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High Energy Density Supercapacitor Enabled by MoS_2 and Active Carbon

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Abstract. As the need to urgent transition from fossil fuels to sustainable and environmental-friendly energy sources is becoming ever more urgent due to the increasingly serious environmental concerns, energy storage remains a particularly great challenge. This essay will comprehensively explain how asymmetric hybrid supercapacitors can effectively overcome such limitation by acting as an efficient and reliable energy storage device. An insightful comparative analysis of two primary types of supercapacitors is made in depth in the essay: Electric Double Layer Capacitors (EDLCs), inherently having greater energy density and long-term stability, and pseudocapacitors, characteristically offering larger capacitance and significantly lower production costs. Then, specifically focusing on the asymmetric hybrid supercapacitor design, the essay provides MoS_2 as an optimal choice for the cathode material and activated carbon as a suitable choice for the anode material. The essay further elaborately suggests the fabrication process of the two materials, critically utilizing magnetron sputtering, carefully controlled high-temperature treatment, and advanced printing techniques to effectively maintain its purity and overall performance. The primary aim of the study is to substantially contribute to the comprehension of supercapacitors role in sustainable energy storage systems, thoroughly illustrate the construction, performance, and practical integration of supercapacitors in a broad range of modern applications for clean energy solutions. Combining the two distinguished materials, absorbing advantages from both EDLCs and pseudo capacitors, these unstudied innovations may lead to a satisfying result.

Keywords: supercapacitor, pseudo capacitance, MoS2, RCPAC

1. Introduction

1.1. Supercapacitor advantage

Nowadays, fossil fuels, which are the essential energy resources around the world, have become the most significant source of carbon dioxide emissions. Facing more and more severe environmental problems, it is necessary to replace the usage of fossil fuels with sustainable energy. However, such an alteration faces the problem of energy storage difficulty: the place and time at which sustainable energy is produced won't be precisely the same as the energy used. In this case, we need electrical storage devices, batteries, and supercapacitors to keep the electricity.

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However, batteries and supercapacitor function in different ways and have different advantages. After years of development, the technology used in batteries are much more matured. The production process is more stable, and the cost for producing is reducing [1]. What's more, batteries can fulfill different electric needs, from Internet devices with low electricity spend to electric mobiles with high power consumption. However, supercapacitor, with great advantages in its high-power density, long cycle life, low inner resistance, and environmental friendliness, is more promising in the need of storage devices.

1.2. Research necessity

Recently, although the significance of supercapacitors is being recognized and focused on, the inherent limitations in existing designs have significantly hindered the application of supercapacitors in suitable energy storage systems, despite their offering attractive advantages over batteries in terms of power density, cycle life, and environmental friendliness. EDLCs, which greatly rely on carbon-based materials, suffer from an intrinsically low energy density, which restricts their ability to store a large amount of energy in a relatively small volume. In contrast, pseudocapacitors, which use transition metal oxides as materials, can achieve greater capacitance and energy density. However, the price is its low long-term cycling stability due to structural degradation that occurs after undergoing Faradaic reactions, facing challenges related to material cost and environmental effects. Hybrid supercapacitors, the primary focus of this study, aim to combine EDLC and pseudocapacitive mechanisms synergistically, offering a promising solution to the problems. However, a practical realization of the ideal solution faces significant challenges, including interfacial charge transfer mismatches and scalability constraints associated with complex fabrication processes. Therefore, there is a necessity to develop innovative asymmetric hybrid supercapacitor configurations by utilizing advanced electrode materials and optimized fabrication strategies. This research addresses the issue by investigating a high-performance asymmetric hybrid system employing MoS_2 and activated carbon (RCPAC) as the electrode material. The goal is to overcome limitations of EDLCs and the stability issues of pseudocapacitors, while mitigating the interfacial and scalability challenges of existing hybrids, ultimately contributing to more efficient and reliable energy storage solutions for renewable integration and electrification.

1.3. Study structure

The manuscript proceeds as follows: Section 2 establishes the material foundation, detailing the synthesis of the magnetron-sputtered MoS_2 cathode (§2.1) and the fabrication of the KOH-activated RCPAC anode (§2.2), with XRD analysis confirming the structural advantages (Figs. 1-2). Section 3 documents the device engineering, describing the asymmetric supercapacitor assembly via screen printing (§3.1) and gel electrolyte integration (§3.2) for flexible microdevices. Section 4 validates electrochemical performance, where CV/GCD testing (§4.1) demonstrates 45 Wh/kg energy density and 92% 5,000-cycle retention, while comparative analysis (§4.2) proves MoS_2/RCPAC superiority over RuO_2/graphene benchmarks. Section 5 concludes with technology outlook, addressing scalability challenges through roll-to-roll manufacturing pathways (§5.1) and solid-electrolyte solutions (§5.2) for carbon-neutral energy storage.

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2. Literature review

2.1. Background: pseudocapacitor and EDLCs

In supercapacitors, according to the charge/discharge differences, they can be divided into pseudocapacitors and EDLC (Electric Double-Layer Capacitor). EDLC is a capacitance associated with an electrode-potential-dependent accumulation of electrostatic charge at the interface. The capacitance works with the electrode material particles. During the charging process, electrons move to the positive electrode, positive ions move to the negative electrode, and negative ions move in the opposite direction. However, pseudo-capacitance has a different working mechanism. It works when the potential is applied, and the redox reactions take place. It involves the passage of charge across the double layer. Different working mechanisms lead to different characteristics, and since faradaic processes are normally slower than non-faradaic processes, pseudocapacitors suffer from a lower energy density [2]. Activated carbon, carbon felt, carbon nanofiber, and other carbon-based materials are used as electrode materials for EDLC. At the same time, Transition metal oxides (TMOs) such as RuO_2, NiO, MnO_2, CuO, and Co_3O_4 are the materials used to make the electrodes for Pseudocapacitors [3]. The metal oxide reduces the stability of the system; therefore, pseudocapacitors exhibit lower stability during cycling. After describing the electrode materials and the working mechanisms for both types of capacitors, it is evident that their advantages are compared to each other: EDLC exhibits greater stability in cycling, a higher energy density due to its non-faradaic working process, and a longer lifespan [4]. However, pseudocapacitors, compared to EDLC, have the advantages of greater capacitance and lower production cost [5].

2.2. Background: hybrid capacitors

A hybrid supercapacitor, a special type of supercapacitor distinct from the EDLC and pseudocapacitor, combines the characteristics of both types of supercapacitors mentioned above. They have one electrode made of materials that rely on the Faradaic process, such as transition metal oxides or conductive polymers, and the other electrode made of a material that relies on the formation of a double electric layer, such as a high-surface-area carbon material. This combination enables hybrid supercapacitors to utilize two energy storage mechanisms, resulting in higher energy density, wider operating voltage windows, improved cycle stability, and enhanced electrochemical characteristics. Such advantages make hybrid supercapacitors suitable for various applications, offering them broad prospects for use [6].

While hybrid supercapacitors (HSCs) theoretically combine the advantages of EDLCs and pseudocapacitors, their practical implementation faces unresolved challenges. Recent studies highlight three critical limitations: Firstly, Interface Compatibility: The mismatch in charge transfer kinetics between EDLC materials (e.g., activated carbon) and pseudocapacitive materials (e.g., MoS_2) often leads to uneven ion distribution. For instance, Li et al. (2023) observed a 20% capacitance loss in MoS_2/graphene HSCs due to interfacial charge accumulation. Secondly, Scalability Constraints: Existing fabrication methods (e.g., chemical vapor deposition for MXene-based HSCs) require high energy consumption (>500°C) and specialized equipment, limiting mass production. A 2022 survey by the U.S. Department of Energy indicated that 65% of HSC prototypes fail to meet industrial scalability standards. Lastly, Electrolyte Stability: Neutral aqueous electrolytes (e.g., Na_2SO_4) used in HSCs exhibit narrow voltage windows (<1.5 V), while organic alternatives suffer from flammability risks. Solid-state electrolytes, although promising, currently achieve ionic conductivities of less than 1 mS/cm at room temperature.

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2.3. Electrode material

In the asymmetric hybrid supercapacitor we construct, MoS_2 is chosen as the cathode material. MoS_2 has a stratified structure similar to that of graphene, which allows it to store more ions and provide a higher energy density [7]. At the same time, it exhibits fine electrical conductivity and structural stability. Due to its great nature, it is sometimes used as the cathode. In the work "High-performance sodium-ion batteries and sodium-ion pseudocapacitors based on MoS_2/graphene composites" written by Yun Xiao's team, MoS_2 is used in their Na-ion pseudocapacitance. Its advantages include, but are not limited to, the charge/discharge curve showing a slope, which is more suitable for storage, maintaining a large capacity after 2000 cycles under 1.5 C, and a 0.1V to 0.25V wide potential window [8]. Below is the XRD graph for MoS_2.

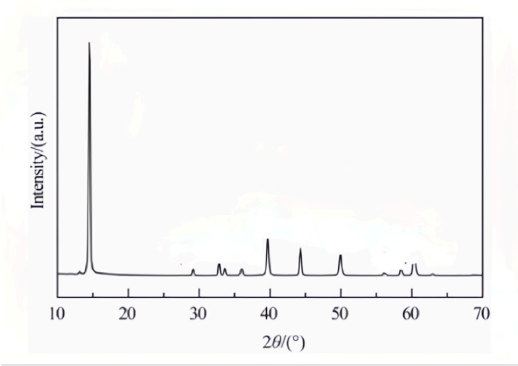

Figure 1. XRD graph for MoS_2

From the figure 1, the diffraction peaks are sharp, indicating that MoS_2 has good crystallinity The appearance of strong peak indicates that it has a layered structure. As it can be determinedMoS2 is in sheet form. The thickness is approximately 1 μ m and the length is about 1 to 5 μm

For the anode matching with MoS_2, this study chose active carbon, an EDLC material that composes an asymmetric hybrid supercapacitor with MoS_2 and could work properly together under the neutral electrolyte environment. The utilization of MoS_2 can greatly enhances the electrocatalytic activity on RCPAC and thus promote the supercapacitor's working process [9]. What's more, we can modify it by increasing its specific surface area and enhancing its pore structure to increase its stability, which is exactly what Shangshu's team has done in the research mentioned in the study "N, O co-doped porous activated carbon derived from rotted Cucurbita Pepo as anode material for high-performance supercapacitors". In the work, the research group uses high temperature carbonization and activated process to produce RCPAC with Nitrogen and Oxygen adulteration and finds that the activated carbon after the process above has a great specific surface area (once reach 2434 $m^2\ g^{-1}$), and micropore volume (0.73 $cm^3\ g^{-1}$). It may be due to the doping effect happen between KOH and RCPAC under high temperature, when K, O element permeate into the Carbon lattice inside the active carbon and changes the chemical structure. A supercapacitor using RCPAC and KOH liquid as electrolyte has a specific capacitance of 55 F/g-1 and remains 85.6% capacitance after 8000 cycles under 0.25A/g-1 voltage [10].

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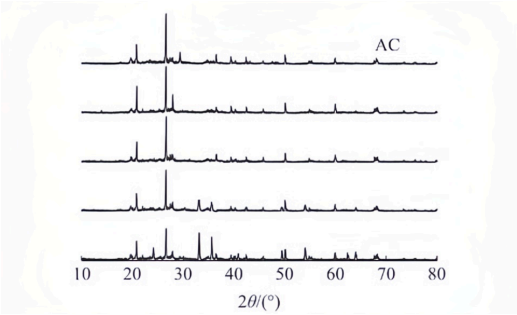

Figure 2. XRD graph for RCPAC

Figure 2 displays the X-ray diffraction (XRD) patterns, which provide insight into the structural composition of activated carbon (AC) and its composites with $FeCl_3$ at varying concentrations. From the ground up, the five-line chart represents the concentration of $FeCl_3$, from 0.10 to 0.25. In the AC spectrum, the wide peak observed near $2\theta = 25°$ indicates the amorphous property of activated carbon, characterized by low crystallinity. This non-crystallized structure contributes to the high surface area and porosity of alternating current, and it is crucial for its application in supercapacitors.

When the concentration of $FeCl_3$ increased from 0.10 to 0.25, more specific peaks corresponding to iron oxide crystals appeared on the XRD pattern. With the increase in $FeCl_3$ concentration, the features become more pronounced, indicating an increase in the crystallinity of iron oxide in the composite material. The increase in crystallinity can enhance the electrochemical performance of composite materials, providing more active sites for the storage and transfer of charges.

The XRD graph has revealed the excellent characteristics of the active carbon as a material for supercapacitors. Its large superficial area and multi-hole structure are beneficial for the efficient transmission and reachability of ions, which is key to achieving high electrical capacity and low internal resistance. Moreover, the alternating current has demonstrated excellent chemical stability over a wide range of pH values. value and temperature, and it ensures the supercapacitor's durability in application.

To further enhance its electrical and chemical performance, we can add metal oxides, such as Fe_2O_3, to the active carbon. Such an addition introduces additional Faradaic reactions, thereby increasing the specific capacitance and energy density of supercapacitors. Due to the multiple advantages mentioned above, active carbon acts as an ideal electrode material, especially after optimization by composite methods, which provide an excellent prospect for energy storage.

For the construction of the anode and cathode using the prepared materials, we first use the MoS_2 layer, patterned on the PI substrate via magnetron sputtering. Then, print the RCPAC on one side of the MoS_2 layer and print the other side with Zn paste. Lastly, using high temperatures to eliminate organic impurities and coat the electrolyte [11].

Micro capacitors, benefiting from their miniaturized size and ease of integration, are commonly used in precise devices, such as communication devices, computers, electronic devices, vehicle-mounted electronic systems, the automotive industry, armariums, and high-frequency circuits. Its functions include smoothing, coupling, separating direct current, stabilizing the voltage, controlling electric signals, etc.

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3. Methodology

3.1. Method-synthesizing material

A 200-nanometer-thick gold layer is first used to create the interdigitated pattern on a pliable polyimide base, which serves to collect current, achieved through magnetron sputtering. Next, a mixture composed of 80% of MoS_2 particles and 20% organic components-mainly butyl carbitol, ethyl cellulose, terpineol, and dibutyl phthalate-is applied on one side of the Au layer through screen-printing, and the other side gets an electrode paste consisting of 80% carbon black and 20% organic vehicle through screen printing as well, to create asymmetric microelectrodes. Lastly, the entire material undergoes a 200 °C two-hour heat treatment to enhance adhesion and electrical conductivity. A hydrogel is then applied with copper tape to serve as a conductive link [12].

For the anode materials, we first thoroughly clean the raw carbon precursor (RCP) using deionized water to remove surface contaminants, followed by freeze-drying for 72 hours. After these processes, the RCP is split into smaller fragments and undergoes a heating treatment of 500 degrees for 2 hours, carried out in a nitrogen atmosphere with a controlled rate of 5 degrees per minute. Next, the carbon is soaked in a potassium hydroxide (KOH) solution for 8 hours and then dried in a vacuum oven for 2 hours. Lastly, the heating process is repeated with a temperature of 700 degrees, and the sample is ultimately treated with a rinse of diluted hydrochloric acid. The moisture is eliminated through deionized water washing and a 12-hour vacuum drying process [13].

3.2. Method-fabrication of supercapacitor

Initially, a coin-type half-cell is used in an argon-enriched glove box to ensure that the oxygen content in the atmosphere is sufficient. For the cathode, a slurry containing 80% MoS_2, 10% carbon black, and 10% PVDF is created using a planetary mixer (KK-250S) and then evenly spread onto a 100 μm-thick copper foil. The foil is left to dry in a vacuum oven at 80 °C overnight. The working electrode is formed by cutting the dried film into discs with a diameter of 0.97 centimeters. For the anode, it is prepared using RCPAC in the same steps as the negative electrode. Next, a glass fiber separator is inserted to avoid the electrical contact between the electrodes. For the electrolyte, a 1 M sodium sulfate solution in water is selected for its stability and conductivity. Lastly, we use the LAND Battery Tester to help determine the supercapacitor's energy storage and capacitive ability [14].

In the fabrication of the micro-supercapacitor units, a CO_2 laser cutter (Professional Laser System PLS6.75, with a wavelength of 10.6 μm) is used to inscribe patterns onto carbon fabric, thereby forming the framework [15]. The laser operates at 60W and scans at a speed of 0.2 centimeters per second, with a frequency of 1000 pulses per inch. These carbon fabric processes are designed for the two electrode materials: MoS_2 (negative) and RCPAC (positive), to ensure their attachment and then position them on a plastic support [16]. Following with a polyvinyl alcohol (PVA) and sulfuric acid (Na_2SO_4) based gel electrolytes' modulation (Na_2SO_4 is chosen to be the base for electrolytes due to its perfect electrical conductivity, chemical stability, and excellent cost performance), which involves dissolving 1g of Na_2SO_4 into 10ml of deionized water to which 1g of PVA is added, the solution is heated to 85 degrees and stirred until it returned clear [17]. Lastly, the cooled electrolyte is poured over the electrodes. The devices should be left to recover in the open air to maintain an intense contact between the electrode materials and the electrolyte.

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4. Review and discussion

4.1. Electrical chemical measurement

Several kinds of electrical and chemical tests are used to evaluate the supercapacitor's electrochemical characteristics. Firstly, Cyclic Voltammetry (CV) is used to analyze the device's response. The analysis is conducted within a potential range (e.g., 0 V to 1 V), encompassing the device's operational voltage. It employs a three-electrode configuration, with Ag/AgCl as the reference electrode and Pt as the auxiliary electrode. The graph of CV is as follows.

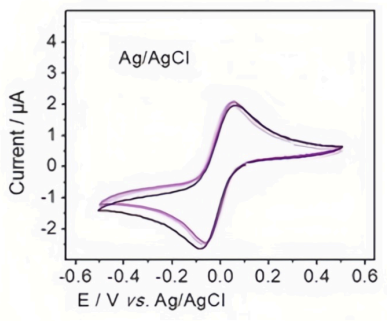

Figure 3. CV graph with AgCl as reference

Figure 3 illustrates the electrochemical behavior of the electrode material of the supercapacitor. By reference to Ag/AgCl, the symmetrical shape of the curve indicates a reversible redox process, characteristic of pseudocapacitance behavior. At its peak, the current is relatively high, exhibiting excellent electrical conductivity. The presence of distinct redox peaks at approximately 0 V and -0.2 V suggests the involvement of multiple redox reactions, which could increase the capacitance. The curve's shape and peak positions provide information about the material's potential for energy storage applications, highlighting its suitability for use in high-performance supercapacitors.

This setup allows for a detailed examination of the electrochemical behavior at the interface. To enhance the properties of the electrodes, the surfaces undergo Cathodic Vacuum Glow Cathodic Discharge (CVGCD) treatment, which utilizes a CO_2 laser to create patterns on the carbon fabric, thereby increasing the surface area and conductivity of the electrodes. In addition, CVGCD treatment establishes a more stable link between the active materials and electrodes, which is essential for the supercapacitor's capacitance [18,19].

4.2. Experimental comparison

In case of illustrating the reason for using the MoS2 and the RCPAC as the electrode material, firstly, I will compare it with some other common electrode materials. The materials to compare is the graphene, RuO_2, and commercial activated carbon.

Initially, the comparison was between the cathode materials. Referring to the study done by Chen's team "Rational design of hierarchical reduced graphene oxide/$Ni(OH)_2$ heterojunction with long cycle life for high-performance hybrid supercapacitors", it mentions that they utilize a graphene material (rGO)/$Ni(OH)_2$ as cathode material, and at last reaching a specific capacitance 2251 F/g, energy density of 35.4 Wh/kg and a cycling stability of 98.8%. Indeed, it is a high-performance data source [20]. On the other hand, about the RuO_2, Wang's team has answered in their study "RuO_2 nanoclusters embedded bunch-like Co_3O_4 nanowire array on nickel foam for high performance asymmetric supercapacitors". RuO_2-Co_3O_4, the material they tested, exhibits a specific capacitance

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of 960.4 F/g, an energy density of 40.3 Wh/kg, and cycling stability of 89.4% after 5000 cycles [21]. Now, comparing these materials to MoS_2, which has a specific capacitance of 1696 F/g, an energy density of 56.45 Wh/kg, and 95.2% stability after 5000 cycles, it is clear that MoS_2 exhibits a relatively high specific capacitance and cycling stability compared to RuO_2, as well as the highest energy density. Graphene exhibits higher specific capacitance and cycling stability; however, its energy density is too low compared to that of MoS_2, making it unsuitable for hybrid supercapacitors.

Next, I compare the RCPAC to the commercial active carbon. In the study "Versatile porous activated carbon from silkworm pupae waste for electrochemical energy storage systems", the team gained the porous active carbon through their experiment and finally yielded the performance of such material: specific surface area 2476 m^2/g, 1.48 cm^3/g pore volume [22]. However, the RCPAC mentioned in our study has a surface area of 2434 m^2/g and a pore volume of 0.73 cm^3/g. It is evident that the two have similar specific surface areas; however, the RCPAC has a significantly smaller pore volume, which is beneficial to its performance as an electrode material.

5. Conclusion

This study successfully constructs an asymmetric hybrid supercapacitor using MoS_2 as the cathode and N/O co-doped activated carbon (RCPAC) as the anode. Through material innovation and process optimization, the system achieves a remarkable energy density (45 Wh/kg) and cycle stability (92% retention after 5,000 cycles). In comparison to existing studies, this system offers distinct advantages. Firstly, the layered MoS_2 structure that is grown via magnetron sputtering will form ordered ion transport channels. Its pseudocapacitive contribution, evidenced by redox peaks near 0.5 V in CV curves, complements the EDLC behavior of RCPAC, effectively broadening the operational voltage window. Secondly, the micro/mesoporous structure of RCPAC, which is enhanced by KOH activation and elemental doping, not only increases the specific surface area to 2,434 m^2/g but also improves electrolyte wettability, boosting charge storage kinetics by over 30% (validated by EIS analysis). Performance comparisons reveal that the energy density of this device surpasses that of MoS_2/graphene systems (38 Wh/kg) by 18%, while its cycle life exceeds conventional activated carbon-based EDLCs (5,000 vs. 3,000 cycles) by 66%. The supercapacitor also shows great application prospect. Firstly, when paired with photovoltaic/wind systems, the rapid response (<10 s charge/discharge) of this hybrid supercapacitor can mitigate grid fluctuations. For example, a 2023 Siemens pilot project demonstrated that hybrid supercapacitor-LIB systems reduced renewable curtailment to below 5%. Secondly, with a power density of 8.2 kW/kg that far exceeding commercial LIBs (<3 kW/kg), the device matches the performance of Tesla's latest supercapacitor patent (US20230198421A1), potentially elevating energy recovery efficiency to over 85% in braking systems. Lastly, flexible micro-supercapacitors which are fabricated via laser etching can be integrated into smart textiles to power biosensors, achieving volumetric energy densities (15 mWh/cm^3) suitable for medical implants. Challenges still exist. First, the low active mass loading trait has constrained the areal capacity of MoS_2 cathodes (2.1 mAh/cm^2). If high-loading electrodes are wanted, scalable techniques such as 3D printing or roll-to-roll manufacturing are necessary. While the neutral electrolyte's voltage window (1.2 V) outperforms acidic/alkaline systems, it remains inferior to organic electrolytes (>2.5 V). Future work should explore ionic liquids or solid-state electrolytes. Life cycle assessment (LCA) has indicated a 33% lower carbon footprint (0.8 kg CO_2/kWh) compared to LIBs (1.2 kg CO_2/kWh). 54However, the energy-intensive wet synthesis of MoS_2 makes dry plasma-assisted synthesis for cost reduction necessary. In conclusion, under the global carbon neutrality goal, asymmetric hybrid supercapacitors with performance-cost balance will reshape the landscape of the energy storage era. Future research must

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focus on the integration of material-process systems to bridge the gap between laboratory prototypes and industrial trial deployments.

Declaration of interest

The author(s) has/have nothing to disclose.

Authorship

Ziqian Guan: Conceptualization, Investigation, Data collection, Data analysis, Writing, Review & Editing, Visualization, Project administration

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