelerated material corrosion induced by the interface barrier layer. The commonly used Fe interface layer possesses a much higher equilibrium potential than the $Mg_3(Sb, Bi)_2$ matrix, accelerating corrosion of the matrix during operation[17]. To overcome this, the team replaced Fe with $Mg_{17}Al_{12}$ as the interface layer. This design maintains low contact resistivity, matched thermal expansion, and chemical stability, which are essential requirements for thermoelectric interface materials[18,19], while simultaneously eliminating interfacial galvanic corrosion and providing cathodic protection. As a result, no performance degradation of the thermoelectric device was observed after 28 days under harsh hot-humid conditions (350 K, 70% RH). The study also reminds us that the galvanic stability, which had been overlooked, should also be included in the design principles of thermoelectric interface materials in future studies.

This work shifts the design paradigm for thermoelectric materials and devices from conventional coating strategies to sacrificial anode protection. It highlights the power of an integrated, multi-scale approach combining electrochemistry, materials science, and device engineering to enhance both stability and functionality. Beyond enabling moisture-resistant and high-performance Mg-based thermoelectric devices, this study establishes a broadly applicable design strategy for corrosion-resistant functional materials.

DECLARATIONS

Authors' contributions

Wrote the manuscript: Yu, Y.
Created the figure: Zhang, Q.
Contributed through helpful discussions: Wuttig, M.

Availability of data and materials

Not applicable.

AI and AI-assisted tools statement

Not applicable.

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Conflicts of interest

Yu, Y. is an Editorial Board Member of the *Advanced Energy Conversion* journal. He had no involvement in the review or editorial process of this manuscript, including but not limited to reviewer selection, evaluation, or the final decision, while the other authors have declared that they have no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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