Supporting Information for "Damage-free removal of residual carbon in a dielectric barrier discharge (DBD) plasma for carbothermal-synthesized materials"

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1. Removal of residual carbon from carbothermal synthesized SnSb alloy anode materials by DBD plasma

Carbothermal method is widely used to produce iron and various non-ferrous metals in industrially. Herein, carbothermal-synthesized SnSb alloy anode materials application for lithium-ion batteries was selected as an example to demonstrate the generality of DBD plasma decarburization technique to be functional for metals.

Experiments: SnSb alloy anode materials were synthesized from tin and antimony oxides by carbothermal reduction method. Typical, raw

materials of SnO_2 (99.5%, Aladdin), Sb_2O_3 (99.5%, Aladdin), and graphite powder (99%, Aladdin) with a molar ratio of Sn: Sb: C = 0.95 : 0.05 : 3 were first weighed and thoroughly ground in an agate mortar. Then the mixture was heated in a horizontal tube furnace at 1550 ℃ for 6 h under a flow of Ar_2 at 400 mL/min. After cooling down to room temperature, the carbothermal-synthesized SnSb alloy was placed within DBD generator to remove the residual carbon. The average voltage and input power of the high voltage generator were tuned to be 10 kV and 200 W during carbon removal. The duration of the plasma treatment was 50 min. For a comparison, the residual carbon for carbothermal-synthesized SnSb alloy was also removed by traditional high-temperature oxidation in air by exposing the materials at 750 ℃ for about 50 min. X-ray power diffraction (XRD) technology was employed to analyze the removal of residual carbon and the damage effect.

Results and discussion: Figure S1 showed the XRD diffraction patterns for the initial carbothermal-synthesized SnSb alloy sample, DBD plasma treatment sample, and 750 ℃ high-temperature oxidation treatment sample, respectively. It was found that the carbothermal-synthesized SnSb alloy sample maintains the crystalline structure of Sn metal. In addition, obvious diffraction peak belongs to residual carbon was indexed for SnSb alloy sample, indicating the excess addition of carbon reductant. For the DBD plasma treated sample, the

diffraction peak intensity of residual carbon decreased dramatically while that of the SnSb alloy had no obvious change, as compared to the initial carbothermal-synthesized sample. This revealed that the residual carbon existing in carbothermal-synthesized SnSb alloy has been removed effectively. With regard to the high-temperature oxidation treated sample, we found that the diffraction peaks of SnSb alloy nearly quenched completely. SnO_2 crystal was indexed as the dominant phase for the high-temperature oxidation treated sample. This observation about the phase component change from SnSb alloy to SnO_2 verified the seriously oxidation damage effect of the traditional high-temperature oxidation route. In other words, DBD plasma decarburization technique showed superiority over the traditional high-temperature decarburization route for carbothermal-synthesized metals.

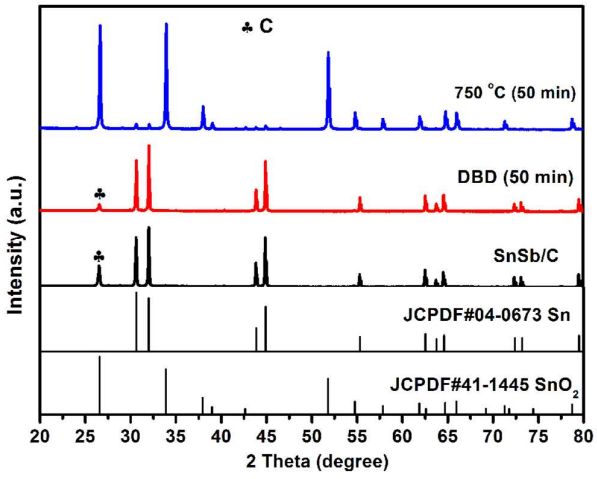

Figure S1 XRD patterns for the initial carbothermal-synthesized SnSb alloy sample, DBD plasma treatment sample, and 750 ℃ high-temperature oxidation treatment sample, respectively.

2. Removal of residual carbon from carbothermal synthesized TiN ceramic powder by DBD plasma

Non-oxides ceramics, such as Si_3N_4, AlN, TiN, SiC, have got much attentions and available in many fields in the past years. TiN ceramic powder was synthesized by carbothermal reduction method and the residual carbon was removed by DBD plasma treatment in this section to show the generality of DBD plasma decarburization technique application for carbothremal-synthesized non-oxides ceramic powders.

Experiments: TiN ceramic powder was synthesized from TiO_2 by carbothermal reduction method. Typical, raw materials of TiO_2 (99.5%, Aladdin) and graphite powder (99%, Aladdin) with a molar ratio of Ti: C = 1: 3 were first weighed and thoroughly ground in an agate mortar. Then the mixture was heated in a horizontal tube furnace at 1550 ℃ for 6 h under a flow of N_2 at 400 mL/min. After cooling down to room temperature, the carbothermal-synthesized TiN ceramic powder was placed within DBD generator to remove the residual carbon. The average voltage and input power of the high voltage generator were tuned to be 10 kV and 300 W during carbon removal. The duration of the plasma treatment was 50 min. For a comparison, the residual carbon for carbothermal-synthesized TiN ceramic powder was also removed by traditional high-temperature oxidation in air by exposing the materials at 750 ℃ for about 50 min. X-ray power diffraction (XRD) technology

was employed to analyze the removal of residual carbon and the damage effect.

Results and discussion: Figure S2 showed the XRD diffraction patterns for the initial carbothermal-synthesized TiN sample, DBD plasma treatment sample, and 750 ℃ high-temperature oxidation treatment sample respectively. Mixture phases of TiN crystalline and residual carbon were indexed for the initial carbothermal-synthesized TiN sample. After high-temperature oxidation treatment, TiN was found to be oxidized into TiO_2 completely accompanying with the removal of residual carbon. The phase transformation from TiN to TiO_2 was ascribed to the serious oxidation damage of high temperature oxidation route. As for the DBD plasma treated sample, the diffraction intensity of residual carbon decreased obviously while that of TiN phase had no obvious change, as compared to the initial TiN/C sample. This implied that the residual carbon for carbothermal-synthesized TiN ceramic powder could be removed effectively. Except to the dominant TiN phase, trace amount of TiO_2 phase was also detected for the DBD plasma treated sample, indicating the occurrence of oxidation damage induced by DBD treatment. By comparing with the XRD profiles, we could find that the damage effect induced by DBD treatment is much lower than that induced by high temperature treatment. These results evidenced that the residual carbon for carbothermal-synthesized TiN ceramic powder could be removed

effectively by DBD plasma treatment with lower damage.

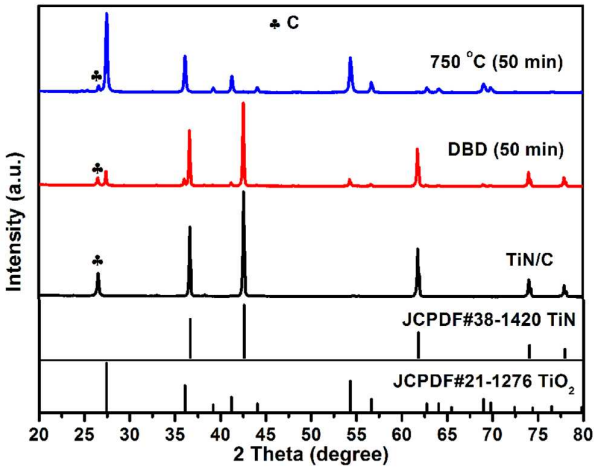

Figure S2 XRD patterns for the initial carbothermal-synthesized TiN ceramic powder sample, DBD plasma treatment sample, and 750 ℃ high-temperature oxidation treatment sample, respectively.