

Response to Reviewer 3 Comments

Dear Reviewer,

Firstly, thanks for your careful review for my paper. Based on your suggestion, we had revised my paper in detail, and the supplemented and revised parts had been marked in purple and blue in revised manuscript.

Detailed corrections are listed below point by point:

Point 1: Usually, scientific papers are written using the simple past tense, whereas the authors describe all result in the present tense. I suggest to change it.

Response 1: Based on your suggestion, the manuscript had been revised carefully for tense. We had used the simple past tense to describe all results. The modified words were highlighted in red, purple and blue in revised manuscript

Point 2: Line 12-13: If you say general metal oxides remove NiO and use the plural (are regarded as). Otherwise say directly nickel oxide instead of transition metal oxides and use the singular.

Response 2: Your suggestion is right. The incorrect sentence had been corrected in revised manuscript (seeing Line 12-13).

Point 3: Line 14: remove unfortunately.

Response 3: The “unfortunately” had been removed (seeing Line 14).

Point 4: Line 14: remove moreover.

Response 4: “Moreover” had been removed (seeing Line 17).

Point 5: Line 19: replace “take the place” with “replace”.

Response 5: “take the place” had been replaced by “replace” (seeing Line 20).

Point 6: Line 23-24: an increased tendency of what? Reformulate the sentence.

Response 6: Based on your suggestion, this sentence had been reformulated in revised manuscript as “When the Ni doping content was less than 3 wt.%, the contents of anatase and NiO were both increased while the rutile content was decreased.” (seeing Line 23-25).

Point 7: Line 28-33: break the sentence and address the factor one by one. Writing them all together with brackets does not help the reading.

Response 7: Based on your suggestion, the sentence had been revised as “Correspondingly, the obtained electronic conductivity of $4.92 \times 10^{-5} \text{ S}\cdot\text{cm}^{-1}$ was also highest, which was about 1.7 times that of pristine TiO_2 . The optimal electrochemical performance was also obtained.

The initial discharge and charge specific capacity was 576 and 264 mAh·g⁻¹ at a current density of 100 mA·g⁻¹. After 100 cycles, the capacity retention reached 48%, and the coulombic efficiency was about 100%. The average discharge specific capacity was 48 mAh·g⁻¹ at a current density of 1000 mA·g⁻¹. About 65.8% of the initial discharge specific capacity was retained when the current density was recovered to 40 mA·g⁻¹.” (seeing Line 29-39).

Point 8: Line 77: Replace “it was due” with “This result was explained in terms of...”

Response 8: Based on your suggestion, “it was due” had been replaced by “This result was explained in terms of...” (seeing Line 83).

Point 9: Line 91: “...TiO₂ nanowires owned a higher specific capacity than bulk TiO₂ (305 vs 240 mAh/g).

Response 9: Based on your suggestion, the sentence had been revised (seeing Line 100).

Point 10: Line 92: In addition to nanomerization, the inclusion of additional substances such as Fe₂O₃, ...into the TiO₂ structure was also found to be an effective method to improve the electrochemical performance.

Response 10: Based on your suggestion, the sentence had been revised (seeing Line 101-104).

Point 11: Line 115: The brackets are not needed when using “such as”.

Response 11: Based on your suggestion, the brackets had been removed (seeing Line 130).

Point 12: Line 117-120: Please reformulate the sentence with clear logic. For instance, “when doping TiO₂ with Sn and Fe, a higher specific capacity was obtained due to...”

Response 12: Based on your suggestion, the sentence had been reformulated as “When doping TiO₂ with Sn and Fe, a higher specific capacity can be obtained due to the formation of SnO₂ and Fe₂O₃, which have a high theoretical specific capacity, typically 781 mAh·g⁻¹ for SnO₂ and 1005 mAh·g⁻¹ for Fe₂O₃.” (seeing Line 132-134).

Point 13: Line 12: annealing treatment at 500C under open air atmosphere.

Response 13: Based on your suggestion, the sentence had been revised (seeing Line 146).

Point 14: Line 155-158: Please reformulate this sentence with correct English and clear logics.

Response 14: Based on your suggestion, the sentence had been reformulated as “The Ni-doped TiO₂ nanofibers may be endowed with the excellent electrochemical performance due to the synergistic effect between the two, which demonstrate the potential application as a prospective anode for high-performance LIBs. However, the above-mentioned researches also have some disadvantages.” (seeing Line 170-175).

Point 15: Line 159: “The present investigation” sounds like the work produced by the author of this manuscript (the one I am reviewing). Please rephrase such as “Second, the effect of nickel concentration of electrochemical performances and was not investigated. As a matter of fact, it is well known that....”.

Response 15: Based on your suggestion, sentence had been rephrased as “First, most NiO-TiO₂ nanocomposites were prepared by the two-step method, namely nanostructured TiO₂ was firstly prepared, then NiO was synthesized on pre-prepared TiO₂. The separated bilayer structure resulted in the interior of TiO₂ free of NiO, and a portion of TiO₂ surfaces uncovered with NiO. This may led to the synergistic effect greatly weakened. Second, the effect of nickel concentration on electrochemical performances and the evolution in phase constituents was not investigated. As a matter of fact, it is well known that as-prepared TiO₂ usually exists in two forms of anatase and rutile.” (seeing Line 176-185).

Point 16: Line 193: Please quantify the specific amount.

Response 16: Based on your suggestion, the specific amount had been quantified as “1 mL” (seeing Line 216).

Point 17: 2.3. Characterization

Please add more details about the analytical measurements and their conditions. Please read other papers to understand what I mean. For XPS add also the reference against which you performed the measurements.

Authors should also describe the cell configuration for the electrochemical experiments.

Response 17: Based on your suggestion, more details about the analytical measurements and their conditions had been added (seeing Line 231-242). We also added the reference [34] for XPS which we performed the measurements (seeing Line 236).

In addition, we had described the cell configuration in details (seeing Line 220-222) and added Figure 1 to show the cell configuration of the battery.

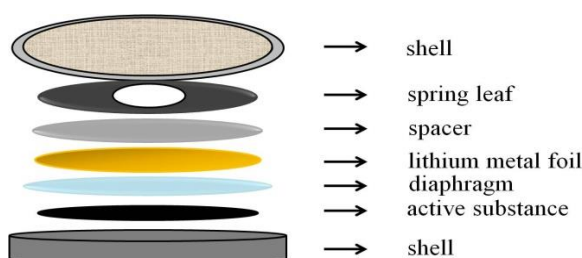


Figure 1. The diagram of the cell configuration

Point 18: Line 259. Please cut the sentence “...doping Ni. This observation can be...”

Response 18: Based on your suggestion, the sentence had been revised (seeing Line 303).

Point 19: Line 261 That “in thermodynamics” is meaningless there.

Response 19: Based on your suggestion, the “in thermodynamics” had been removed. (seeing Line 304).

Point 20: Line 267-268: I suggest to modify as “The content of anatase TiO₂ and rutile TiO₂ in the pristine TiO₂ nanofiber was 11.5 and 88.5%, respectively.”

Response 20: Based on your suggestion, the sentence had been modified (seeing Line 311-312).

Point 21: Line 272-272: The sentence is completely wrong. I suggest the following: “THE Ni atoms INCLUDED in the lattice can cause lattice distortion, thus resulting in stress field”. Also, could the author provide a literature reference for this statement and what follows up to the line 277?

Response 21: Your suggestion is right. The incorrect sentence had been corrected in revised manuscript (seeing Line 317-318). The literature reference for this statement had been provided [55] (seeing Line 318). And the content what followed up to the line 277 had been added (seeing Line 329-338).

Point 22: Line 275: Remove INVOLVED (The content in NiO content in the nanofibers...).

Response 22: Based on your suggestion, the “INVOLVED” had been removed (seeing Line 320).

Point 23: Line 277-279: The sentence is written so badly that is even difficult to understand. Please reformulate.

Response 23: Based on your suggestion, the sentence had been reformulated as “However, when the doping content of Ni exceeded 3 wt%, the content of NiO in the nanofibers presented a downward tendency with the increase in Ni doping content. At the same time, the ratio of anatase to rutile was also reduced. These may be because the formation of NiTiO₃, which would consume NiO and TiO₂.” (seeing Line 329-335).

Point 24: Line 280: I would replace “remained” with “retained”

Response 24: Based on your suggestion, “remained” had been replaced by “retained.” (seeing Line 329).

Point 25: Line 281-282: However, the contrary change in intensity ratio for samples with more than 3 wt.% Ni almost has no relationship with Ni atoms solubilized in TiO₂ due to the solid solution content of Ni reaching the saturation value. This sentence is also very poorly written. Please fix it. I cannot do it without risking to change the meaning.

Response 25: Based on your suggestion, the sentence had been reformulated as “However, when the doping content of Ni exceeded 3 wt%, the content of NiO in the nanofibers presented a downward tendency with the increase in Ni doping content.” (seeing Line 329-331).

Point 26: Line 287-288: Add TiO₂ before rutile and anatase TiO₂(rutile) + ...

Response 26: Based on your suggestion, TiO₂ had been added before rutile and anatase (seeing Line 340-341).

Point 27: Line 284-293: “In order to determine which TiO₂ (anatase TiO₂ or rutile TiO₂) preferentially participates in the formation of NiTiO₃, the changes in standard Gibbs free energy of the following reactions were estimated by thermodynamic calculations.....This provides a strong evidence that the reduction in anatase TiO₂ content is closely related to the formation of NiTiO₃ when more than 3 wt.% Ni is doped”.

The logics here must be improved. The decrease in anatase TiO₂ above 3% Ni was an experimental evidence. The thermodynamic calculations do not provide any direct evidence about it. Thermodynamics can support this finding but are not proof of it at all since (i) both reactions with negative free energy change can happen and (ii) kinetics might be also involved. I suggest to remove the “in order to determine...” (285-285), start from “the free energy change....” And show directly the results of the calculations. Then state that “these results can somehow support the experimental findings about anatase being....

Response 27: Your suggestion is right, “in order to determine...” had been removed (seeing Line 335).

Based on your suggestion, the sentence had been reformulated as “These results can somehow support the experimental finding about anatase being consumed by Reaction (2).” (seeing Line 306-307).

Point 28: Figure 2: Changes in standard Gibbs free energy (ΔG^0) of (a) reaction (2) and (b) reaction (3). The caption written in this way might generate confusion as the word “change” seems to refer to the trend (change of the y variable with the x variable). Instead, I guess that here the word “change” refers to the y variable, as ΔG^0 is the Gibbs free energy change. I would suggest “Gibbs free energy change for reactions (2) and (3). Then also replace directly (a) and (b) in the graph with (2) and (3).

Response 28: Your suggestion is right, the description had been revised (seeing Line 362), and Figure. 3 had been revised too.

Point 29: Line 313-314: Please adjust as “XPS analysis was performed on samples with Ni content set at 3 and 6% (*mentioning it in the body of the text helps the reader*). The narrow and sharp peaks in the XPS spectra from the survey scans confirm that both samples contain Ti, Ni, O, and C.”

Please replace also a1, a2, b1, b2, etc. with a, b, c, d, etc. There is no meaning in having the subscripts and the esthetic result is not even nice.

Response 29: Based on your suggestion, the description of XPS analysis had been adjusted. (seeing Line 369-372), and Figure. 4 had been revised.

Point 30: Line 315-316: “The carbon may derive from the carbon contamination of the surface during the sample preparation”. This sentence leaves space to confusion and misinterpretations and might suggest a not real understanding about how XPS work. That sharp peak is not simple contamination during preparation, like if something we do not know happened. As also stated in the reference [48], the sharp peak of C 1s can be ascribed to the

adventitious carbon-based contaminant from the XPS instrument itself. The meaning is completely different.

Response 30: Your suggestion is right, the sentence had been revised as “the sharp peak of C 1s can be ascribed to the adventitious carbon-based contaminant from the XPS instrument itself or from the sample preparation.” (seeing Line 373-374).

Point 31: Line 318-325: Please add a reference

Response 31: Based on your suggestion, the reference [63] and [48] had been added (seeing Line 379-482).

Point 32: Line 364: All sample PRESENT.

Response 32: Based on your suggestion, the sentence had been revised (seeing Line 430-440).

Point 33: Line 384: “Quite a few” is not an expression to be used in scientific papers.

Response 33: Based on your suggestion, “Quite a few” had been removed (seeing Line 450).

Point 34 : Please replace the caption with subscript indexes in the FE-SEM results too.

Response 34: Based on your suggestion, the caption with subscript indexes in the FE-SEM results had been replaced (seeing Figure 6).

Point 35: Line 408: Comparatively speaking, Ni is further enriched in the samples with an increase in Ni doping content.

It is not Ni to be enriched in the samples. It's the samples who are enriched in Ni. Please adjust it.

Response 35: Your suggestion is right, the sentence had been revised as “Comparatively speaking, the sample who were enriched in Ni showed more nickel” (seeing Line 477).

Point 36: Line 424-425: For the pristine TiO₂ nanofibers, a pair of characteristic redox peaks at 2.24 and 1.6 V correspond to extraction and insertion of Li⁺ from anatase TiO₂.

Where does that Li come from? Suddenly the authors mention the Li insertion without having described the cell configuration, the electrochemical set-up, and so on. Please add all required information.

Response 36: Based on your suggestion, the detailed description about the process of Li⁺ insertion and deintercalation had been supplemented (seeing Line 501-506), which were also supported by the cell configuration (seeing Figure 1).

Point 37: Line 428: A sentence cannot start with “in which”... Please replace with “where”.

Response 37: Based on your suggestion, “in which” had been replaced by “Where” (seeing Line 509).

Point 38: Line 439: “More twice than the...” Maybe the authors mean “More than twice than...”

Response 38: Based on your suggestion, the sentence had been revised as “The discharge capacity of NiO was more than twice than that of TiO₂” (seeing Line 521).

Point 39: Please replace the figure caption with subscripts in the electrochemical results too.

Response 39: Based on your suggestion, the figure caption with subscripts in the electrochemical results had been replaced (seeing Figure 10).

Point 40: Line 519: This is due...

Response 40: Based on your suggestion, the sentence had been revised (seeing Line 610).

Point 41: Line 521-525: This is very poorly written. It is wrong in grammar and contains so many redundant information Please fix it. I suggest the following: The first change is associated with the largest lattice distortion of TiO₂, resulting in the inhibition of transformation from anatase to rutile. The second one is related to the large number of uniformly distributed NiO particles retarding the atomic diffusion and recombination [55], which in turn reduce the nucleation and growth rates of anatase, and intensely suppress its transformation to rutile.

Response 41: Your suggestion is right, the sentence had been revised (seeing Line 613-619).

Point 42: Line 627: replace “acquired” with “observed”.

Response 42: Based on your suggestion, “acquired” had been replaced by “observed” (seeing Line 626).

Point 43: Please revise also the conclusions. The English level is as low as the whole paper. Besides, conclusions is not a summary or a repetition of the abstract. Conclusions must include some comment about the work, future development, significance etc.

Response 43: Based on your suggestion, the conclusions had been re-written, in which some comments about the work, future development and significance had been supplemented (seeing Line 703).

Yours sincerely
Jun Li