nature portfolio

Peer Review File

A rolling-mode triboelectric nanogenerator with multi-tunnel grating electrodes and opposite-charge-enhancement for wave energy harvesting

Editorial Note: Parts of this Peer Review File have been redacted as indicated to remove third-party material where no permission to publish could be obtained.

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The authors have reported the power generation with MO-TENG and have demonstrated the applicability of the system for random and harmonic waves. They authors have demonstrated experimental results, but no significant novelty can be found. The design and experiments are more or less regular. Hence, the manuscript is not suitable for Nat Comm due to the lack significant.

Reviewer #2:
Please see attached PDF.

Reviewer #3 (Remarks to the Author):

The authors report a triboelectric nanogenerator for wave energy harvesting, incorporating some unique and novel features to enhance power output. The authors should be commended for the detailed manuscript, with a thorough discussion of the data included. However, there are major revisions that are required, as listed below.

1. The first sentence of the Abstract refers to a specific geopolitical event. It should be removed/replaced. The authors should focus on discussing the motivation to their work in a general scientific way.

2. Lines 122-125, the figures for transferred charge and output current appear to be compared against the F-TENG only. Please revise the wording to make this clear since you are not comparing against the M-TENG.

3. A few sub-figures in Fig. 1 are probably not needed since they do not add much information. Specifically, Fig. 1b and 1i.

4. Line 141, the architecture of the TENG should be described as cuboid or hexahedral, not cubic.

5. There appear to be some mistakes in Equations (1), (2) and (3). Please check them carefully. For L_(1,F-TENG), the expression lacks the gaps between the balls as shown in Fig. 2b. For H (all three designs), the terms in the brackets should be (delta_1 + delta_2).

6. Define Q_sc in line 174, in the same way that you define the other important parameters.

7. Clarify how you apply the external excitation mentioned in lines 210-211. You could point your reader to a figure showing the set up.

8. What is the power consumption of the power management module? Can you discuss that against the overall power output that you obtain from the stacked MO-TENG?

9. Define VCG (line 368).

Reviewer #2 (Attachment):

Title: **A Rolling-mode Triboelectric Nanogenerator with Multi-tunnel Grating Electrodes and Opposite-charge-enhancement for Wave Energy Harvesting**

Number: **NCOMMS-24-05596**

This study described an approach combining rating electrodes and ternary tribo-materials to greatly improves rolling mode TENG for ocean wave energy harvesting. Additionally, the authors present a self-driven ocean sensing (SOS) system capable of fully self-powered water quality monitoring and wireless communication under random wave conditions, which provides a good contribution towards practical TENG applications. However, the manuscript still has some questions that need to be revised and answered before consideration of publication.

1. The authors should thoroughly discuss the novelty of the design and fabrication compared to existing literature.

2. It appears that TENG with PTFE exhibits significantly higher performance compared to other materials. Please clarify this point. Why did you investigate PDMS?

4. While POM yields higher TENG performance than Nylon, the reason for choosing Nylon needs clarification. Furthermore, a scientific discussion on the charge transfer mechanisms between PTFE and Nylon should be included.

5. A systematic investigation of TENG design parameters, such as grating electrode dimensions and ball size, should be conducted and reported.

6. Including simulations would enhance the content of this work and provide additional insights.

7. Considering the potential for enhancing performance, the manuscript could explore the impact of using balls with porous or patterned surface structures.

Response to Review Comments

We are thankful for the time and efforts of the editor and reviewers. The review comments are valuable for us to improve the quality of our manuscript. We have considered and addressed all the comments from the reviewers, and the detailed point-to-point response is given below. It is worth noting that bookmarks have been generated for the reviewers to quickly navigate to the responses to specific comments.

Response to Reviewer #1:

General Comment: The authors have reported the power generation with MO-TENG and have demonstrated the applicability of the system for random and harmonic waves. They authors have demonstrated experimental results, but no significant novelty can be found. The design and experiments are more or less regular. Hence, the manuscript is not suitable for Nat Comm due to the lack significant.

Response: First and foremost, we would like to express our gratitude to **Reviewer #1** for the time and effort in reviewing our manuscript. We appreciate the feedback on the perceived lack of novelty in our work. However, we believe some key aspects and contributions of our research may have been overlooked. In this response, we will provide a clear elaboration on the novelties and contributions of our work. We hope the reviewer can reconsider and revisit our work in light of the additional details we provide.

Reviewer #1 also raised a concern about the device design. It is well-known that compact and straightforward design is the inherent characteristic and the most significant advantage of TENGs, contributing to their robustness, low-frequency response, and high durability. Utilizing complex structures and transmission mechanisms will weaken the nature and benefits of TENGs and lead to a convergence with the design strategies of electromagnetic generators. Thus, our design MO-TENG inherits the simple design of rolling mode TENGs. Following this principle, we leveraged multi-tunnel grating electrodes and the opposite charge enhancement strategy to enhance the output performance of a TENG for the first time. We have conducted experiments to evaluate the performance of this device and proven its superiority. Despite the more or less regular nature of the design and experiments, the obtained results greatly surpassed those reported in the literature in this field.

The revised details in response to your comments have been highlighted in pink in the manuscript and response letter.

This study introduces an innovative approach to greatly enhance TENGs' outputs. The proposed TENG demonstrated excellent performance in wave energy harvesting, and we applied it to realize a fully TENG-powered IoT node in an actual ocean environment. The novelty and significance are summarized and highlighted from three aspects:

Novelty #1: For the first time, we have developed a rolling mode TENG by integrating and combining the design strategies of Multi-tunnel Grating Electrodes and Opposite-Charge-Enhancement.

Rolling mode TENGs represent an effective and potent option for harnessing ambient mechanical energy, particularly in the context of high-entropy blue energy in oceanic environments. Although many robust rolling-mode TENG devices have been proposed, most of them still have the problem of low output current (< 5 μA) and high internal resistance (about 1 GΩ) [*ACS Nano* **2019**, 13, 1932−1939; *Nano Energy* **2021**, 84, 105920; *ACS Energy Lett.* **2021**, 6, 2809; *Energy Environ. Sci.* **2023**, 16(7), 3040]. Some researchers employed tunnel design to increase the effective electrode area [*J. Mar. Sci. Eng.* **2022**, 10, 455.; *Adv. Energy Mater.* **2023**, 13, 2301665], resulting in a significant improvement in charge transfer. However, the issues of low current output and too large internal resistance persist without any good solutions.

Grating electrodes were specifically developed to increase the charge transfer velocity in sliding-mode TENGs, thus boosting the current output. Though some rolling-mode TENGs incorporated the design of grating electrodes in bearings, those configurations necessitate cage-like structures to maintain an appropriate gap between cylinders and balls, averting charge neutralization between electrodes, as shown in Figure R1. More details can be found in the literature [*ACS Energy Lett.* **2021**, 6, 2343; *Nano Energy* **2023**, 109, 108308]. Nonetheless, the above approach is impracticable for the present study because spatial constraints prevent cage installation. In this study, we have, for the first time, developed a rolling-mode TENG that utilizes grating electrodes **without requiring any cages**.

The ternary electrification layered architecture [*ACS Nano* **2020**, 14(7), 9050; *Nat. Comm.*, ***2021**, 12, 5470*], as shown in Figure R2, was also proposed to enhance the output, marking a significant advance in this field. However, such architectures were initially and exclusively for sliding-mode TENG. The increased wear in sliding-mode TENGs due to this mechanism may lead to a significant decline in performance and structural integrity over extended periods of operation, posing a crucial issue that cannot be ignored [*Nat. Comm.*, ***2021**, 12, 5470*].

To address the above challenges, we have, **for the first time**, integrated the multi-tunnel design and grating electrodes in a rolling mode TENG through an innovative design approach, as shown in Figure 2h. Moreover, unlike the conventional design using a cage, we encase two types of balls with identical diameters but opposite electron affinities within the device (Figure 2i). This setup effectively mitigates the charge neutralization effect and maintains a compact structure, concurrently enhancing triboelectrification and electrostatic induction effects without reliance on a fixed frame. The results (Revised Figure 1e) show that the MO-TENG demonstrates a significant improvement compared to a Flat Structure TENG (F-TENG) and a Multi-Tunnel TENG (M-TENG), with respectively, a 7-fold and 2.3-fold increase in transferred charge, as well as a 51.6-fold and 14.95-fold increase in current output. The photograph of the F-TENG and M-TENG are shown in Figure S1.

Last but not least, due to the merits of the rolling mode, the wear issue was significantly reduced.

Figure 4g showcases the scanning electron microscopic (SEM) images of the surfaces of the PTFE ball, Nylon ball, and Cu film before usage (sub-images 1, 3, and 5 in the first row) and after 10 days of testing (sub-images 2, 4, and 6 in the second row). A comparison of the images in the two rows finds minor interface abrasion, which proves the high durability of the developed MO-TENG.

[REDACTED]

Figure R1: Structures of bearing-like TENGs. (a) *ACS Energy Lett.* **2021**, 6, 2343. (b) *Nano Energy* **2023**, 109, 108308.

a

[REDACTED]

b

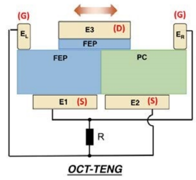


Figure R2: TENGs with ternary electrification layered architectures. (a) *ACS Nano* **2020**, 14(7), 9050. (b) *Nat. Comm.*, ***2021***, *12, 5470*.

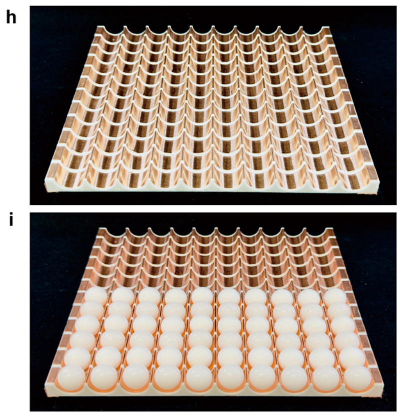
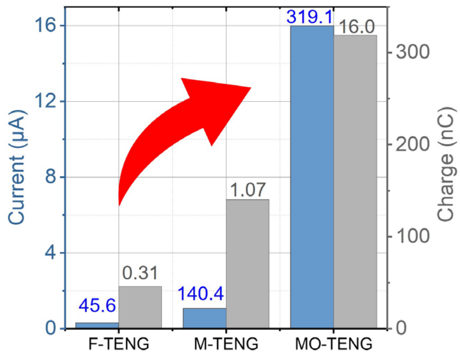


Figure 2(h-i): Photograph of the multi-tunnel grating electrodes without/with dielectric balls.



Revised Figure 1f: Comparison of the transferred charge and current generated by the F-TENG, M-TENG, and MO-TENG.

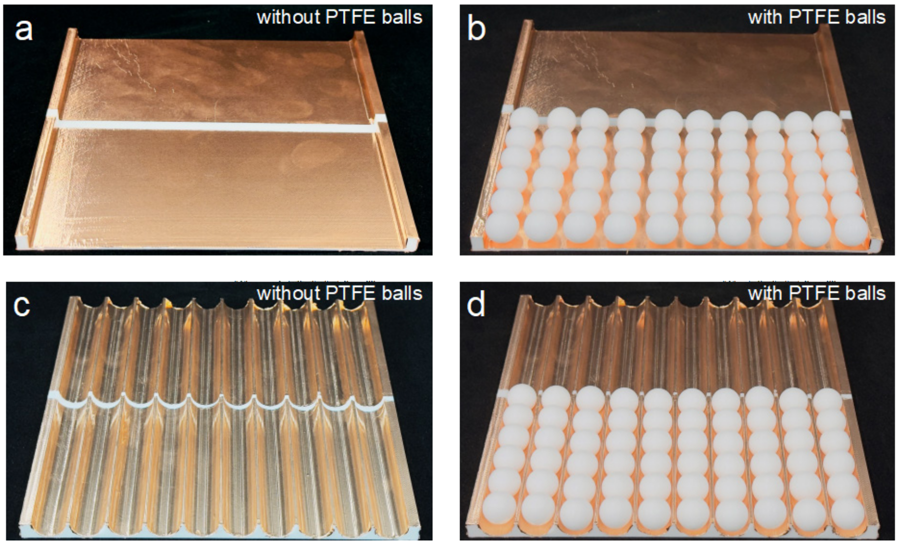


Figure S1: Photographs of (a-b) the F-TENG and (c-d) the M-TENG.

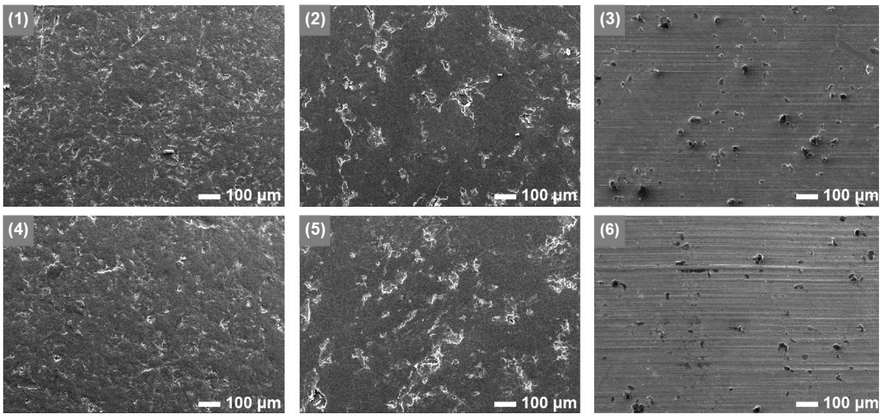


Figure 4g: SEM images of the PTFE ball, Nylon ball, and Cu of unused (sub-graphs in the first row: 1, 3, 5) and after 10 days (sub-graphs in the second row: 2, 4, 6).

Novelty #2: The proposed TENG design has achieved a low optimal resistance and an unprecedented power density.

Leveraging the multi-tunnel grating electrodes and opposite-charge-enhancement design, the MO-TENG produces significantly large output, with instantaneous and RMS power densities of 185.4 W/m^3·Hz and 10.92 W/m^3·Hz at 200 MΩ. These results represent a significant breakthrough in rolling-mode TENGs, as we have achieved an unprecedented power density at a remarkably low optimal resistance (as shown in Figure 1f). Compared with other prototypes for wave energy harvesting applications, the proposed MO-TENG also exhibits superiority in terms of the peak and RMS power densities. As shown in Figure S25. The output performance is much higher than previously reported Charge Pumping TENG [126.8 W/m^3·Hz in peak and 1.4 W/m^3·Hz in RMS, *Nat. Comm.*, **2020**, 11, 4203], and Chiral networks TENG [7.88 W/m^3·Hz in peak and 2.18 W/m^3·Hz in RMS, *Energy Environ. Sci.*, **2023**, 16, 3040], active resonance TENG [16.6 W/m^3·Hz in peak, *Joule*, **2021**, 5, 1613], etc. Other detailed comparative data can be found in Table S1.

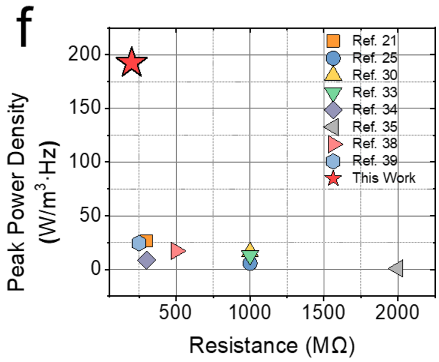


Figure 1g: Comparison of the peak power and optimal resistance of the MO-TENG with other designs[21,25,30,33–35,38,39].

[REDACTED]

Figure S1: Comparison of the peak and RMS power densities in this work with others[1–8].

Table S1. Summary of previously reported TENGs for blue energy harvesting.

	Year	Name	Working Mode	Features	Typical Output				Volume	Application	Ref
					Q_{sc}	Power (mW)	Power Density ($W/m^3 \cdot Hz$)	Match Resistance			
1	2023	MO-TENG	Rolling Mode	Multi-tunnel; Grating Electrodes; Opposite Charge Enhancement	**379.03 nC** (Motor, 0.5 Hz)	**Peak: 37.17** (Motor, 0.5 Hz, Per unit) **RMS: 2.19** (Motor, 0.5 Hz, Per unit)	**Peak: 185.4** (Motor, 0.5 Hz, Per unit) **RMS: 10.92** (Motor, 0.5 Hz, Per unit)	**200 MΩ** (Motor, 1 Hz)	**400.98 cm^3**	Navigation Light; Self-powered TDS Sensing; Self-powered LoRa Communication	This work
2	2023	SR-TENG	Rolling mode		**810 nC** (Motor, 2 Hz)	**Peak: 16** (Motor, 2 Hz) **RMS: 2.9** (Motor, 2 Hz) (ten units)	**Peak: 26.435** (Motor, 2 Hz) **RMS: 5.04** (Motor, 2 Hz) (ten units)	**300 MΩ** (Motor, 2 Hz)	**302.63 cm^3**	LEDs; Temperature and Humidity Sensor;	[9]
3	2023	FE-TENG	Rolling mode	Bio-inspired	**N/A**	**N/A**	**Peak: 5.6** (Motor, 1.25 Hz)	**1000 MΩ** (Motor, 1.25 Hz)	**N/A**	LEDs; Thermometer	[10]
4	2019	Self-assembly TENG	Rolling mode	Self-assembly	**520 nC** (Motor, 2 Hz, one unit)	**Peak: 8.75** (Motor, 1.67 Hz)	**Peak: 19.52** **RMS: 5.20** (Motor, 1.67 Hz)	**1000 MΩ** (Motor, 1.67 Hz)	**268 cm^3**	LEDs; Thermometer; Wireless transmitter;	[11]
5	2023	Chiral Network of Triboelectric Nanogenerator	Rolling mode	Chiral networks	**540 nC** (Motor, 1.5 Hz, Per Unit)	**Peak: 7.88** (motor, 1.5 Hz) **RMS: 2.18** (motor, 1.5 Hz)	**Peak: 19.61** (motor, 1.5 Hz) **RMS: 5.42** (motor, 1.5 Hz)	**1000 MΩ** (Motor, 1 Hz)	**267.95 cm^3**	LEDs; T&H Sensing (Wireless)	[7]

6	2021	Spherical TENG	Rolling mode	Polacrylate Balls	**820 nC** (Motor, 5.9 Hz, one unit)	**Peak: 10.7** (Motor, 5.9 Hz)	**Peak: 3.47** (Motor, 5.9 Hz)	**300 MΩ** (Motor, 5.9 Hz)	**527 cm^3**	LEDs; Timer	[12]
7	2019	T-TENG	Rolling mode	Tower-like	**46 nC** (Motor, 1.2 Hz) (one unit)	**Peak: 0.143** (Motor, 1.2 Hz) (one unit) **Peak: 14.734** (Motor, 1.2 Hz) (ten units)	**Peak: 1.03** (Motor, 1.2 Hz) (one unit) **Peak: 10.6** (Motor, 1.2 Hz) (ten units)	**2000 MΩ** (Motor, 1.2 Hz) (one unit) **300 MΩ** (Motor, 1.2 Hz) (ten units)	**1390 cm^3**	LEDs; Temperature Sensor	[13]
8	2021	S-TENG	Rolling mode	Sandwich-shape	**600 nC** (Motor, 2 Hz) (one unit)	**Peak: 25.22** (Motor, 2 Hz) (one unit)	**Peak: 17.325** (Motor, 2 Hz) (one unit)	**500 MΩ** (Motor, 2 Hz)	**727.85 cm^3**	LEDs;	[14]
9	2022	S-TENG	Rolling mode	Multi-tunnel	**14.715 µC** (Motor, 2 Hz, 48 layers)	**N/A**	**Peak: 24.535** (Motor, 2 Hz)	**300 MΩ** (Motor, 2 Hz)	**N/A**	LEDS; PH Sensor; Salinity Sensor	[15]
10	2021	WT-TENG	Sliding Mode	Solid-liquid Electrification	**47 nC** (Motor, 1 Hz)	**N/A**	**Peak: 13.1** (Motor, 1 Hz) **RMS: 3.8** (Motor, 1 Hz)	**Peak: 10 GΩ** **RMS: 4 GΩ** (Motor, 1 Hz)	**5.2 cm^3**	LEDs;	[1]
11	2020	CS-TENG	Contact-Separation	Charge Pumping	**3.3 µC** (Motor, 0.7 Hz)	**Peak: 126.8** (Motor, 1 Hz) **RMS: 1.4** (Motor, 1 Hz)	**Peak: 17.81** (Motor, 1 Hz) **RMS: 0.19** (Motor, 1 Hz)	**517 kΩ** (Motor, 0.7 Hz)	**488.79 cm^3**	LEDs; Thermometer; Barometer	[2]

12	2019	OB-TENG	Contact-Separation	Multi-layer	**10.5 µC** (Motor, 1 Hz)	**Peak: 38.7** (Motor, 1 Hz) **RMS: 3.45** (Motor, 1 Hz)	**Peak: 9.675** (Motor, 1 Hz) **RMS: 0.863** (Motor, 1 Hz)	**13.8 MΩ** (Motor, 1 Hz)	**4000 cm^3**	LEDs; Thermometer;	[3]
13	2023	DE-TENG	Sliding Mode	Solid-liquid Electrification	**0.55 µC** (Motor, 0.5 Hz)	**Peak: 0.85** (Motor, 0.5 Hz) **RMS: 0.14** (Motor, 0.5 Hz)	**Peak: 61.9** (Motor, 0.5 Hz) **RMS: 9.88** (Motor, 0.5 Hz)	**500 MΩ**	**27.3 cm^3**	LEDs; Thermometer; TDS sensing	[4]
14	2023	CS-TENG	Sliding Mode	Frequency Up-Conversion	**1.1 µC** (Motor, 1.75 Hz)	**Peak: 14.36** (motor, 1.75 Hz) **RMS: 3.53** (motor, 1.75 Hz)	**Peak: 19.71** (motor, 1.75 Hz) **RMS: 4.85** (motor, 1.75 Hz)	**560 kΩ** (Motor, 1.75 Hz)	**416 cm^3**	LEDs; Thermometer; Anemometer	[5]
15	2019	TD-TENG	Sliding Mode	Eccentric Pendulum	**3.3 µC** (Motor, 1 Hz)	**Peak: 29.4** (Motor, 1 Hz) **RMS: 7** (Motor, 1 Hz)	**Peak: 30.21** (Motor, 1 Hz) **RMS: 7.19** (Motor, 1 Hz)	**1 MΩ** (Motor, 1 Hz)	**973.3 cm^3**	LEDs; TDS Sensing	[6]
16	2023	WLM-TENG	Sliding Mode		**1.1 µC** (Wave, 1 Hz)	**Peak: 50** (Wave, 1 Hz) **RMS: 2.99** (Wave, 1 Hz)	**Peak: 14.1** (Wave, 1 Hz) **RMS: 0.84** (Wave, 1 Hz)	**200 kΩ** (Wave, 1 Hz)	**3546.09 cm^3**	LEDs; Thermometer; Barometer; TDS sensing	[8]

Novelty #3: We have designed a fully functional system, i.e., the SOS system, consisting of ultra-low power consumption energy management and wireless LoRA communication modules, to complete complex self-powered sensing tasks.

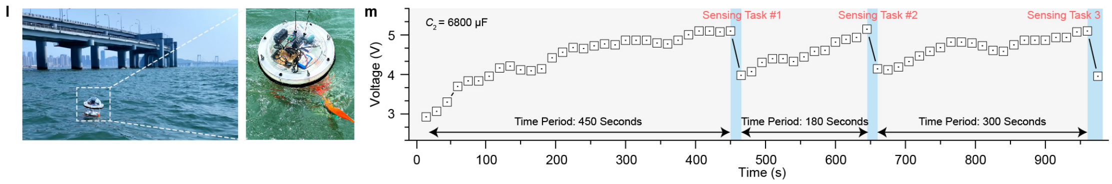
Self-powered sensing, a pivotal application scenario for TENGs, still requires further advancement toward practical implementation. Presently, there are three critical challenges in the realm of self-powered sensing applications that demand groundbreaking solutions.

i. **The issue of cold-start capability has always been overlooked in previous works.** TENG acts as an energy harvester and needs to be coupled with auxiliary circuits to realize the establishment of self-powered nodes. The circuit needs to enable energy management, sensor signal acquisition, and most importantly, cold-start capability. The absence of cold-start functionality inhibits their utility in practical scenarios [*ACS Nano* **2019**, 13, 1932−1939; *Nano Energy* **2021**, 84, 105920; *Nat. Comm.*, **2020**, 11, 4203]. Despite a great advance in output optimization in some works, the lack of a cold start during a self-powered demonstration remains unconvincing.

ii. **The singularity of sensor information.** Temperature and humidity sensors are often used in application demonstrations due to their low power consumption and accessibility. However, such easily accessible information cannot provide a comprehensive understanding of marine environments. Hence, there is a need for more advanced sensing capabilities to gain more diverse information.

iii. **Limitation of data transmission distance.** The vast range of marine environments makes Bluetooth technology, which is typically used for short-range data transmission, inadequate [*Adv. Energy Mater.* **2023**, 13, 2301665; *Nano Energy* **2020**, 76, 105052]. This puts forward the demand for a more robust energy harvesting foundation and effective power management to support communication modules that cater to higher energy demands and extended ranges.

To summarize, the issues mentioned above primarily arise from **the unsatisfactory performance of the energy harvesters** and **the lack of a well-designed power management circuit**. In this work, we employed innovative design principles to significantly enhance the output of the TENG, rendering this energy harvester more adept at fulfilling marine sensing requirements. We further developed a power management module (PMM) to optimize the capacitor charging performance and proposed a fully functional self-driven ocean sensing (SOS) system that regulates energy flow, enables cold-start, and integrates commercial sensors and a long-distance wireless communication module. For MO-TENGs to survive in the ocean as well as to serve, two variants of marine buoys equipped with MO-TENGs for wireless water quality sensing have been prototyped to operate under regular and random wave excitations. In the revision stage, we have also conducted an in-situ marine field test for water quality sensing, as shown in Figure 6k-l and Video S6. Results show that the marine buoy with stacked MO-TENGs can be deployed in ocean environments to monitor the water quality. The entire wireless sensing process was carried out autonomously, without the need for external power sources and manual oversight, thus marking a substantial stride towards the practical deployment of TENG technology in real-world applications.



Revised figure 6 (l-m): Photograph and voltage history of the self-powered sensing process in the real ocean environment.

In response to **Reviewer #1**'s feedback, we have highlighted and refined the innovation and original contributions of this work. Correspondingly, we have revised and restructured the **ABSTRACT**, **INTRODUCTION**, and **CONCLUSION** sections of our manuscript to reflect that. In addition, to strengthen the credibility of our methodology, we further studied the influences of the geometrical parameters on MO-TENG's outputs, evaluated its outputs on a 6-degree-of-freedom platform and a wave tank, addressed circuit power consumption management of the power management module (PMM), and updated the **RESULTS** section for increased clarity and impact. Last but not least, we carried out an in-situ ocean test, which represents an advanced showcase of TENG-powered sensing according to the authors' best knowledge. The results have been supplemented into the revised manuscript. This field test significantly highlighted the significance and contributions of our work in advancing marine technology applications. The details of the revisions made in the manuscript are as follows:

Revision to the manuscript:

1. In the **ABSTRACT**, lines 20 to 21, the sentence "However, the high internal resistance and low-current features of TENGs limit the power outputs, especially as the ocean environment usually induces low-frequency motion." **has been revised as** "However, the high internal resistance, low current, and friction-induced abrasion issues of TENGs limit their performance and practical applications." to **introduce the research gap in this stage**.
2. In the **ABSTRACT**, lines 21 to 31, the sentences "In this paper, we designed and prototyped a rolling mode TENG (MO-TENG) to harvest the wave energy by leveraging the multi-tunnel grating electrodes and the opposite-charge-enhancement effect, with instantaneous and RMS power densities of 185.4 W/m^3·Hz and 10.92 W/m^3·Hz at 200 MΩ. The applicability of the MO-TENG has been well explored and demonstrated via a self-powered water quality sensing application under harmonic and random waves. We envision that our design will pave the way for TENG-based energy harvesting in the marine IoT." **has been revised as** "This work presents a novel rolling mode TENG, referred to as MO-TENG, that utilizes multi-tunnel grating electrodes and the opposite-charge-enhancement effect to harvest wave energy efficiently. The MO-TENG showed impressive instantaneous and RMS power densities of 185.4 and 10.92 W/m^3·Hz, respectively. By utilizing stacked MO-TENGs and an exclusively designed power management module, a self-powered ocean sensing (SOS) system with sensing, computing, and long-range wireless communication (0.8 km) capabilities was developed. Laboratory and in-situ ocean tests were conducted to assess and validate the SOS

system. This work signifies a substantial breakthrough in wave energy harvesting using TENGs, offering solutions for the previously challenging deployment of marine self-powered sensing nodes." to **point out the contribution and significance of this work**.

3. In the **INTRODUCTION**, lines 37 to 40, the sentences "Global population growth and resource scarcity are driving demand for ocean exploration. Unfortunately, marine pollution and climate change seriously threaten marine resources[2]." **has been revised as** "The urgency for ocean exploration is escalating because of global population growth and the pressing need for resources. However, significant threats such as marine pollution and climate change present significant challenges to the marine environment and resources[2]."
4. In the **INTRODUCTION**, lines 40 to 45, the sentences "Therefore, real-time and long-term monitoring technology is necessary to maintain ocean sustainability[3,4]. However, due to the harsh environment and vast expanse of the ocean, using battery and/or cord power supplies always limits the life span and deployment range of monitoring equipment. Harnessing in situ energy from the ocean environment to power ocean observation nodes is a promising method to address such issues[5–7]." **has been revised as** "Therefore, deploying real-time and long-term monitoring technologies is imperative for ensuring ocean sustainability[3,4]. Nevertheless, the harsh and expansive nature of the oceanic environment often restricts the lifespan and deployment scope of monitoring equipment with heavy reliance on batteries and/or cord power supplies. Harnessing in situ energy from the marine environment to energize ocean observation nodes presents a viable solution to overcome these limitations[5–7].".
5. In the **INTRODUCTION**, lines 53 to 55, the sentence "Using TENGs for blue energy harvesting has gained significant attention[15–18]. In the past decade, numerous prototypes have been designed, and they can be classified into two main types: open and enclosed configurations." **has been revised as** "The utilization of TENGs for harvesting blue energy has attracted considerable interest[15–18]. Over the last decade, a variety of prototypes have been developed and can be broadly classified into two primary categories: open and enclosed configurations.".
6. In the **INTRODUCTION**, lines 57 to 59, the sentences "These harvesters have simple structures and are lightweight. However, the lifespans of the contact material and electrodes of open-structure TENGs are short due to the corrosive nature of seawater." **have been revised as** "These harvesters have simple structures and are lightweight. Nonetheless, the durability of the contact materials in open-type TENGs is significantly reduced due to seawater's corrosive effect.".
7. In the **INTRODUCTION**, lines 60 to 65, the sentences "By incorporating a protective shell, TENGs can be shielded from the harsh ocean environment, resulting in improved output performance and prolonged operational lifespan[23–26]. Notably, using rolling balls as inertial bodies is currently esteemed as an ideal scheme in this field[27,28]. Due to the low friction and ease of activation in the rolling mode, these TENGs exhibit high durability, seamless integration, and good output performance[29,30]." **have been revised as** "Equipping TENGs with a protective casing can shield them from harsh ocean conditions, thereby boosting their outputs and extending their service lifespans[23–26]. Employing rolling balls as inertial bodies is esteemed as an ideal scheme in this field[27,28]. The low friction and effortless activation characteristic of the rolling balls contribute to significantly enhancing durability, seamless integration, and

superior output performance of the rolling-mode TENGs[29,30].".

8. In the **INTRODUCTION**, lines 72 to 74, the sentence "In recent years, researchers have further improved the SS-TENG through materials selection[21] and electrode treatment[25,32–34], promoting it toward practical applications in the ocean." **has been revised as** "Recent advancements in the field have enhanced the SS-TENG through materials selection[21], electrode design[25,32–34], and building harvester networks[35], promoting it toward practical applications in the ocean.".
9. In the **INTRODUCTION**, the sentence "Moreover, based on flow-induced vibration (FIV) phenomena, structures have also been developed to harvest flow energy[35,36]." **has been deleted**.
10. In the **INTRODUCTION**, lines 74 to 80, the sentences "Previous studies have proven that TENGs designed in rolling mode can be easily excited by waves to generate high-voltage outputs. However, under low-frequency wave excitations in the ocean, most TENGs produce outputs of only a few microamps (< 5 μA) but have optimal resistances of giga-ohms (GΩ), which makes it hard and impractical to directly power electronics in the marine environment." **have been revised as** "While it has been proven that TENGs, particularly those in rolling mode, can be effectively driven by wave to generate substantial voltage output, the prevalent low-frequency wave dynamics in marine environments typically lead them to produce very limited current outputs (<5 μA) at optimal resistances reaching giga-Ohms (GΩ). This limitation poses significant challenges for directly employing such TENGs to power electronic devices in the marine environment.".
11. In the **INTRODUCTION**, lines 80 to 86, the sentences "Some researchers attempted to address these issues by designing appropriate mechanical mechanisms to upconvert the motion frequency, known as the frequency up-conversion (FUC) strategy. Whereas, the FUC strategy often weakens the characteristics of TENGs in the low-frequency region and leads to the sweet zone of electromagnetic generators. Regulating the outputs of TENGs by power management circuits is another means to mitigate the low-current, high-internal resistance issue. But unfortunately, the power outputs of TENGs eventually degrade after circuit regulation since electronics in power management circuits consume power." **have been revised as** "Grating electrodes are employed to boost current output and decrease internal resistance by accelerating charge transfer[36], typically in conjunction with sliding mode TENGs[3] and frequency up-conversion strategies (FUC)[24]. However, the inherent sliding friction in sliding mode TENGs significantly deteriorates their output performance and durability[37]. On the other hand, although the FUC strategy can optimize the performance of piezoelectric and electromagnetic generators, it diminishes the efficacy of TENGs in low-frequency applications.".
12. In the **INTRODUCTION**, lines 87 to 91, the sentences "In this work, a rolling mode TENG with multi-tunnel grating electrodes and opposite-charge-enhancement (MO-TENG), as shown in Figure 1a, is proposed for the first time to address the challenges mentioned above in ocean wave energy harvesting. The rolling mode enables low friction between the interfaces for an extended lifespan and makes the TENG effortlessly excited by the wave. The multi-tunnel structure improves the effective electrode area of the TENG and thus enhances the electrostatic induction effect. Instead of the FUC strategy, this study combines the grating electrodes and ternary tribo-materials to boost the charge transfer rate, triboelectrification, and

electrostatic induction processes. The integration of these strategies and approaches led to a great optimization of the rolling-mode TENG in the blue energy harvesting area, producing an instantaneous power density of 192.34 W/m^3·Hz and a root-mean-square (RMS) power density of 11.33 W/m^3·Hz. **have been revised as** "In this work, a novel rolling mode TENG named MO-TENG is designed and proposed, that utilizes multi-tunnel grating electrodes and the opposite-charge-enhancement effect to harvest wave energy efficiently, as illustrated in Figure 1a and Figure 1b-d. These strategies improve TENGs' performance leading to an instantaneous power density of 185.4 W/m^3·Hz and an RMS power density of 10.92 W/m^3·Hz.".

13. In the **INTRODUCTION**, lines 94 to 96, the sentence "Furthermore, we developed a self-driven ocean sensing (SOS) system that regulate energy, and integrates commercial sensors and a wireless communication module." **has been revised as** "Furthermore, a self-driven ocean sensing (SOS) system is developed using stacked MO-TENGs and an ultra-low power consumption circuit with the cold-start function to realize wireless self-powered sensing.".
14. In the **INTRODUCTION**, lines 96 to 102, the sentences "When exposed to random wave excitations, the SOS system can successfully realize fully self-powered water quality monitoring and wireless communication. This marks the first instance of TENG accomplishing a self-powered task in real wave conditions." **have been revised as** "The impressive performance of the MO-TENG and the well-thought-out design of the energy management circuit endow the SOS system with excellent self-powered sensing and communication capabilities in the marine environment. Remarkably, a buoy fitted with 30 stacked MO-TENG units was tested in a wave tank and the real ocean. This smart buoy successfully realized fully self-powered water quality monitoring and wireless communication tasks and demonstrated significant potential for practical deployment.".
15. In the **INTRODUCTION**, lines 102 to 106, the sentences "Overall, this novel design effectively enhances the TENG's performance and provides a reliable solution for application in the marine Internet of Things (Marine IoT). Our work represents a substantial step forward, bringing TENGs closer to practical application." **have been revised as** "Overall, the design methodology presented in this work notably improves the TENG's performance and offers a reliable solution for use within the Marine Internet of Things (IoT) domain. This study marks a significant advancement, promoting the research of TENGs towards practical applications.".
16. In the **Conclusion**, lines 426 to 435, the sentences "Using stacked MO-TENGs, a self-powered ocean sensing (SOS) system is designed to monitor water quality remotely and wirelessly. A buoy installed with 10 stacked MO-TENGs was submerged in the water tank that has the wave generation ability. Under the simulated wave excitation, the stacked MO-TENGs in the buoy, together with those auxiliary circuit modules, accomplished the tasks of powering navigation light, water quality sensing, data collecting, and LoRa communication. Significantly, the self-powered sensing task was well demonstrated in a random wave tank, with a buoy stacked with 30 MO-TENG units. The in situ TDS values were collected and transmitted to the receiver thrice in 270 seconds. And the transmission distance can be up to 1.13 kilometers." **have been revised as** "Furthermore, a self-powered ocean sensing (SOS) system featuring cold start capability has been developed for remote and wireless water quality monitoring. At the same time, it enables the scalable construction of system-level self-powered nodes of MO-TENG-based devices for maritime applications, and has been presented and validated under various

laboratory conditions. It is worth noting that a buoy equipped with stacked MO-TENGs and the SOS system was tested in a real ocean environment. The intelligent buoy accomplished the self-powered sensing task three times within 920 seconds with a maximum transmission distance of 0.8 km, which marks the pioneering instance of a TENG-based self-powered device serving in a real ocean environment.”.

Response to Reviewer #2:

General Comment: This study described an approach combining rating electrodes and ternary tribo-materials to greatly improves rolling mode TENG for ocean wave energy harvesting. Additionally, the authors present a self-driven ocean sensing (SOS) system capable of fully self-powered water quality monitoring and wireless communication under random wave conditions, which provides a good contribution towards practical TENG applications. However, the manuscript still has some questions that need to be revised and answered before consideration of publication.

Response: We sincerely thank **Reviewer #2** for carefully reviewing our manuscript and raising professional comments to help further deepen the study and improve the quality of the manuscript. In particular, we highly appreciate the reviewer's positive comment, i.e., "which provides a good contribution towards practical TENG applications". Detailed revisions following your comments have been highlighted in green in the manuscript and response letter.

Comment 1: The authors should thoroughly discuss the novelty of the design and fabrication compared to existing literature.

Response: The authors thank **Reviewer #2** for the comments regarding the novelty of this work. After thoroughly referring to the previous works in this field, we have carefully reorganized the highlights to enhance the persuasion of this paper.

This work presents an innovative methodology to improve the output of TENG to facilitate a great performance in wave energy harvesting and realize fully self-powered IoT nodes. The novelty and significance can be highlighted in three aspects:

Novelty #1: For the first time, we have developed a rolling mode TENG by integrating and combining the design strategies of Multi-tunnel Grating Electrodes and Opposite-Charge-Enhancement.

Rolling mode TENGs represent an effective and potent option for harnessing ambient mechanical energy, particularly in the context of high-entropy blue energy in oceanic environments. Although many robust rolling-mode TENG devices have been proposed, most of them still have the problem of low output current (< 5 μA) and high internal resistance (about 1 GΩ) [*ACS Nano* **2019**, 13, 1932−1939; *Nano Energy* **2021**, 84, 105920; *ACS Energy Lett.* **2021**, 6, 2809; *Energy Environ. Sci.* **2023**, 16(7), 3040]. Some researchers employed tunnel design to increase the effective electrode area [*J. Mar. Sci. Eng.* **2022**, 10, 455.; *Adv. Energy Mater.* **2023**, 13, 2301665], resulting in a significant improvement in charge transfer. However, the issues of low current output and too large internal resistance persist without any good solutions.

Grating electrodes were specifically developed to increase the charge transfer velocity in sliding-mode TENGs, thus boosting the current output. Though some rolling-mode TENGs incorporated the design of grating electrodes in bearings, those configurations necessitate cage-like structures to maintain an appropriate gap between cylinders and balls, averting charge neutralization between electrodes, as shown in Figure R1. More details can be found in the literature [*ACS Energy Lett.* **2021**, 6, 2343; *Nano Energy* **2023**, 109, 108308]. Nonetheless, the above approach is impracticable for the present study because spatial constraints prevent cage installation. In this study, we have, for the first time, developed a rolling-mode TENG that utilizes grating electrodes **without requiring any cages**.

The ternary electrification layered architecture [*ACS Nano* **2020**, 14(7), 9050; *Nat. Comm.*, ***2021**, 12, 5470*], as shown in Figure R2, was also proposed to enhance the output, marking a significant advance in this field. However, such architectures were initially and exclusively for sliding-mode TENG. The increased wear in sliding-mode TENGs due to this mechanism may lead to a significant decline in performance and structural integrity over extended periods of operation, posing a crucial issue that cannot be ignored [*Nat. Comm.*, ***2021**, 12, 5470*].

To address the above challenges, we have, **for the first time**, integrated the multi-tunnel design and grating electrodes in a rolling mode TENG through an innovative design approach, as shown in Figure 2h. Moreover, unlike the conventional design using a cage, we encase two types of balls with identical diameters but opposite electron affinities within the device (Figure 2i). This setup effectively mitigates the charge neutralization effect and maintains a compact structure, concurrently enhancing triboelectrification and electrostatic induction effects without reliance on a fixed frame. The results (Revised Figure 1e) show that the MO-TENG demonstrates a significant improvement compared to a Flat Structure TENG (F-TENG) and a Multi-Tunnel TENG (M-TENG), with respectively, a 7-fold and 2.3-fold increase in transferred charge, as well as a 51.6-fold and 14.95-fold increase in current output. The photograph of the F-TENG and M-TENG are shown in Figure S1.

Last but not least, due to the merits of the rolling mode, the wear issue was significantly reduced. Figure 4g showcases the scanning electron microscopic (SEM) images of the surfaces of the PTFE ball, Nylon ball, and Cu film before usage (sub-images 1, 3, and 5 in the first row) and after 10 days of testing (sub-images 2, 4, and 6 in the second row). A comparison of the images in the two rows finds minor interface abrasion, which proves the high durability of the developed MO-TENG.

[REDACTED]

Figure R1: Structures of bearing-like TENGs. (a) *ACS Energy Lett.* **2021**, 6, 2343. (b) *Nano Energy* **2023**, 109, 108308.

a

[REDACTED]

b

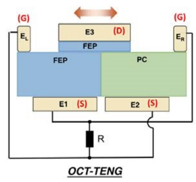


Figure R2: TENGs with ternary electrification layered architectures. (a) *ACS Nano* **2020**, 14(7), 9050. (b) *Nat. Comm.*, ***2021***, *12, 5470*.

h

i

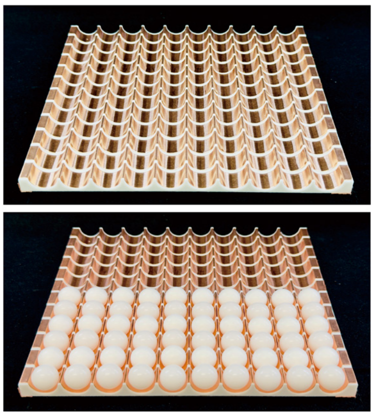
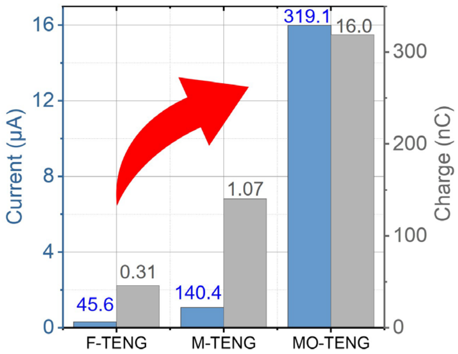


Figure 2(h-i): Photograph of the multi-tunnel grating electrodes without/with dielectric balls.



Revised Figure 1f: Comparison of the transferred charge and current generated by the F-TENG, M-TENG, and MO-TENG.

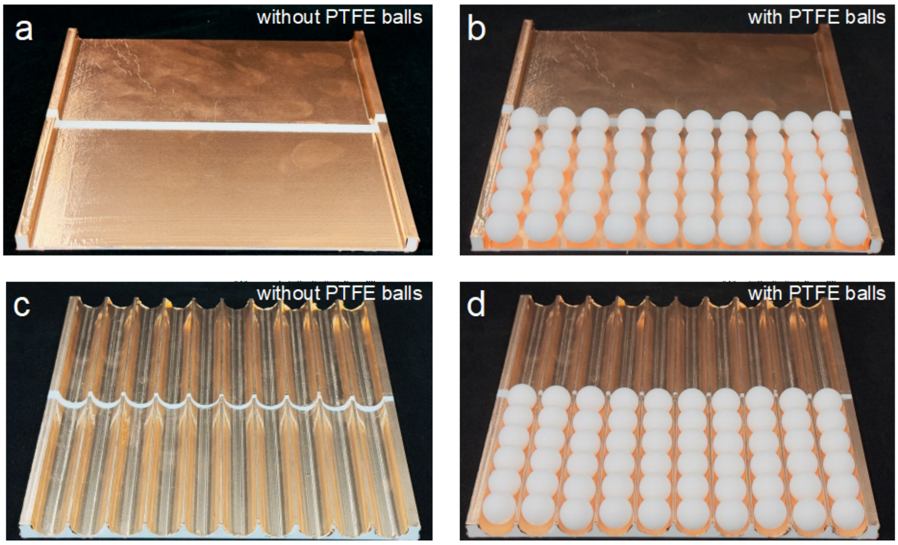


Figure S1: Photographs of (a-b) the F-TENG and (c-d) the M-TENG.

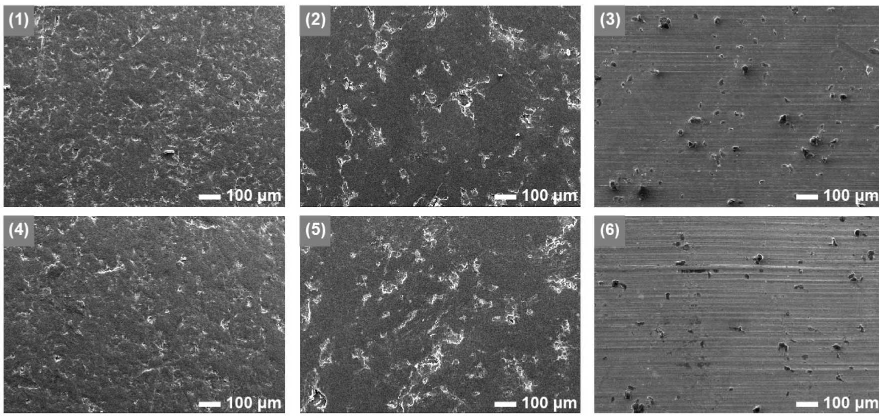


Figure 4g: SEM images of the PTFE ball, Nylon ball, and Cu of unused (sub-graphs in the first row: 1, 3, 5) and after 10 days (sub-graphs in the second row: 2, 4, 6).

Novelty #2: The proposed TENG design has achieved a low optimal resistance and an unprecedented power density.

Leveraging the multi-tunnel grating electrodes and opposite-charge-enhancement design, the MO-

TENG produces significantly large output, with instantaneous and RMS power densities of 185.4 $W/m^3 \cdot Hz$ and 10.92 $W/m^3 \cdot Hz$ at 200 MΩ. These results represent a significant breakthrough in rolling-mode TENGs, as we have achieved an unprecedented power density at a remarkably low optimal resistance (as shown in Figure 1f). Compared with other prototypes for wave energy harvesting applications, the proposed MO-TENG also exhibits superiority in terms of the peak and RMS power densities. As shown in Figure S25. The output performance is much higher than previously reported Charge Pumping TENG [126.8 $W/m^3 \cdot Hz$ in peak and 1.4 $W/m^3 \cdot Hz$ in RMS, *Nat. Comm.*, **2020**, 11, 4203], and Chiral networks TENG [7.88 $W/m^3 \cdot Hz$ in peak and 2.18 $W/m^3 \cdot Hz$ in RMS, *Energy Environ. Sci.*, **2023**, 16, 3040], active resonance TENG [16.6 $W/m^3 \cdot Hz$ in peak, *Joule*, **2021**, 5, 1613], etc. Other detailed comparative data can be found in Table S1.

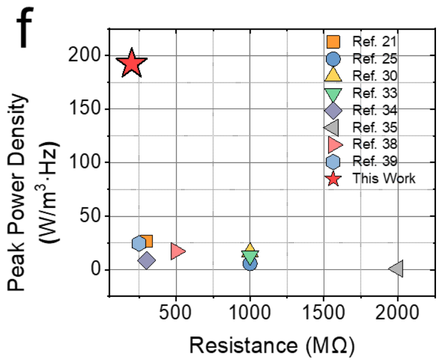


Figure 1g: Comparison of the peak power and optimal resistance of the MO-TENG with other designs[21,25,30,33–35,38,39].

[REDACTED]

Figure S2: Comparison of the peak and RMS power densities in this work with others[1–8].

Table S1. Summary of previously reported TENGs for blue energy harvesting.

	Year	Name	Working Mode	Features	Typical Output				Volume	Application	Ref
					Q_{sc}	Power (mW)	Power Density ($W/m^3 \cdot Hz$)	Match Resistance			
1	2023	MO-TENG	Rolling Mode	Multi-tunnel; Grating Electrodes; Opposite Charge Enhancement	**379.03 nC** (Motor, 0.5 Hz)	**Peak: 37.17** (Motor, 0.5 Hz, Per unit) **RMS: 2.19** (Motor, 0.5 Hz, Per unit)	**Peak: 185.4** (Motor, 0.5 Hz, Per unit) **RMS: 10.92** (Motor, 0.5 Hz, Per unit)	**200 MΩ** (Motor, 1 Hz)	**400.98 cm^3**	Navigation Light; Self-powered TDS Sensing; Self-powered LoRa Communication	This work
2	2023	SR-TENG	Rolling mode		**810 nC** (Motor, 2 Hz)	**Peak: 16** (Motor, 2 Hz) **RMS: 2.9** (Motor, 2 Hz) (ten units)	**Peak: 26.435** (Motor, 2 Hz) **RMS: 5.04** (Motor, 2 Hz) (ten units)	**300 MΩ** (Motor, 2 Hz)	**302.63 cm^3**	LEDs; Temperature and Humidity Sensor;	[9]
3	2023	FE-TENG	Rolling mode	Bio-inspired	**N/A**	**N/A**	**Peak: 5.6** (Motor, 1.25 Hz)	**1000 MΩ** (Motor, 1.25 Hz)	**N/A**	LEDs; Thermometer	[10]
4	2019	Self-assembly TENG	Rolling mode	Self-assembly	**520 nC** (Motor, 2 Hz, one unit)	**Peak: 8.75** (Motor, 1.67 Hz)	**Peak: 19.52** **RMS: 5.20** (Motor, 1.67 Hz)	**1000 MΩ** (Motor, 1.67 Hz)	**268 cm^3**	LEDs; Thermometer; Wireless transmitter;	[11]
5	2023	Chiral Network of Triboelectric Nanogenerator	Rolling mode	Chiral networks	**540 nC** (Motor, 1.5 Hz, Per Unit)	**Peak: 7.88** (motor, 1.5 Hz) **RMS: 2.18** (motor, 1.5 Hz)	**Peak: 19.61** (motor, 1.5 Hz) **RMS: 5.42** (motor, 1.5 Hz)	**1000 MΩ** (Motor, 1 Hz)	**267.95 cm^3**	LEDs; T&H Sensing (Wireless)	[7]

6	2021	Spherical TENG	Rolling mode	Polacrylate Balls	**820 nC** (Motor, 5.9 Hz, one unit)	**Peak: 10.7** (Motor, 5.9 Hz)	**Peak: 3.47** (Motor, 5.9 Hz)	**300 MΩ** (Motor, 5.9 Hz)	**527 cm^3**	LEDs; Timer	[12]
7	2019	T-TENG	Rolling mode	Tower-like	**46 nC** (Motor, 1.2 Hz) (one unit)	**Peak: 0.143** (Motor, 1.2 Hz) (one unit) **Peak: 14.734** (Motor, 1.2 Hz) (ten units)	**Peak: 1.03** (Motor, 1.2 Hz) (one unit) **Peak: 10.6** (Motor, 1.2 Hz) (ten units)	**2000 MΩ** (Motor, 1.2 Hz) (one unit) **300 MΩ** (Motor, 1.2 Hz) (ten units)	**1390 cm^3**	LEDs; Temperature Sensor	[13]
8	2021	S-TENG	Rolling mode	Sandwich-shape	**600 nC** (Motor, 2 Hz) (one unit)	**Peak: 25.22** (Motor, 2 Hz) (one unit)	**Peak: 17.325** (Motor, 2 Hz) (one unit)	**500 MΩ** (Motor, 2 Hz)	**727.85 cm^3**	LEDs;	[14]
9	2022	S-TENG	Rolling mode	Multi-tunnel	**14.715 μC** (Motor, 2 Hz, 48 layers)	**N/A**	**Peak: 24.535** (Motor, 2 Hz)	**300 MΩ** (Motor, 2 Hz)	**N/A**	LEDS; PH Sensor; Salinity Sensor	[15]
10	2021	WT-TENG	Sliding Mode	Solid-liquid Electrification	**47 nC** (Motor, 1 Hz)	**N/A**	**Peak: 13.1** (Motor, 1 Hz) **RMS: 3.8** (Motor, 1 Hz)	**Peak: 10 GΩ** **RMS: 4 GΩ** (Motor, 1 Hz)	**5.2 cm^3**	LEDs;	[1]
11	2020	CS-TENG	Contact-Separation	Charge Pumping	**3.3 μC** (Motor, 0.7 Hz)	**Peak: 126.8** (Motor, 1 Hz) **RMS: 1.4** (Motor, 1 Hz)	**Peak: 17.81** (Motor, 1 Hz) **RMS: 0.19** (Motor, 1 Hz)	**517 kΩ** (Motor, 0.7 Hz)	**488.79 cm^3**	LEDs; Thermometer; Barometer	[2]

12	2019	OB-TENG	Contact-Separation	Multi-layer	**10.5 µC** (Motor, 1 Hz)	**Peak: 38.7** (Motor, 1 Hz) **RMS: 3.45** (Motor, 1 Hz)	**Peak: 9.675** (Motor, 1 Hz) **RMS: 0.863** (Motor, 1 Hz)	**13.8 MΩ** (Motor, 1 Hz)	**4000 cm^3**	LEDs; Thermometer;	[3]
13	2023	DE-TENG	Sliding Mode	Solid-liquid Electrification	**0.55 µC** (Motor, 0.5 Hz)	**Peak: 0.85** (Motor, 0.5 Hz) **RMS: 0.14** (Motor, 0.5 Hz)	**Peak: 61.9** (Motor, 0.5 Hz) **RMS: 9.88** (Motor, 0.5 Hz)	**500 MΩ**	**27.3 cm^3**	LEDs; Thermometer; TDS sensing	[4]
14	2023	CS-TENG	Sliding Mode	Frequency Up-Conversion	**1.1 µC** (Motor, 1.75 Hz)	**Peak: 14.36** (motor, 1.75 Hz) **RMS: 3.53** (motor, 1.75 Hz)	**Peak: 19.71** (motor, 1.75 Hz) **RMS: 4.85** (motor, 1.75 Hz)	**560 kΩ** (Motor, 1.75 Hz)	**416 cm^3**	LEDs; Thermometer; Anemometer	[5]
15	2019	TD-TENG	Sliding Mode	Eccentric Pendulum	**3.3 µC** (Motor, 1 Hz)	**Peak: 29.4** (Motor, 1 Hz) **RMS: 7** (Motor, 1 Hz)	**Peak: 30.21** (Motor, 1 Hz) **RMS: 7.19** (Motor, 1 Hz)	**1 MΩ** (Motor, 1 Hz)	**973.3 cm^3**	LEDs; TDS Sensing	[6]
16	2023	WLM-TENG	Sliding Mode		**1.1 µC** (Wave, 1 Hz)	**Peak: 50** (Wave, 1 Hz) **RMS: 2.99** (Wave, 1 Hz)	**Peak: 14.1** (Wave, 1 Hz) **RMS: 0.84** (Wave, 1 Hz)	**200 kΩ** (Wave, 1 Hz)	**3546.09 cm^3**	LEDs; Thermometer; Barometer; TDS sensing	[8]

Novelty #3: We have designed a fully functional system, i.e., the SOS system, consisting of ultra-low power consumption energy management and wireless LoRA communication modules, to complete complex self-powered sensing tasks.

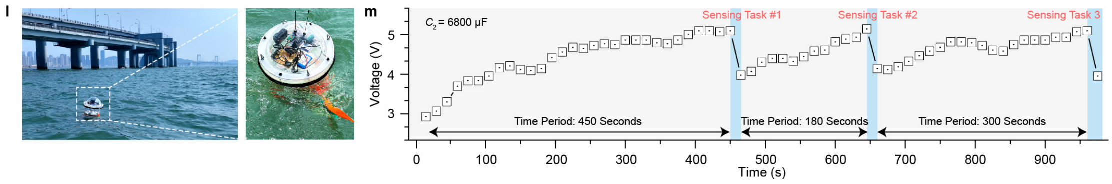
Self-powered sensing, a pivotal application scenario for TENGs, still requires further advancement toward practical implementation. Presently, there are three critical challenges in the realm of self-powered sensing applications that demand groundbreaking solutions.

iv. **The issue of cold-start capability has always been overlooked in previous works.** TENG acts as an energy harvester and needs to be coupled with auxiliary circuits to realize the establishment of self-powered nodes. The circuit needs to enable energy management, sensor signal acquisition, and most importantly, cold-start capability. The absence of cold-start functionality inhibits their utility in practical scenarios [*ACS Nano* **2019**, 13, 1932−1939; *Nano Energy* **2021**, 84, 105920; *Nat. Comm.*, **2020**, 11, 4203]. Despite a great advance in output optimization in some works, the lack of a cold start during a self-powered demonstration remains unconvincing.

v. **The singularity of sensor information.** Temperature and humidity sensors are often used in application demonstrations due to their low power consumption and accessibility. However, such easily accessible information cannot provide a comprehensive understanding of marine environments. Hence, there is a need for more advanced sensing capabilities to gain more diverse information.

vi. **Limitation of data transmission distance.** The vast range of marine environments makes Bluetooth technology, which is typically used for short-range data transmission, inadequate [*Adv. Energy Mater.* **2023**, 13, 2301665; *Nano Energy* **2020**, 76, 105052]. This puts forward the demand for a more robust energy harvesting foundation and effective power management to support communication modules that cater to higher energy demands and extended ranges.

To summarize, the issues mentioned above primarily arise from **the unsatisfactory performance of the energy harvesters** and **the lack of a well-designed power management circuit**. In this work, we employed innovative design principles to significantly enhance the output of the TENG, rendering this energy harvester more adept at fulfilling marine sensing requirements. We further developed a power management module (PMM) to optimize the capacitor charging performance and proposed a fully functional self-driven ocean sensing (SOS) system that regulates energy flow, enables cold-start, and integrates commercial sensors and a long-distance wireless communication module. For MO-TENGs to survive in the ocean as well as to serve, two variants of marine buoys equipped with MO-TENGs for wireless water quality sensing have been prototyped to operate under regular and random wave excitations. In the revision stage, we have also conducted an in-situ marine field test for water quality sensing, as shown in Figure 6k-l and Video S6. Results show that the marine buoy with stacked MO-TENGs can be deployed in ocean environments to monitor the water quality. The entire wireless sensing process was carried out autonomously, without the need for external power sources and manual oversight, thus marking a substantial stride towards the practical deployment of TENG technology in real-world applications.



Revised figure 6 (l-m): Photograph and voltage history of the self-powered sensing process in the real ocean environment.

In response to your comments, we have highlighted and refined the innovation and original contributions of this work. Correspondingly, we have revised and restructured the **ABSTRACT**, **INTRODUCTION**, and **CONCLUSION** sections of our manuscript to reflect that. In addition, to strengthen the credibility of our methodology, we further studied the influences of the geometrical parameters on MO-TENG's outputs, evaluated its outputs on a 6-degree-of-freedom platform and a wave tank, addressed circuit power consumption management of the power management module (PMM), and updated the **RESULTS** section for increased clarity and impact. Last but not least, we carried out an in-situ ocean test, which represents an advanced showcase of TENG-powered sensing according to the authors' best knowledge. The results have been supplemented into the revised manuscript. This field test significantly highlighted the significance and contributions of our work in advancing marine technology applications. Since **Reviewer #1** also commented about the novelty and significance, the highlights in the below "Revision to the manuscript" are marked in pink to minimize any misunderstanding.

Revision to the manuscript:

1. In the **ABSTRACT**, lines 20 to 21, the sentence "However, the high internal resistance and low-current features of TENGs limit the power outputs, especially as the ocean environment usually induces low-frequency motion." **has been revised as** "However, the high internal resistance, low current, and friction-induced abrasion issues of TENGs limit their performance and practical applications. to **introduce the research gap in this stage**.
2. In the **ABSTRACT**, lines 21 to 31, the sentences "In this paper, we designed and prototyped a rolling mode TENG (MO-TENG) to harvest the wave energy by leveraging the multi-tunnel grating electrodes and the opposite-charge-enhancement effect, with instantaneous and RMS power densities of 185.4 W/m^3·Hz and 10.92 W/m^3·Hz at 200 MΩ. The applicability of the MO-TENG has been well explored and demonstrated via a self-powered water quality sensing application under harmonic and random waves. We envision that our design will pave the way for TENG-based energy harvesting in the marine IoT." **has been revised as** "This work presents a novel rolling mode TENG, referred to as MO-TENG, that utilizes multi-tunnel grating electrodes and the opposite-charge-enhancement effect to harvest wave energy efficiently. The MO-TENG showed impressive instantaneous and RMS power densities of 185.4 and 10.92 W/m^3·Hz, respectively. By utilizing stacked MO-TENGs and an exclusively designed power management module, a self-powered ocean sensing (SOS) system with sensing, computing, and long-range wireless communication (0.8 km) capabilities was

developed. Laboratory and in-situ ocean tests were conducted to assess and validate the SOS system. This work signifies a substantial breakthrough in wave energy harvesting using TENGs, offering solutions for the previously challenging deployment of marine self-powered sensing nodes." to **point out the contribution and significance of this work**.

3. In the **INTRODUCTION**, lines 37 to 40, the sentences "Global population growth and resource scarcity are driving demand for ocean exploration. Unfortunately, marine pollution and climate change seriously threaten marine resources[2]." **has been revised as** "The urgency for ocean exploration is escalating because of global population growth and the pressing need for resources. However, significant threats such as marine pollution and climate change present significant challenges to the marine environment and resources[2]."
4. In the **INTRODUCTION**, lines 40 to 45, the sentences "Therefore, real-time and long-term monitoring technology is necessary to maintain ocean sustainability[3,4]. However, due to the harsh environment and vast expanse of the ocean, using battery and/or cord power supplies always limits the life span and deployment range of monitoring equipment. Harnessing in situ energy from the ocean environment to power ocean observation nodes is a promising method to address such issues[5–7]." **has been revised as** "Therefore, deploying real-time and long-term monitoring technologies is imperative for ensuring ocean sustainability[3,4]. Nevertheless, the harsh and expansive nature of the oceanic environment often restricts the lifespan and deployment scope of monitoring equipment with heavy reliance on batteries and/or cord power supplies. Harnessing in situ energy from the marine environment to energize ocean observation nodes presents a viable solution to overcome these limitations[5–7].".
5. In the **INTRODUCTION**, lines 53 to 55, the sentence "Using TENGs for blue energy harvesting has gained significant attention[15–18]. In the past decade, numerous prototypes have been designed, and they can be classified into two main types: open and enclosed configurations." **has been revised as** "The utilization of TENGs for harvesting blue energy has attracted considerable interest[15–18]. Over the last decade, a variety of prototypes have been developed and can be broadly classified into two primary categories: open and enclosed configurations.".
6. In the **INTRODUCTION**, lines 57 to 59, the sentences "These harvesters have simple structures and are lightweight. However, the lifespans of the contact material and electrodes of open-structure TENGs are short due to the corrosive nature of seawater." **have been revised as** "These harvesters have simple structures and are lightweight. Nonetheless, the durability of the contact materials in open-type TENGs is significantly reduced due to seawater's corrosive effect.".
7. In the **INTRODUCTION**, lines 60 to 65, the sentences "By incorporating a protective shell, TENGs can be shielded from the harsh ocean environment, resulting in improved output performance and prolonged operational lifespan[23–26]. Notably, using rolling balls as inertial bodies is currently esteemed as an ideal scheme in this field[27,28]. Due to the low friction and ease of activation in the rolling mode, these TENGs exhibit high durability, seamless integration, and good output performance[29,30]." **have been revised as** "Equipping TENGs with a protective casing can shield them from harsh ocean conditions, thereby boosting their outputs and extending their service lifespans[23–26]. Employing rolling balls as inertial bodies is esteemed as an ideal scheme in this field[27,28]. The low friction and effortless activation characteristic of

the rolling balls contribute to significantly enhancing durability, seamless integration, and superior output performance of the rolling-mode TENGs[29,30].".

8. In the **INTRODUCTION**, lines 72 to 74, the sentence "In recent years, researchers have further improved the SS-TENG through materials selection[21] and electrode treatment[25,32–34], promoting it toward practical applications in the ocean." **has been revised as** "Recent advancements in the field have enhanced the SS-TENG through materials selection[21], electrode design[25,32–34], and building harvester networks[35], promoting it toward practical applications in the ocean.".
9. In the **INTRODUCTION**, the sentence "Moreover, based on flow-induced vibration (FIV) phenomena, structures have also been developed to harvest flow energy[35,36]." **has been deleted**.
10. In the **INTRODUCTION**, lines 74 to 80, the sentences "Previous studies have proven that TENGs designed in rolling mode can be easily excited by waves to generate high-voltage outputs. However, under low-frequency wave excitations in the ocean, most TENGs produce outputs of only a few microamps (< 5 μA) but have optimal resistances of giga-ohms (GΩ), which makes it hard and impractical to directly power electronics in the marine environment." **have been revised as** "While it has been proven that TENGs, particularly those in rolling mode, can be effectively driven by wave to generate substantial voltage output, the prevalent low-frequency wave dynamics in marine environments typically lead them to produce very limited current outputs (<5 μA) at optimal resistances reaching giga-Ohms (GΩ). This limitation poses significant challenges for directly employing such TENGs to power electronic devices in the marine environment.".
11. In the **INTRODUCTION**, lines 80 to 86, the sentences "Some researchers attempted to address these issues by designing appropriate mechanical mechanisms to upconvert the motion frequency, known as the frequency up-conversion (FUC) strategy. Whereas, the FUC strategy often weakens the characteristics of TENGs in the low-frequency region and leads to the sweet zone of electromagnetic generators. Regulating the outputs of TENGs by power management circuits is another means to mitigate the low-current, high-internal resistance issue. But unfortunately, the power outputs of TENGs eventually degrade after circuit regulation since electronics in power management circuits consume power." **have been revised as** "Grating electrodes are employed to boost current output and decrease internal resistance by accelerating charge transfer[36], typically in conjunction with sliding mode TENGs[3] and frequency up-conversion strategies (FUC)[24]. However, the inherent sliding friction in sliding mode TENGs significantly deteriorates their output performance and durability[37]. On the other hand, although the FUC strategy can optimize the performance of piezoelectric and electromagnetic generators, it diminishes the efficacy of TENGs in low-frequency applications.".
12. In the **INTRODUCTION**, lines 87 to 91, the sentences "In this work, a rolling mode TENG with multi-tunnel grating electrodes and opposite-charge-enhancement (MO-TENG), as shown in Figure 1a, is proposed for the first time to address the challenges mentioned above in ocean wave energy harvesting. The rolling mode enables low friction between the interfaces for an extended lifespan and makes the TENG effortlessly excited by the wave. The multi-tunnel structure improves the effective electrode area of the TENG and thus enhances the electrostatic induction effect. Instead of the FUC strategy, this study combines the grating

electrodes and ternary tribo-materials to boost the charge transfer rate, triboelectrification, and electrostatic induction processes. The integration of these strategies and approaches led to a great optimization of the rolling-mode TENG in the blue energy harvesting area, producing an instantaneous power density of 192.34 $W/m^3 \cdot Hz$ and a root-mean-square (RMS) power density of 11.33 $W/m^3 \cdot Hz$. **have been revised as** "In this work, a novel rolling mode TENG named MO-TENG is designed and proposed, that utilizes multi-tunnel grating electrodes and the opposite-charge-enhancement effect to harvest wave energy efficiently, as illustrated in Figure 1a and Figure 1b-d. These strategies improve TENGs' performance leading to an instantaneous power density of 185.4 $W/m^3 \cdot Hz$ and an RMS power density of 10.92 $W/m^3 \cdot Hz$.".

13. In the **INTRODUCTION**, lines 94 to 96, the sentence "Furthermore, we developed a self-driven ocean sensing (SOS) system that regulate energy, and integrates commercial sensors and a wireless communication module." **has been revised as** "Furthermore, a self-driven ocean sensing (SOS) system is developed using stacked MO-TENGs and an ultra-low power consumption circuit with the cold-start function to realize wireless self-powered sensing.".
14. In the **INTRODUCTION**, lines 96 to 102, the sentences "When exposed to random wave excitations, the SOS system can successfully realize fully self-powered water quality monitoring and wireless communication. This marks the first instance of TENG accomplishing a self-powered task in real wave conditions." **have been revised as** "The record-breaking performance of the MO-TENG and the well-thought-out design of the energy management circuit endow the SOS system with excellent self-powered sensing and communication capabilities in the marine environment. Remarkably, a buoy fitted with 30 stacked MO-TENG units was tested in a wave tank and the real ocean. This smart buoy successfully realized fully self-powered water quality monitoring and wireless communication tasks and demonstrated significant potential for practical deployment.".
15. In the **INTRODUCTION**, lines 102 to 106, the sentences "Overall, this novel design effectively enhances the TENG's performance and provides a reliable solution for application in the marine Internet of Things (Marine IoT). Our work represents a substantial step forward, bringing TENGs closer to practical application." **have been revised as** "Overall, the design methodology presented in this work notably improves the TENG's performance and offers a reliable solution for use within the Marine Internet of Things (IoT) domain. This study marks a significant advancement, promoting the research of TENGs towards practical applications.".
16. In the **Conclusion**, lines 426 to 435, the sentences "Using stacked MO-TENGs, a self-powered ocean sensing (SOS) system is designed to monitor water quality remotely and wirelessly. A buoy installed with 10 stacked MO-TENGs was submerged in the water tank that has the wave generation ability. Under the simulated wave excitation, the stacked MO-TENGs in the buoy, together with those auxiliary circuit modules, accomplished the tasks of powering navigation light, water quality sensing, data collecting, and LoRa communication. Significantly, the self-powered sensing task was well demonstrated in a random wave tank, with a buoy stacked with 30 MO-TENG units. The in situ TDS values were collected and transmitted to the receiver thrice in 270 seconds. And the transmission distance can be up to 1.13 kilometers." **have been revised as** "Furthermore, a self-powered ocean sensing (SOS) system featuring cold start capability has been developed for remote and wireless water quality monitoring. At the same time, it enables the scalable construction of system-level self-powered nodes of MO-TENG-

based devices for maritime applications, and has been presented and validated under various laboratory conditions. It is worth noting that a buoy equipped with stacked MO-TENGs and the SOS system was tested in a real ocean environment. The intelligent buoy accomplished the self-powered sensing task three times within 920 seconds with a maximum transmission distance of 0.8 km, which marks the pioneering instance of a TENG-based self-powered device serving in a real ocean environment.”.

Comment 2: It appears that TENG with PTFE exhibits significantly higher performance compared to other materials. Please clarify this point. Why did you investigate PDMS?

Response:

The **Reviewer #2** may have unintentionally made a misspelling in the above question, and we guess the reviewer may want to ask, "Why did you investigate PTFE?".

Due to its distinctive negative electron affinity, PTFE has been extensively utilized in developing and constructing TENGs. Its exceptional electron capture capacity and outstanding durability make it one of the best choices for researchers in this area. The choice of PTFE in this study can be justified by two main reasons. First, PTFE balls are easily accessible and pretty robust. Second, previous rolling-mode works using PTFE balls as the rolling part yielded excellent outputs. By following previous research [*ACS Nano* **2019**, 13, 1932−1939; *Nano Energy* **2021**, 84, 105920] and also to facilitate the comparison with them, PTFE is used in this work. The experiment that compares the design using different material combinations should be able to address the reviewer's concern. As shown in Figure 3d, this choice yielded excellent output performance compared with other choices.

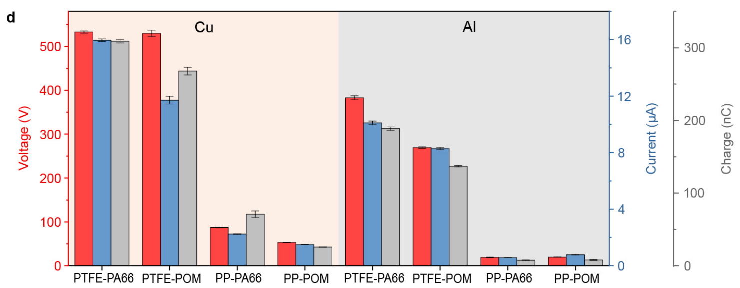


Figure 3d: Output performance of MO-TENGs using different material combinations. Output performance of MO-TENGs using different material combinations.

Comment 3: While POM yields higher TENG performance than Nylon, the reason for choosing Nylon needs clarification. Furthermore, a scientific discussion on the charge transfer mechanisms between PTFE and Nylon should be included.

Response:

The authors thank **Reviewer #2** for providing opinions regarding the materials selections and triboelectrification between interfaces. From the triboelectric series, Polyformaldehyde (POM) indeed exhibits better positive electron affinity than Polyamide 66 (Nylon). However, it is worth noting that both POM and Nylon are at the top of the positive direction with an adjacent ranking. This makes both of them potential material candidates for losing electrons in the friction process. As we all know, the electron transfer process between interfaces is complicated and impacted by various parameters. Even two interfaces with the same materials, like FEP, might cause an evident charge transfer phenomenon [*Nat. Comm.*, **2021**, 12, 5470]. Therefore, in order to determine the optimal material for the friction interface, several prototypes with different materials were fabricated to test the electrification capability as well as the output performance of different materials and material combinations (Figure 3a-c and Figure 3d). From Figure 3d, through the data values of V_{oc}, I_{sc}, and Q_{sc}, the MO-TENG prototype produces a higher output when Nylon is used as the electropositive material, compared to POM.

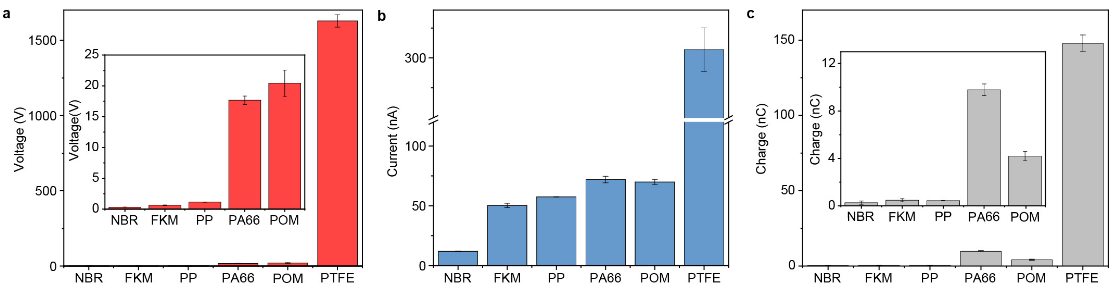


Figure 3a-c: V_{oc}, I_{sc}, and Q_{sc} of the M-TENG using different materials.

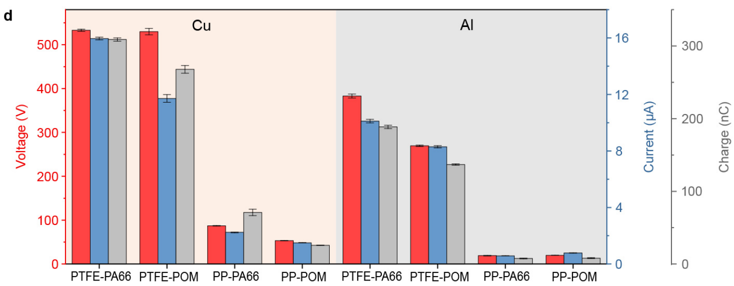
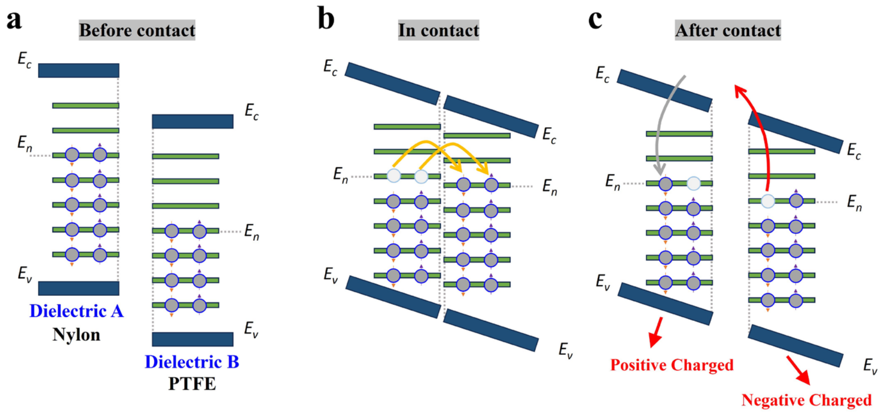


Figure 3d: Output performance of MO-TENGs using different material combinations. Output performance of MO-TENGs using different material combinations.

As for the transfer mechanisms between the PTFE and Nylon interface, the process can be classified as contact-electrification, which occurs during the rolling procedure. Revised Figure S3 shows the contact-electrification between Nylon and PTFE materials, which can be explained using the surface state model.



Revised Figure S3: Contact electrification between two different dielectrics. (a) Before contact, due to their different valence and conduction band structures, Nylon's occupied surface states possess higher energy than PTFE's unoccupied surface states. (b) In contact, some electrons transfer from the Nylon's surface to the PTFE's surface, leading to contact electrification. (c) After contact, positive and negative electrostatic charges stay on Nylon's and PTFE's surfaces, respectively. These charges are indefinitely stable under undisturbed conditions. However, thermal fluctuations and elevated temperatures can cause these electrons to be released from their surface states.

Revision to the manuscript:

1. Figure S3 regarding the contact electrification **has been provided** in the revised **Supporting Information.**
2. In the **Results and discussion**, lines 193 to 194, "The triboelectrification that occurs between two materials, such as PTFE and Nylon, are illustrated in Figure S3, utilizing the surface state model[40]." **has been added**.

Comment 4: A systematic investigation of TENG design parameters, such as grating electrode dimensions and ball size, should be conducted and reported.

Response:

The authors sincerely thank **Reviewer #2** for this constructive comment. We have conducted further experiments and thoroughly discussed the structural parameters of the MO-TENG. Initially, we utilized charge weight density as a metric to assess the impact of dielectric ball weight on charge transfer, thus helping select an appropriate ball diameter. Subsequently, we fabricated prototypes of rolling mode TENGs featuring 4-segment grating electrodes and varying tunnel diameters. Among these, a tunnel diameter of 13.5 mm was selected due to its superior performance. Additionally, we explored the effect of increasing the number of grating electrode segments, finding that higher segments enhance the TENG's current output. The details have been added in the Supplementary Note 1.

Revision to the manuscript:

1. In the **Revised Supporting information**, Supplementary Note 1 **has been added** to discuss the structural parameter impactions. Details are below:

Supplementary Note 1. The diameter selection of dielectric balls and multi-tunnel grating electrodes

PTFE balls are commonly utilized in rolling mode TENGs due to their wide accessibility. Balls of different diameters have different weights. Contact force, as a key parameter in triboelectrification, is related to the weights of balls in rolling mode. To determine an appropriate diameter, transfer charge density was studied with various PTFE balls with different diameters. As presented in Figure N1, we utilized five PTFE balls with different diameters ranging from 5 mm to 18 mm to prototype rolling mode TENGs with different dimensions. The housing of these TENGs was fabricated using a 3D printer, and then one pair of copper films was attached as electrodes. For a fair comparison, we let the width of the copper electrodes in this prototype match that of the PTFE balls, and the copper electrode length equals five diameters. Additionally, the gap between the two electrodes is uniformly set at 1 cm. The details can be found in Figure N2. Here, we define the charge weight density to identify the impact of weight on the output and then help select the diameter of the ball. The charge weight density is calculated as $\frac{the\ transfer\ charge\ (nC)}{weight\ of\ five\ PTFE\ balls\ (g)}$. Figure N3 depicts the dependence between the diameter of the balls used and the weight of five balls, the transfer charge output, and the charge weight density. As the diameter of the balls increases, both the weight of the five PTFE balls and the transfer charge output exhibit similar growth trends. Nonetheless, the charge weight density initially increases with the diameter and then stabilizes once the diameter reaches 12.7 mm. Therefore, the dielectric ball with a diameter of 12.7 mm was selected for subsequent experiments.

Furthermore, we examined the impact of the multi-tunnel grating electrodes' dimensions (tunnel diameter and electrode segments) on the performance of MO-TENGs. Figure N4 presents the waveforms of the short current transfer charge (Q_{sc}) of MO-TENGs with different tunnel diameters and four electrode segments. Among the results in Figure N4, TENG fabricated with a 13.5 mm tunnel diameter yields the best performance. There are two reasons behind this. First, the 13mm diameter tunnel is nearly the same as the 12.7mm diameter of the dielectric ball, potentially hindering the ball's rolling motion. Second, as the diameter of the tunnel increases further, the space between the electrode and the dielectric ball rises, which can diminish the effectiveness of electrostatic induction. Figure N5 shows the I_{sc} output of the TENG with different electrode segments, and inserted photographs depict the corresponding prototype. With the electrode segments increasing from 4 to 14, I_{sc} rises from 0.15 μA to 1.57 μA. This result is attributed to two key factors: Firstly, increasing the number of segments extends the tunnel length, allowing it to accommodate more dielectric balls. Secondly, adding more electrode segments accelerates the charge transfer rate, thereby boosting the short-circuit current output. Therefore, the diameter of the dielectric ball and the multi-tunnel are selected as 12.7 mm and 13.5 mm, respectively, for the subsequent experiments. For the segmentation of grating electrodes, it is beneficial to maximize the number of gratings, ensuring that the dimensions still allow for high integration and ease of fabrication.

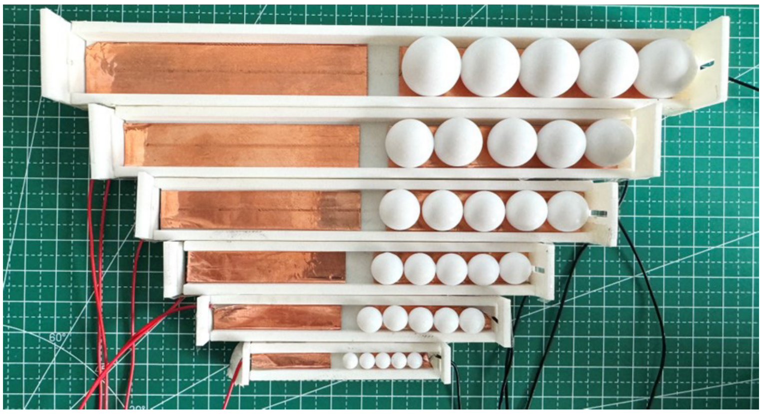


Figure N1: Photograph of rolling mode TENGs fabricated using PTFE balls with different diameters.

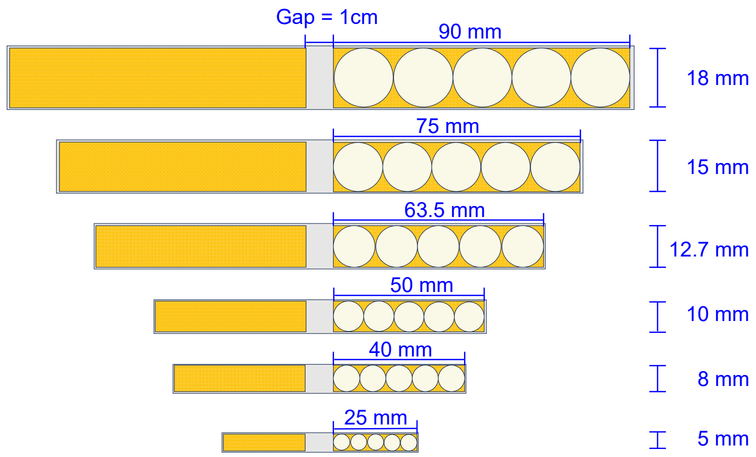


Figure N2: The parameter illustration of rolling mode TENGs fabricated using PTFE balls with different diameters.

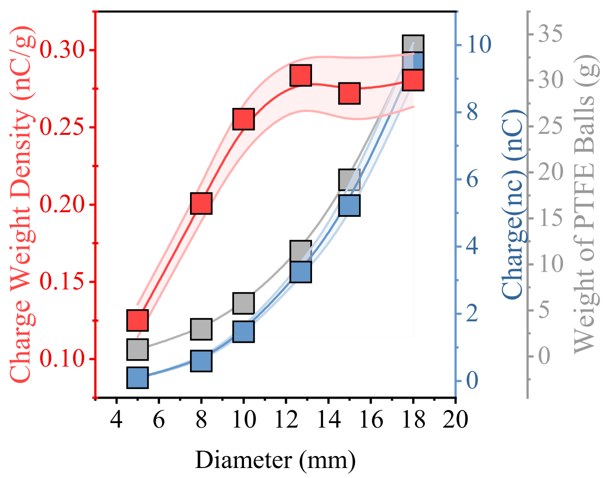


Figure N3: The weight, transfer charge, and charge weight densities of rolling mode TENGs using PTFE balls of various diameters.

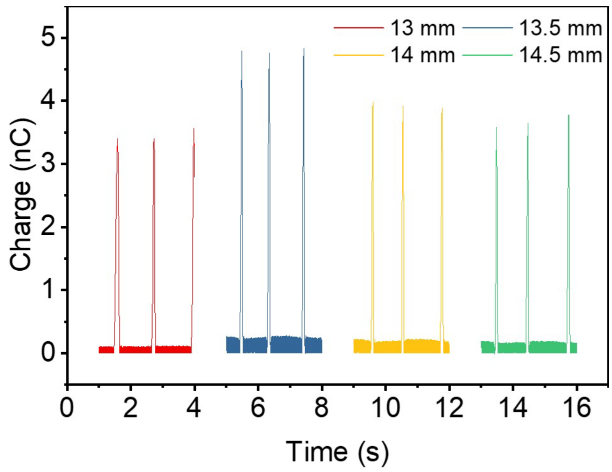


Figure N4: The transfer charge of rolling mode TENGs fabricated with one tunnel and 4 segment electrodes of various tunnel diameters.

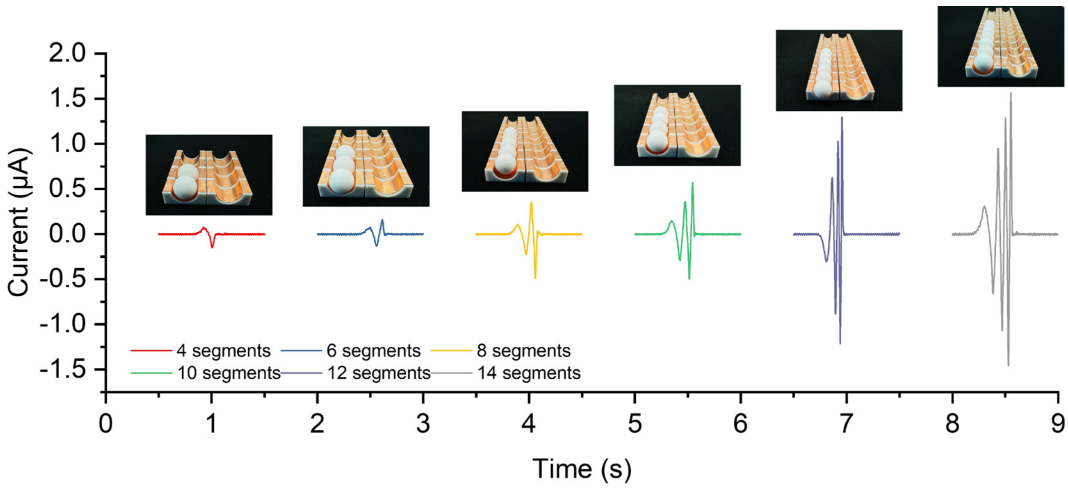
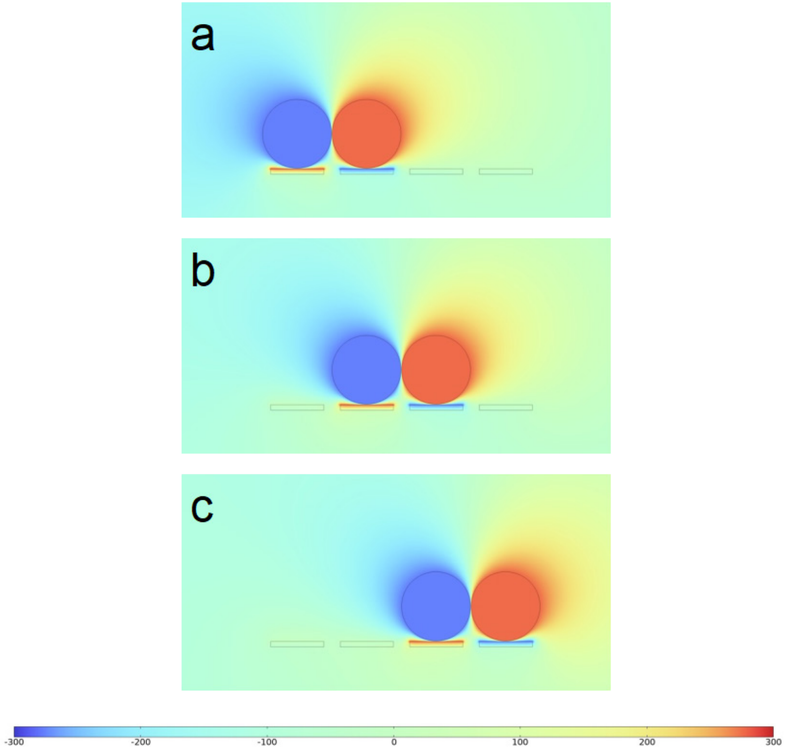


Figure N5: The current output of one tunnel grating electrode TENGs with different electrode segments.

Comment 5: Including simulations would enhance the content of this work and provide additional insights.

Response: The authors thank **Reviewer #2**'s constructive suggestion of conducting simulations to deepen the persuasion of this work, and we fully agree with this viewpoint. We have designed and conducted the related simulation through Comsol Multiphysics to exhibit the mechanisms and output performance of TENGs. The potential distribution simulations of MO-TENGs with two dielectric balls and six dielectric balls were conducted below:



Revised Figure S3: Electric potential distributions of MO-TENGs with two dielectric balls at different positions simulated using COMSOL Multiphysics software.

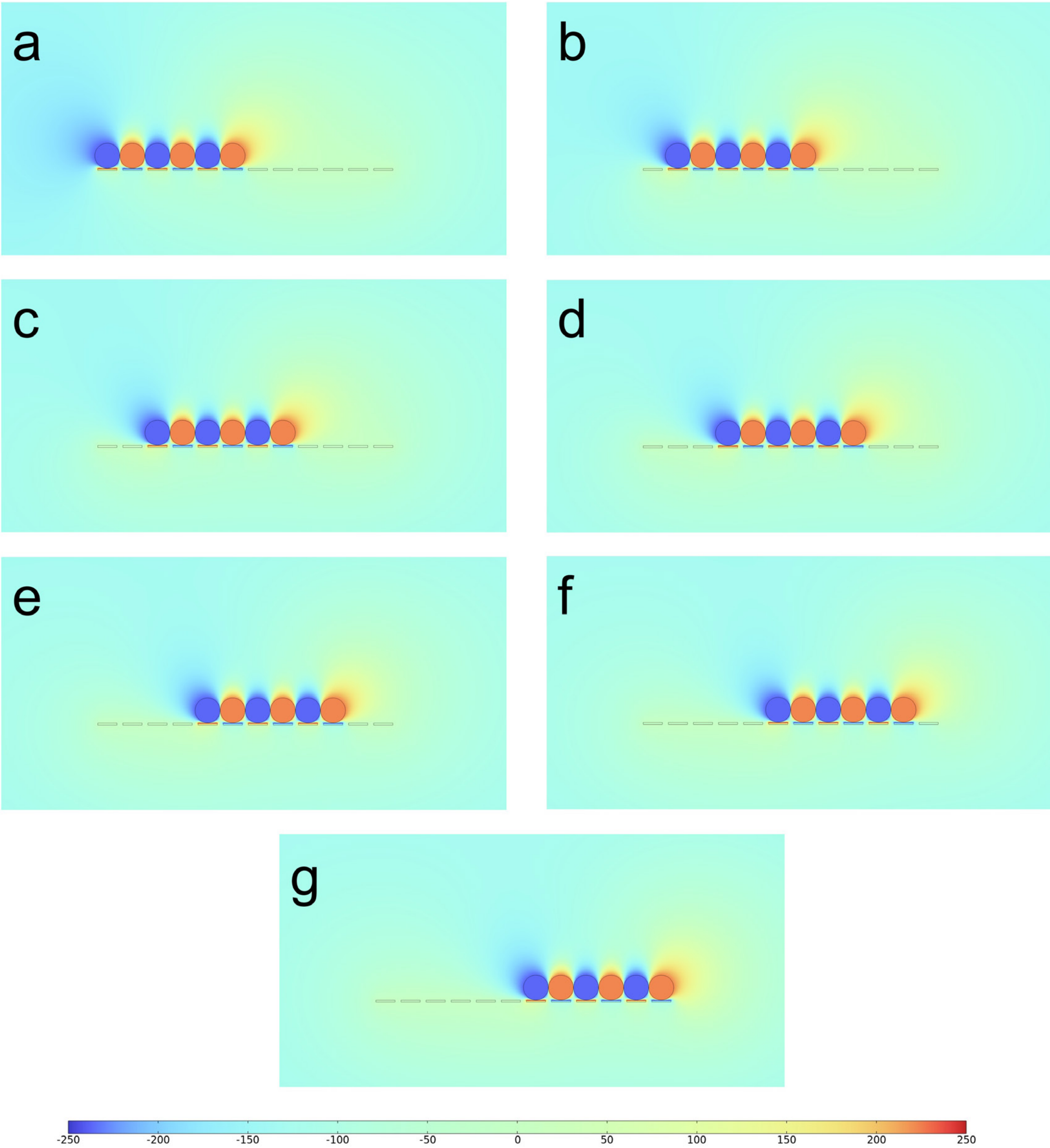


Figure S4: Electric potential distributions of MO-TENGs with six dielectric balls at different positions simulated using COMSOL Multiphysics software.

Revision to the manuscript:

1. Figure S5 and Figure S6 regarding the finite element simulation **have been added** in the revised **Supporting Information**.
2. In the **Results and discussion**, lines 214 to 217, "To further substantiate the proposed mechanism, the potential distributions of the MO-TENG with two and six dielectric balls were simulated using COMSOL (Figure S5-6). It should be noted that the results were obtained under open-circuit conditions." **has been added**.

Comment 6: Considering the potential for enhancing performance, the manuscript could explore the impact of using balls with porous or patterned surface structures.

Response: The authors thank **Reviewer #2** for the insightful recommendation to assess the influence of nanostructure modification on the MO-TENG's performance, highlighting a crucial strategy for improving TENG devices within the realm of material science design.

Due to equipment constraints in our laboratory, preliminary experiments were conducted by employing sandpaper to texture the surface of the dielectric (PTFE and Nylon) balls, which was also a method used in previous works [*Adv. Energy Mater.* **2019**, 9, 1902824; *Nat. Comm.*, **2022**, 13, 3325]. As illustrated in Figure R3, we evaluated the MO-TENG's short-circuit transfer charge following the surface modification of the dielectric balls over durations ranging from 2 to 10 minutes, using 10,000 grit sandpaper. The findings indicate that the alterations introduced did not significantly enhance performance compared to other studies. This outcome could be attributed to two primary factors: 1) the difficulty in precisely controlling and quantifying the modification process undertaken in this trial and 2) due to the inherent durability of those dielectric spheres (PTFE and Nylon), the impact of surface texturing via sandpaper is relatively negligible, as substantiated by the observations presented in Figure 4e.

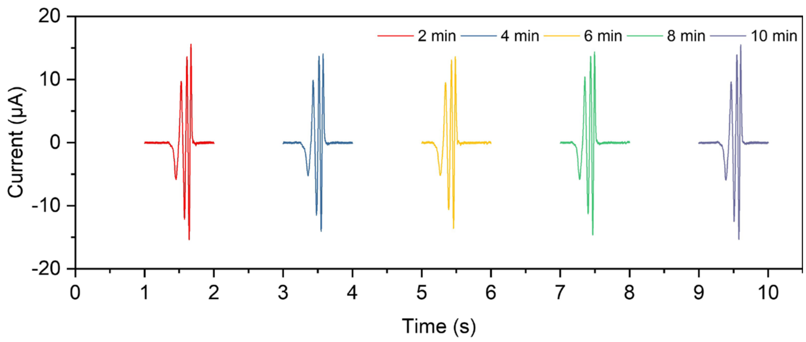


Figure R3: The comparison of the transfer charge of dielectric with different friction treatment times.

According to the previous research, the modification object of the TENGs is always the dielectric or electrode films [*Adv. Energy Technol.* **2021**, 6, 2000916; *Energy Environ. Sci.* **2024**, 17, 885]. However, there exists a notable gap in research concerning the surface modification of dielectric balls, underscoring the urgency for investigation in this area. Thus, comprehensive efforts are imperative to address the challenges associated with this innovative approach, which holds substantial promise for the advancement of rolling TENG technology. This domain will constitute the primary focus of our future research endeavors, and we will explore its critical importance for promoting TENGs' applications.

We extend our gratitude to **Reviewer #2** for your valuable suggestion again, which offered profound insights. This paper has concentrated on improving performance through structural design, leading to significant advancements and results. While previous literature has extensively reported the benefits of surface structural modifications on TENGs' performance, the complexity and many variables involved, particularly the scarcity of modifications oriented towards dielectric spheres, warrant further exploration. We intend to delve deeper into this area in our future research endeavors.

Response to Reviewer #3:

General Comment: The authors report a triboelectric nanogenerator for wave energy harvesting, incorporating some unique and novel features to enhance power output. The authors should be commended for the detailed manuscript, with a thorough discussion of the data included. However, there are major revisions that are required, as listed below.

Response:

We sincerely thank **Reviewer #3** for the compliment on our work. The constructive suggestions are invaluable for us to make significant improvements. We've carefully reviewed each point and offer detailed responses below, aiming to enhance the clarity and impact of our study. The revised details for your specific comments have been highlighted in blue in the manuscript and response letter.

Comment 1: The first sentence of the Abstract refers to a specific geopolitical event. It should be removed/replaced. The authors should focus on discussing the motivation to their work in a general scientific way.

Response:

Thank you for commenting on us to refocus our abstract. We have revised the first sentence to emphasize the broader environmental significance of oceans and the urgent need for innovative energy-harvesting technologies for their preservation. Additionally, we've made further revisions to enhance the overall fluency of the abstract.

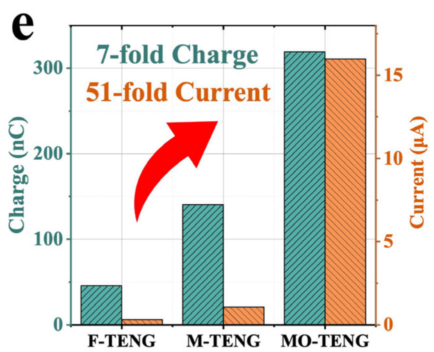
Revision to the manuscript:

1. In the **ABSTRACT**, lines 15 to 20, the sentence "Following the release of nuclear wastewater from the Fukushima power plant, there are increasing concerns about radioactive substance concentration in the Pacific Ocean. Under this circumstance, extensive water quality monitoring will be an urgent need in Asia & Pacific." **has been revised as** "In light of the crucial role of marine ecosystems and the escalating environmental conservation challenges, it is essential to conduct marine monitoring to help implement targeted environmental protection measures efficiently. Energy harvesting technologies, particularly triboelectric nanogenerators (TENGs), have great potential for prolonging the lifespan and enhancing the reliability of sensors in remote areas.".

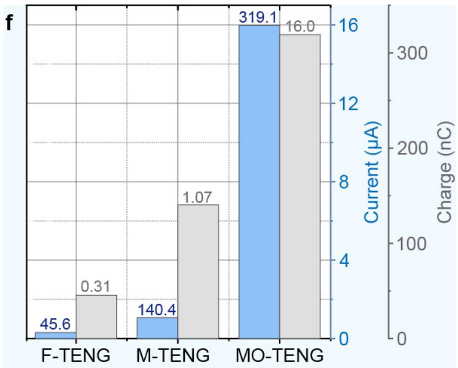
Comment 2: Lines 122-125, the figures for transferred charge and output current appear to be compared against the F-TENG only. Please revise the wording to make this clear since you are not comparing against the M-TENG.

Response:

The authors sincerely thank **Reviewer #3** for this careful and rigorous comment. We have revised Figure 1e to avoid misunderstandings for readers and incorporated precise data values to improve readability.



Original Figure 1e: Comparison of charge and current with F-TENG, M-TENG and MO-TENG.



Revised Figure 1f: Comparison of the transferred charge and current generated by the F-TENG, M-TENG, and MO-TENG.

Revision to the manuscript:

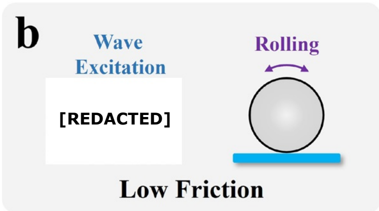
1. In the manuscript, Figure 1 **has been revised and updated.**
2. In the **ABSTRACT**, lines 124 to 129, the sentences "As depicted in Figure 1e, compared with the flat structure TENG (F-TENG)[33] and the multi-tunnel TENG (M-TENG)[32], the transferred charge and output current of the MO-TENG increased to 7 and 51 times, respectively." **has been revised as** "Figure 1f illustrates a substantial enhancement of the MO-TENG's performance, showcasing a transferred charge of 319.1 nC and an output current of 16 μA. Compared with the Flat Structure TENG (F-TENG)[33] and Multi-Tunnel TENG (M-TENG)[32], the MO-TENG exhibits a 7-fold and 2.3-fold increase in transferred charge and a 51.6-fold and 14.95-fold increase in current output, respectively."

Comment 3: A few sub-figures in Fig. 1 are probably not needed since they do not add much information. Specifically, Fig. 1b and 1i.

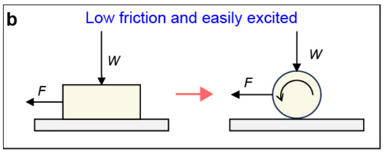
Response:

Thank the reviewer for the valuable suggestion. We have accordingly updated our figures to improve the clarity, logical flow, and readability of the presented information.

Figure 1b-d are designed to list the innovative aspects and principal design attributes of the MO-TENG. Acknowledging **Reviewer #3**'s critique, the original Figure 1b didn't adequately convey the device's features or merits. Thus, we have amended Figure 1b. This revision highlights the rolling mode mechanism's advantages, specifically its susceptibility to activation and the reduced interface friction.

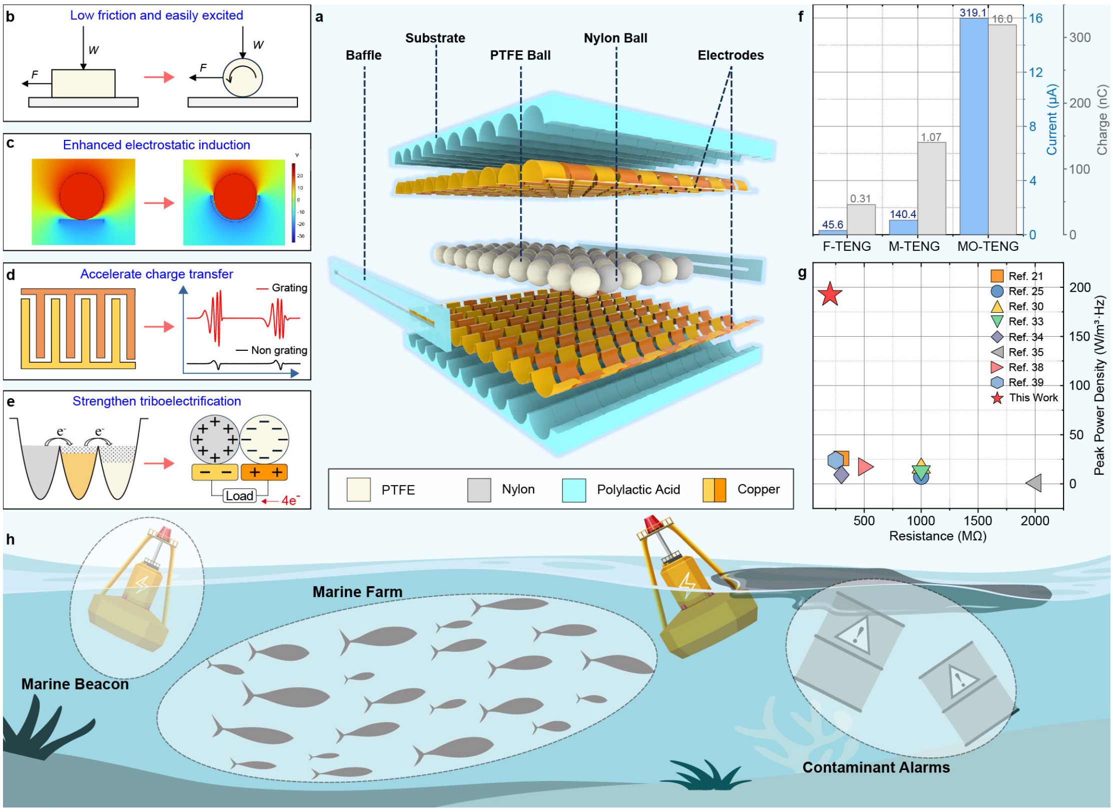


Original Figure 1b: The performance improvement strategy adopted in this work.



Revised Figure 1b: The performance improvement strategy adopted in this work.

Compared with the application scenarios of marine beacons and farms in Figure 1g and Figure 1h, Marine IoT indeed lacks specific situations. We have revised Figure 1g-i into one sub-figure based on the superior performance of MO-TENG and its endowed fully self-powered marine, which can be applied to marine beacons, marine farms, and contamination alarms in ocean environments.



Revised Figure 1: Design and boosting strategies of the MO-TENG.

(a) Structure scheme of the MO-TENG.

(b-e) The performance improvement strategies adopted in this work.

(f) Comparison of the transferred charge and current generated by the F-TENG, M-TENG, and MO-TENG.

(g) Comparison of the peak power and optimal resistance of the MO-TENG with other designs[21,25,30,33–35,38,39].

(h) Potential application scenarios of the proposed MO-TENG.

Revision to the manuscript:

1. In the **RESULTS AND DISCUSSION**, Figure 1 **has been revised.**
2. In the **RESULTS AND DISCUSSION**, lines 114 to 117, the sentences "Dielectric (PTFE and Nylon) balls, as the inertial bodies, can roll forth and back in the housing tunnel under the wave excitation, as shown in Figure 1b." **has been revised as** "Dielectric (PTFE and Nylon) balls serve as inertial bodies that can roll back and forth in the housing tunnel. The rolling mode design, illustrated in Figure 1b, ensures that the TENG can be easily activated and also offers the benefits of low friction and improved durability.".
3. In the **RESULTS AND DISCUSSION**, lines 129 to 131, the sentences "Compared with previous studies in rolling mode design, the MO-TENG utilizes multi-tunnel grating electrodes and opposite-charge-enhancement, which feature a small optimal resistance and a much higher peak power density, as shown in Figure 1f[21,25,30,33–35,38,39]." **has been revised as** "Compared with previous studies of rolling mode TENGs, the MO-TENG has a small internal resistance

and can produce a much larger peak power density, as shown in Figure 1g[21,25,30,33–35,38,39].”.

4. In the **RESULTS AND DISCUSSION**, lines 131 to 135, the sentences “The blueprints of this device in ocean applications are depicted in Figure 1g-i. The MO-TENG can be integrated with the buoys to transform random, low-frequency, high-entropy wave energy into electricity. By powering LEDs, sensors, and communication units, such MO-TENGs can serve the purpose of achieving self-powered ocean node devices, e.g., marine beacons (Figure 1g), marine farms (Figure 1h), and marine Internet of Things (Figure 1i).” **has been revised as** “The blueprints of this device in ocean applications are depicted in Figure 1h. Integrated with buoys, MO-TENGs can be able to convert chaotic, low-frequency, high-entropy wave energy into electricity, powering LEDs, sensors, and communication modules serving marine beacons, farms, and contaminant alarms.”.

Comment 4: Line 141, the architecture of the TENG should be described as cuboid or hexahedral, not cubic.

Response:

The authors sincerely thank **Reviewer #3** for this careful comment. We have substituted the term "cubic" with "hexahedra" for a more precise and visual description. Additionally, we have revised the text to maintain clarity in logic and structure.

Revision to the manuscript:

1. In the **INTRODUCTION**, lines 69 to 72, the sentence "Unlike the RF-TENG with a spherical structure, the SS-TENG was designed with a cubic structure to accommodate more dielectric balls for generating a higher power output.", **has been revised as** "Unlike the RF-TENG, which features a spherical configuration, the SS-TENG was engineered with a hexahedral architecture. This design choice allows for additional dielectric balls, facilitating a greater power output.".
2. In the **RESULTS AND DISCUSSION**, lines 146 to 148, the sentence "In this work, the housing of the rolling mode TENG can be simplified as a cubic, as shown in Figure 2a.", **has been revised as** "In this study, the housing of the rolling mode TENG is conceptually simplified to a hexahedral shape, as depicted in Figure 2a.".

Comment 5: There appear to be some mistakes in Equations (1), (2) and (3). Please check them carefully. For L_(1, F-TENG), the expression lacks the gaps between the balls as shown in Fig. 2b. For H (all three designs), the terms in the brackets should be (delta_1 + delta_2).

Response:

The authors appreciate **Reviewer #3**'s constructive suggestion on parameter research within this study. We have revised Equation (1-3) to clarify and address any potential misunderstandings related to this work.

For the F-TENG, which lacks the *Y*-direction multi-tunnel constraint, the spacing between dielectric balls, as viewed in the *X-Y* plane, varies unpredictably during movement, making evaluation challenging. To address this, a parameter δ_c is introduced to quantify the total gap distance in the X direction of the F-TENG, excluding the space occupied by dielectric balls. Furthermore, we have corrected and updated the calculation of the height for the three designs. We extend our appreciation once more for this delicate and constructive feedback. Furthermore, we have revised the text to maintain clarity in logic and structure, and details are shown below.

Original equation (1-3):

$$\begin{cases} L_{1,F-TENG} = n_1 \mathrm{d} \\ L_{2,F-TENG} = 2n_2 d + g \\ H_{F-TENG} = d + 2(\delta_2 + \delta_3) \end{cases} \quad (1)$$

$$\begin{cases} L_{1,M-TENG} = n_1 (d + 2\delta_2 + \delta_3) \\ L_{2,M-TENG} = 2n_2 d + g \\ H_{M-TENG} = d + 2(\delta_2 + \delta_3) \end{cases} \quad (2)$$

$$\begin{cases} L_{1,MO-TENG} = n_1 (d + 2\delta_2 + \delta_3) \\ L_{2,MO-TENG} = 2n_2 (d + g) \\ H_{MO-TENG} = d + 2(\delta_2 + \delta_3) \end{cases} \quad (3)$$

Revised equation (1-3):

$$\begin{cases} L_{1,F-TENG} = n_1 \mathrm{d} + \delta_c \\ L_{2,F-TENG} = 2n_2 d + g \\ H_{F-TENG} = d + 2(\delta_1 + \delta_2) \end{cases} \quad (4)$$

$$\begin{cases} L_{1,M-TENG} = n_1 (d + 2\delta_2 + \delta_3) \\ L_{2,M-TENG} = 2n_2 d + g \\ H_{M-TENG} = d + 2(\delta_1 + \delta_2) \end{cases} \quad (5)$$

$$\begin{cases} L_{1,MO-TENG} = n_1(d + 2\delta_2 + \delta_3) \\ L_{2,MO-TENG} = 2n_2(d + g) \\ H_{MO-TENG} = d + 2(\delta_1 + \delta_2) \end{cases} \tag{6}$$

Revision to the manuscript:

1. In the **RESULTS AND DISCUSSION**, lines 158 to 160, the revised equation (1-3) **has been added**.
2. In the **RESULTS AND DISCUSSION**, lines 161 to 165, the sentence "Referring to Equation (1-2), the multi-tunnel design introduces the parameters δ_2 and δ_3. These denote the difference in L_1 and L_2 between the F-TENG and the M-TENG, respectively." **has been revised as** "where δ_2 and δ_3 represent the differences in L_1 and L_2 between the F-TENG and the M-TENG, respectively. Assessing the spacing between dielectric balls in the *X-Y* plane presents a challenge. To address this, the parameter δ_c is introduced to quantify the total gap distance along the *X* direction in the F-TENG, excluding the space occupied by dielectric balls.".
3. In the **RESULTS AND DISCUSSION**, lines 179 to 182, the sentence "Substituting the design values into Equation (4) yields $S_{F\text{-}TENG}$ = 44790 mm^2, $S_{M\text{-}TENG} \approx$ 59568 mm^2, and $S_{MO\text{-}TENG} \approx$ 56496 mm^2, indicating that the effective electrode areas of the M-TENG and MO-TENG are enlarged by 33% and 26%, respectively, compared with the F-TENG." **has been revised as** "Substituting the design values into Equation (4) yields $S_{F\text{-}TENG}$ = 44490 mm^2, $S_{M\text{-}TENG} \approx$ 59167 mm^2, and $S_{MO\text{-}TENG} \approx$ 51708 mm^2, indicating that the effective electrode areas of the M-TENG and the MO-TENG are enlarged by 33% and 16%, respectively, compared with the F-TENG.".

Comment 6: Define Q_sc in line 174, in the same way that you define the other important parameters.

Response:

We thank the **Reviewer #3** for pointing out the oversight. We have now clearly defined Q_{sc} in the revised manuscript, making it consistent with the definitions of other parameters to ensure clarity for our readers. Furthermore, we have meticulously reviewed and revised the text to ensure clarity and consistency throughout the manuscript. Details are below.

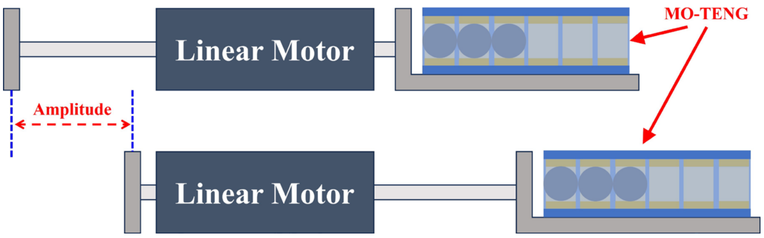
Revision to the manuscript:

1. In the **RESULTS AND DISCUSSION**, lines 185 to 186, the sentence "The Q_{sc} of the two TENGs is evaluated and compared in Figure S2. The Q_{sc} of the two TENGs is evaluated and compared in Figure S2.", **has been revised as** "The short current transfer charge (Q_{sc}) of the two TENGs is evaluated and compared in Figure S2.".
3. In the **INTRODUCTION**, lines 91 to 93, the sentence "A power management circuit is constructed to optimize the capacitor charging performance, resulting in an approximately 150-fold increase in stored energy.", **has been revised as** "A power management module (PMM) is designed to optimize the capacitor charging performance, resulting in an approximately 150-fold increase in stored energy.".
2. In the **RESULTS AND DISCUSSION**, lines 390 to 392, the sentence "The capsule-like buoy will be ballasted (with iron pans near the baseline) to a draft of 0.6m and a VCG of 0.25m (from the baseline).", **has been revised as** "The capsule-shaped buoy was weighted down (using iron blocks positioned near its base) to achieve a draft of 0.6 meters and maintain a vertical center of gravity (VCG) located 0.25 meters above the baseline.".

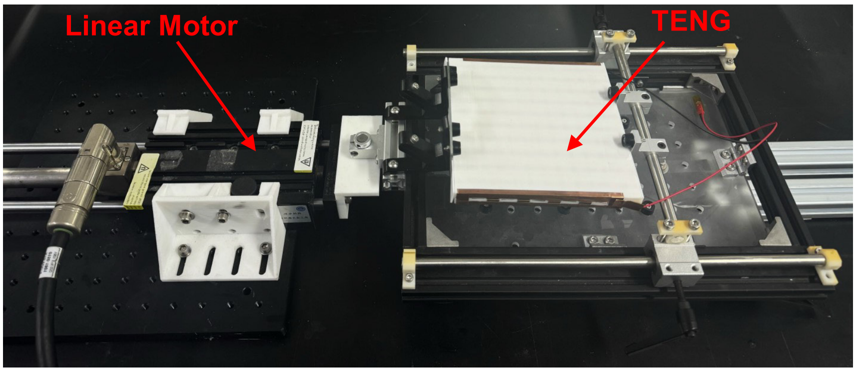
Comment 7: Clarify how you apply the external excitation mentioned in lines 210-211. You could point your reader to a figure showing the setup.

Response:

We thank **Reviewer #3** for highlighting the need for clarity regarding how the external excitation is applied. We have elaborated on this in the manuscript and have directed readers to Figure S10-11, which illustrates the experimental setup in detail.



Revised Figure S10: Illustration of the linear motor test bench designed for generating desired external excitations in the experiment.



Revised Figure S11: The photograph of the linear motor test bench used in the experiment for generating desired external excitations.

Revision to the manuscript:

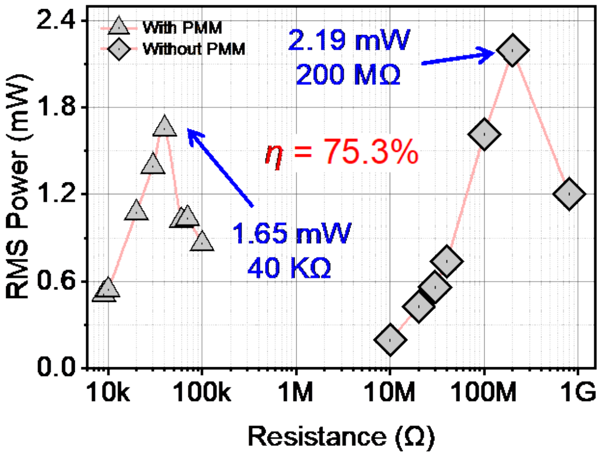
1. In the **SUPPORTING INFORMATION**, the Revised Figure S10-11 **has been added**.
2. In the **RESULTS AND DISCUSSION**, lines 224 to 226, the sentence "A stable external excitation (0.5 Hz, 90 mm peak-to-peak amplitude) is applied by a linear motor.", **has been revised as** "A consistent external excitation with a frequency of 0.5 Hz and a peak-to-peak amplitude of 90 mm is generated using a linear motor, as depicted in Figure S10-11.".

Comment 8: What is the power consumption of the power management module? Can you discuss that against the overall power output that you obtain from the stacked MO-TENG?

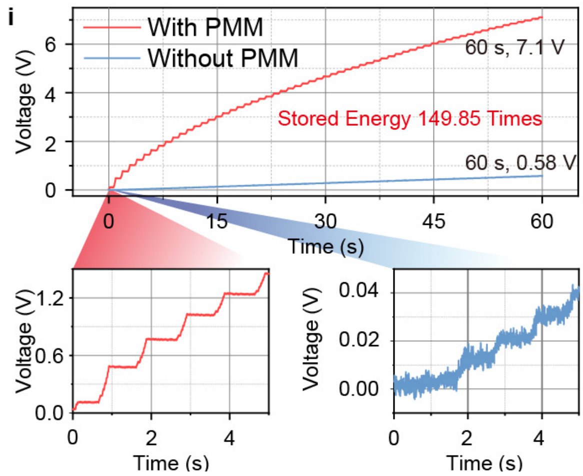
Response:

We are very grateful for your professional advice. We have conducted and added the related experiments in the manuscript and Supporting Information, and the details are shown below:

The resistance dependence of the RMS power of a single MO-TENG unit using and without using PMM was evaluated, as depicted in Figure 4h. The energy transfer efficiency of the PMM is 78.5%, bringing an effective optimal resistance decrease and a significant improvement in capacitor charging performance (Figure 4i).

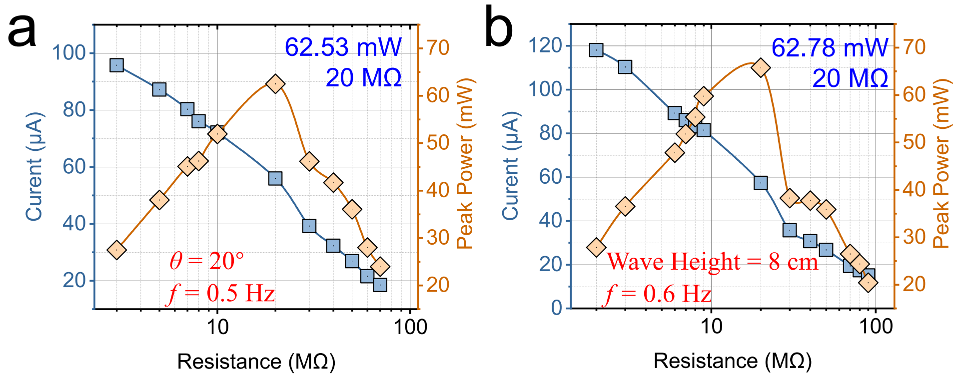


Revised Figure 4h: The resistance dependence of the RMS power of a single MO-TENG unit using and without using PMM.

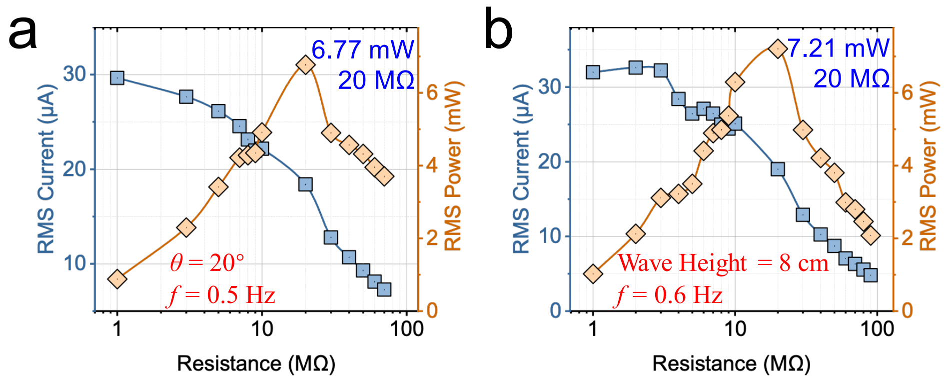


Revised Figure 4i: Comparison of the capacitor (100 μF) charging speeds of the MO-TENG using and without using PMM.

Our research extended to evaluating the performance of a stacked MO-TENG configuration with ten units under the excitation generated by a 6-degree-of-freedom platform and in a wave tank. As illustrated in Revised Figure S26, the stacked MO-TENG achieved a peak output power of 62.53 mW at an optimal resistance of 20 MΩ, under external excitation at 0.5 Hz and 20° on the 6-degree-of-freedom platform. Similarly, in the wave tank environment, the peak power achieved was 62.78 mW at the same optimal resistance of 20 MΩ, under the excitation condition of a 0.6 Hz frequency and 8 cm height regular wave. The RMS output power of the stacked MO-TENG is detailed in Revised Figure S27, showing RMS powers of 6.77 mW and 7.21 mW when excited by the 6-degree-of-freedom platform and wave tank, respectively. The optimal resistance for both conditions is 20 MΩ.



Revised Figure S26: Peak power outputs of a stacked MO-TENG with 10 units excited by a 6-degree-of-freedom platform (a) and in the wave tank (b).



Revised Figure S27: RMS power outputs of a stacked MO-TENG with 10 units excited by a 6-degree-of-freedom platform (a) and in the wave tank (b).

Revision to the manuscript:

1. In the **RESULTS AND DISCUSSION**, the Revised Figure 4h **has been added**.
2. In the **SUPPORTING INFORMATION**, the Revised Figure S26-27 **has been added**.
3. In the **RESULTS AND DISCUSSION**, lines 314 to 324, the sentence "Although a single MO-TENG unit has a limited output, multiple MO-TENG units can be stacked and connected in parallel to boost the output and lower the internal resistance. The resistance dependence of the

peak and RMS power outputs of a stacked MO-TENG having 10 units are shown in Figure 4f and Figure S26-S27. It was tested on a three-degree-of-freedom (3-DOF) swing platform (Figure S28) and in a wave tank (Figure S29). This approach offers a practical solution to meet the power requirements of various electronic devices. However, one issue that lies in the stacked configuration is that the asynchronous movements of the inertial balls across different layers may lead to the neutralization of current, thereby reducing the overall effectiveness. To address this challenge, we have developed a power management module (PMM), to mitigate these adverse effects and maximize the power output. The circuit diagram and other details are shown in Figure 4g and the **Methods** section." **has been added**.

4. In the **RESULTS AND DISCUSSION**, lines 325 to 332, the sentence "As depicted in Figure 4h, the RMS power outputs of a single MO-TENG unit using PMM and without using PMM are evaluated. Under the excitation of 0.5 Hz and 30°, the RMS power is 1.65 mW at a low optimal resistance of 40 KΩ when using PMM, and it is 2.19 mW at 200 MΩ without using PMM. Thus, the energy efficiency of the PMM is about 78.5%, calculated using the following equation: $\eta = P_{RMS,\ with\ PMM}/P_{RMS,\ without\ PMM}$. Despite the decline of the RMS power, the significant decrease in internal resistance due to the use of PMM will facilitate impedance matching and enhance charging speed." **has been added**.

Comment 9: Define VCG (line 368).

Response:

We appreciate the **Reviewer #3**'s attention to detail. The abbreviation VCG stands for vertical center of gravity, while defined in the Supporting Information, was inadvertently omitted in the main manuscript. We have corrected this oversight and replaced it with a more easy-to-understand word.

Revision to the manuscript:

1. In the **RESULTS AND DISCUSSION**, lines 390 to 392, the sentence "The capsule-like buoy will be ballasted (with iron pans near the baseline) to a draft of 0.6m and a VCG of 0.25m (from the baseline).", **has been revised as** "The capsule-shaped buoy was weighted down (using iron blocks positioned near its base) to achieve a draft of 0.6 meters and maintain a vertical center of gravity (VCG) located 0.25 meters above the baseline.".

REVIEWERS' COMMENTS

Reviewer #2 (Remarks to the Author):

The authors have revised the manuscript satisfactorily. I recommend this paper for publication in Nature Communication.

Reviewer #3 (Remarks to the Author):

The authors have carefully addressed my comments and they have improved the manuscript to emphasise the contributions/novelty of their work. I recommend the manuscript for publication.