Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The authors present a short overview on the recent progress of MOFs for electrochemical energy storage devices, which is a hot topic. This article is timely. The manuscript is written well. I am pleased to recommend its publication.

Some minor points:
1) As this article deals with only MOFs, not including MOF-derived materials, the title “MOF Design Strategies for Functional Materials in Electrochemical Energy Storage Devices” might be modified because it may mislead readers as “for Functional Materials” here may include “MOF-derived materials” in the MOF/energy field.
2) The authors may briefly mention MOF-derived materials (at least a paragraph with some literature references) even if not giving a specific section.
3) Some recent comprehensive reviews on this topic may benefit readers’ understanding of this field (for example, Chem, 2, 52-80 (2017); Joule, 2, 2235-2259 (2018)).
4) Better to give more detailed discussions on the limit/drawbacks/bottle necks and possible solutions regarding the use of MOFs for energy storage.

Reviewer #2 (Remarks to the Author):

I have reviewed the review article by Thoi and co-workers on "Elucidating Design Strategies for Functional Materials in Electrochemical Storage Devices". I enjoyed the review article and found it easy to follow. I also learned a lot about these systems and why porous materials offer something unique that non-porous materials would suffer from.

I have a few suggestions that I think would help improve the text.

1) Page 1 line 17/18 states "Current technologies are fundamentally limited by materials properties and require new, creative solutions." Do the authors mean "existing materials". Every application is dependent on materials properties. The sentence needs some repair.

2)on line 26 of page 1, the authors state that MOFs are class of porous crystalline materials. However, as evident by the itation of Bennett's work, the author is clearly aware that MOFs are not always crystalline but they still have application. For a manuscript, I wouldn't mind the use of crystalline. However, for a review article, I think it's best to keep the intro broad and covering all the recent literature.

3) I think it would be worth stating in the intro why porosity is important. This should come in early on. Otherwise, the field of coordination polymers fits all the same criterion without needing porosity. I wouldn't attack coordination polymers in the paper, but simply state the need for porosity that other organic-inorganic hybrid materials could not support.

4) In section 2.3, the authors discuss M2dsbdc and M2dobdc. I think the authors have over-simplified the data here. If M-S bonds are so much better, then why is the M-O only 10x worse than the Fe-S while the Fe-S is a million-fold better than the Mn-S. There seems to be more going on than the importance of soft-soft interactions. I think it is worth exploring or clarifying.

5) It is difficult to say due to the reference style having "Et. al" in it. However, I would like to ensure that proton conductivity includes references from Shimizu who has put a lot of work into this field.

6) Under Porosity for tuning ionic conductivity, the authors state "likely due to the insufficient pore size to accommodate Na+ transport. to address this issue ..." I struggle with this. There is no data to support these statements. It's not clear from the writing that the cited references tried Na+ and it failed. There is no discussion of pore aperture and ionic radius to support this thought. It is likely that Na+ would fit but the authors didn't try and the review article is assuming an issue and a solution to an issue that doesn't exist. This should be cleared up otherwise it reads like a fabricated problem and solution.

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We thank the reviewer for pointing this to our attention. We have changed title to "MOF Design and Functionalization Strategies for Advanced Electrochemical Energy Storage Devices."

2) The authors may briefly mention MOF-derived materials (at least a paragraph with some literature references) even if not giving a specific section.

We thank the reviewer for this suggestion and have added a brief description of MOF-derived materials in the introduction part.

"Moreover, the plethora of multidisciplinary characterization techniques applicable to MOFs allow a multitude of experiments to illuminate underlying determinants of performance and provide key insight for development of next-generation materials. The facile tunability of MOFs also enables their utilization as precursors and templates in creating functional materials with desired chemical composition and unique morphologies. MOF-derived materials, including metallic compounds, porous carbons, and their nanocomposites are being widely investigated.[1] Tuning MOF precursor composition and manipulating conversion processes are two main strategies for chemical and structural control. MOF derivatives have shown great potential in electrocatalysis and energy storage devices.[2,3]"

3) Some recent comprehensive reviews on this topic may benefit readers' understanding of this field (for example, Chem, 2, 52-80 (2017); Joule, 2, 2235-2259 (2018)).

We thank the reviewer for this suggestion and have added some recent reviews, including the suggested ones.

4) Better to give more detailed discussions on the limit/drawbacks/bottle necks and possible solutions regarding the use of MOFs for energy storage.

The concluding section for each strategy headlined "design criteria and opportunities" succinctly lists areas for improvement and possible solutions using MOFs. We have further expanded several of these sections to better define the drawbacks/bottleneck of currently reported MOF-based devices.

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We thank the reviewer for this suggestion and have amended the sentence in question.

"Batteries and supercapacitors are among the most promising technologies for electrical energy storage owing to their portability and compact size for on-demand usage. Despite their promise, chemical and physical limitations of existing materials hinder performance and require new, creative solutions. For instance, polymers and conductive carbon materials are relatively inexpensive, scalable, and synthetically tunable but can lack physical and chemical stability for device implementation…"

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The reviewer brings up an excellent point. We have removed the word "crystalline" from the description.

"MOFs are a class of porous ~~crystalline~~ materials composed…"

3) I think it would be worth stating in the intro why porosity is important. This should come in early on. Otherwise, the field of coordination polymers fits all the same criterion without needing porosity. I wouldn't attack coordination polymers in the paper, but simply state the need for porosity that other organic-inorganic hybrid materials could not support.

This is a great suggestion. We have highlighted the importance of porosity early on in the introduction.

"With creative synthetic design, properties such as porosity, stability, particle morphology, and conductivity can be tailored for specific applications. As the needs of each energy storage device are different, this synthetic versatility of MOFs provides a method to optimize materials properties to combat inherent electrochemical limitations. Porosity, a defining characteristic of MOFs, is also highly important for guest/ion storage and transport."

4) In section 2.3, the authors discuss M2dsbdc and M2dobdc. I think the authors have over-simplified the data here. If M-S bonds are so much better, then why is the M-O only 10x worse than the Fe-S while the Fe-S is a million-fold better than the Mn-S. There seems to be more going on than the importance of soft-soft interactions. I think it is worth exploring or clarifying.

We thank the reviewer for identifying this oversimplification. We have revised the section to reflect contributions from the metal and linker identities ultimately determine the electronic conductivity of the M_2(debdc) series.

"To overcome this challenge, significant research efforts are aimed at tailoring the electronic structure via both metal and linker contributions using an approach termed "through-bond" conductivity. For instance, utilizing the loosely bound d-electrons in Fe^{2+} and soft sulfur linkers led to a million-fold enhancement in electron conductivity compared to the analogous Mn-S bonds in M_2(dsbdc) MOFs and ten times improvement compared to metal-oxygen linkages in M_2(dobdc).[14]"

5) It is difficult to say due to the reference style having "Et. al" in it. However, I would like to ensure that proton conductivity includes references from Shimizu who has put a lot of work into this field.

We have included recent work from Shimizu in Reference 22.

6) Under Porosity for tuning ionic conductivity, the authors state "likely due to the insufficient pore size to accommodate Na+ transport. to address this issue ..." I struggle with this. There is no data to support these statements. It's not clear from the writing that the cited references tried Na+ and it failed. There is no discussion of pore aperture and ionic radius to support this thought. It is likely that Na+ would fit but the authors didn't try and the review article is assuming an issue and a solution to an issue that doesn't exist. This should be cleared up otherwise it reads like a fabricated problem and solution.

We thank the reviewer for this comment and have modified our discussion to better reflect the claim that larger pore geometries are better suited to accommodate the large ionic radius of Na^+

"**Porosity for Tuning Ionic Conductivity.** The long-range order and porosity of MOFs can play a unique function in modulating ion transport. In SIBs, the large ionic radius of Na^+ leads to slow diffusion compared to Li^+, making it critical to have well-designed anode materials with available ion transport pathways.[46,47] Of the MOFs studied for LIB anodes, most frameworks utilize relatively short linkers such as 1,4-benzenedicarboxylate acid (bdc), fumaric acid, and formic acid that yield MOFs with small pore sizes.[48–50] To improve the sluggish diffusion of Na^+, the common bdc linker was extended using 4,4'-biphenlydicarboxylic acid (bpdc) to form an isoreticular Co-based MOF.[51] The authors showed that the longer linker results in a MOF with larger pores that is better suited to facilitate Na^+ diffusion. In SIB devices, this MOF anode exhibited stable capacities of ~200 mAh g^{-1} over 1,000 cycles at a charge rate of

100 mA g^{-1}. Isoreticular MOFs and structurally flexible frameworks thus presents a rare opportunity to correlate porosity and chemical functionality with metal ion transport."

In addition, referee #2 argues that more discussion on the terminologies used in article would be beneficial for the readers. (from Editor's letter)

This is an excellent suggestion. We have defined terminologies related to device performance metrics, which will assist the reader throughout the review.

"The largest challenges in current metal-ion anodes are their low specific capacity and limited rate performance. Specific capacity is defined by the total charge that can be stored in the device per mass of active material with typical units of mAh g^{-1}. The device performance is also measured by rate capability (specific capacity as a function of charge and discharge rates) and capacity retention upon extended cycling (percentage of the maximum specific capacity as a function of cycle number). Most LIBs utilize a graphite anode which relies on the reversible intercalation of Li ions in the layered carbon material."