

Use of Tantalum Oxynitrides as Carbon-free Cathodes for Li-O₂ Batteries



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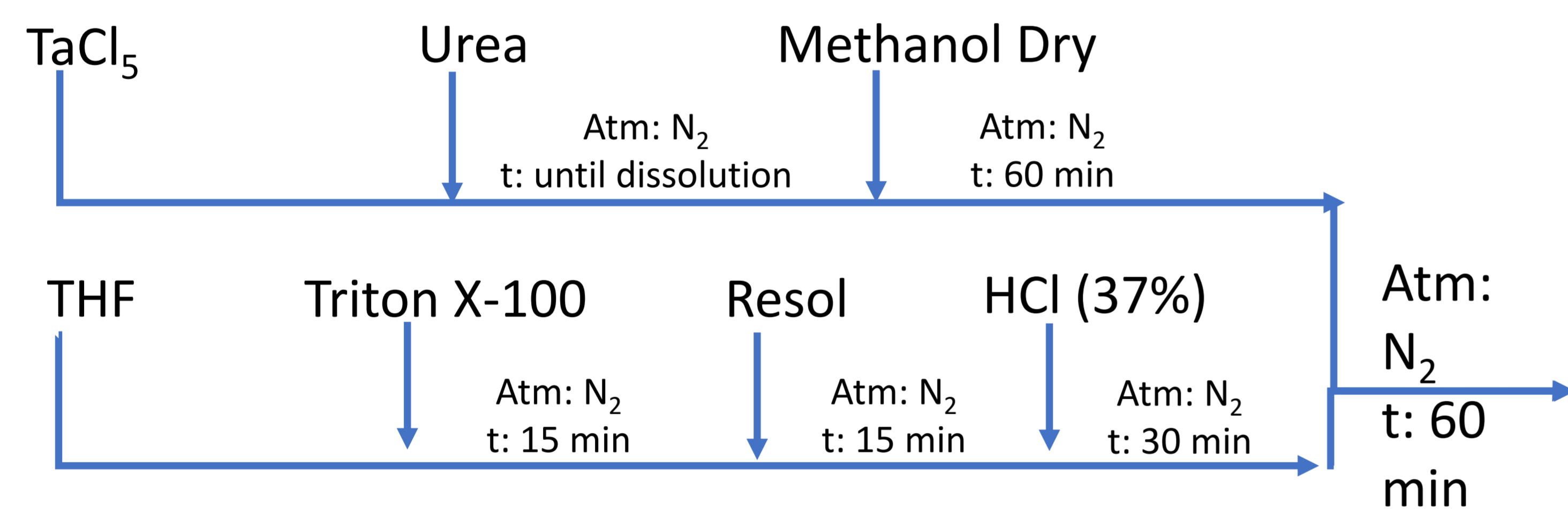
Abstract

Rechargeable batteries based on Li-O₂ with carbon-free cathodes have proven the potential of increasing meaningfully the energy density of batteries and allowing a useful operational life compared to other technologies now implemented. Carbon-free cathodes can be based on several transition metal oxynitrides.

Tantalum oxynitrides were synthesized by sol-gel using tantalum chlorates with the addition of urea, methanol and organics components. The needed porosity was obtained through the application of a thermal treatment in order to eliminate the organic components and the separation of the hydrophobic and the hydrophilic phase. The final product of this process was applied to nickel foam in order to allow a better electric conductivity and a better reactivity of the O₂ in the Oxygen Reduction Reaction (ORR) and Oxygen Evolution Reaction (OER).

The characterization of the sample involved the evaluation of the porosity, with the analysis of the surface area and pore size through the BET technique, Energy-dispersive X-ray spectroscopy (EDS), scanning electron microscopy and ORR/OER evaluation.

Synthesis of Tantalum Oxynitrides



Methodology

Tantalum oxynitrides were synthesized with the molar ratio shown in the table below, apart from Resol which was used at a ratio (1:0.4) to tantalum chloride.

| Reactive | Urea | Metanol Dry | TaCl5 | THF | Triton x100 | Resol | HCl (c) |
|-------------|-------|-------------|-------|-------|-------------|-------|---------|
| Mass | 2,09 | 9,48 | 2,88 | 35,56 | 1,152 | ----- | ----- |
| Volume | ----- | 12 | ----- | 40 | 1,07 | ----- | ----- |
| Molar ratio | 4 | 37 | 1 | 61 | 0,23 | ----- | 4,5 |

Once the synthesis was completed, the final product was left for 74 hours in an oven at 50°C until the product was sufficiently viscous to be applied to the nickel foam. Once the product is applied to the strips, they are treated at 250°C in vacuum for 6 hours. After which it is treated at 450°C in order to ensure the elimination of the organic compounds used in the synthesis in order to create the needed porosity.

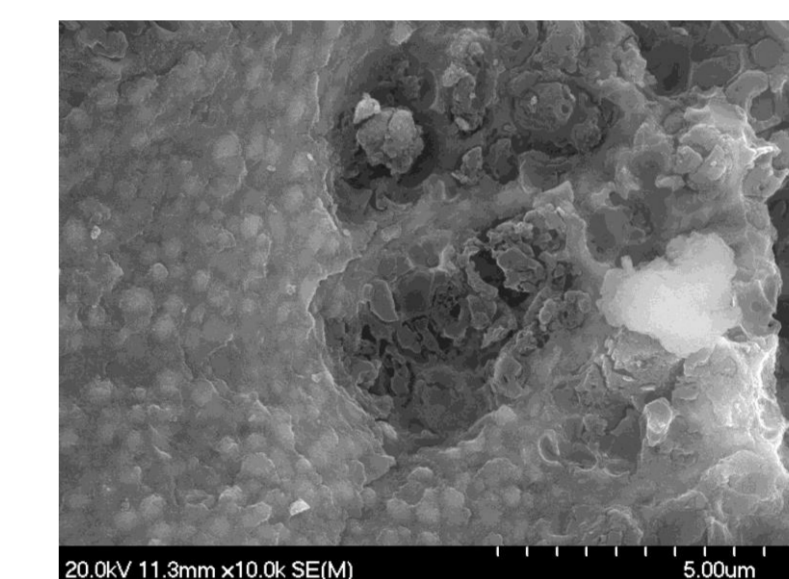
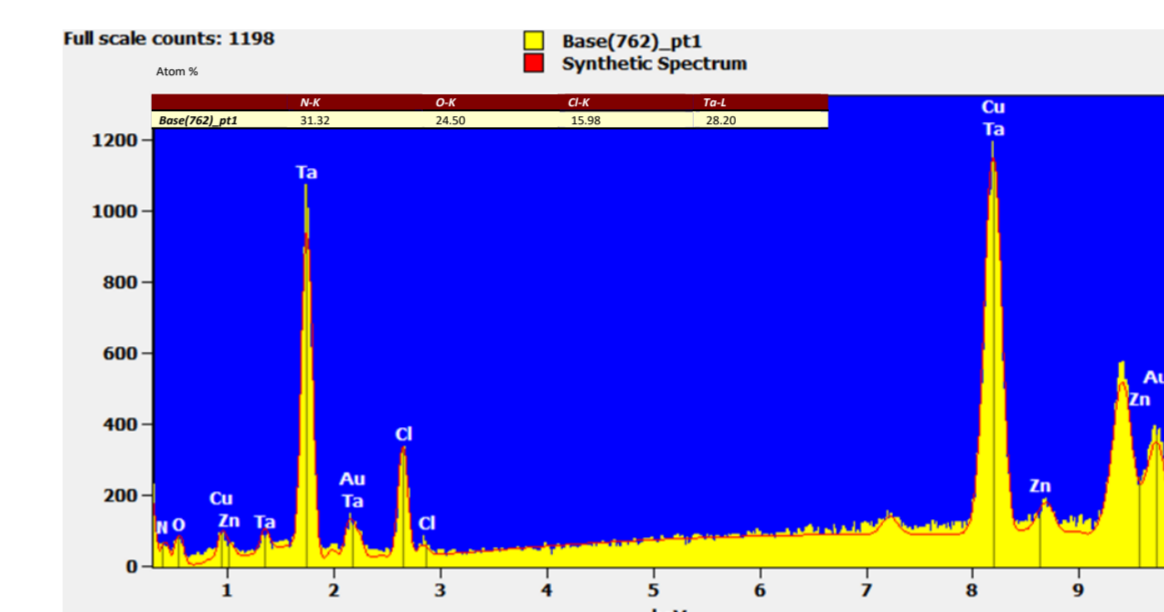
After the heat treatment, the ORR and OER were measured in one of the strips while the others are subjected to heat treatment at 800°C in the presence of a flow of 0.5 l/s of nitrogen. After which, the ORR and OER are measured again.

Finally, powders from the gel are obtained after a thermal treatment at 250°C in vacuum and 450°C in air. A subsequent grinding for the analysis of the surface area through the BET technique was performed.

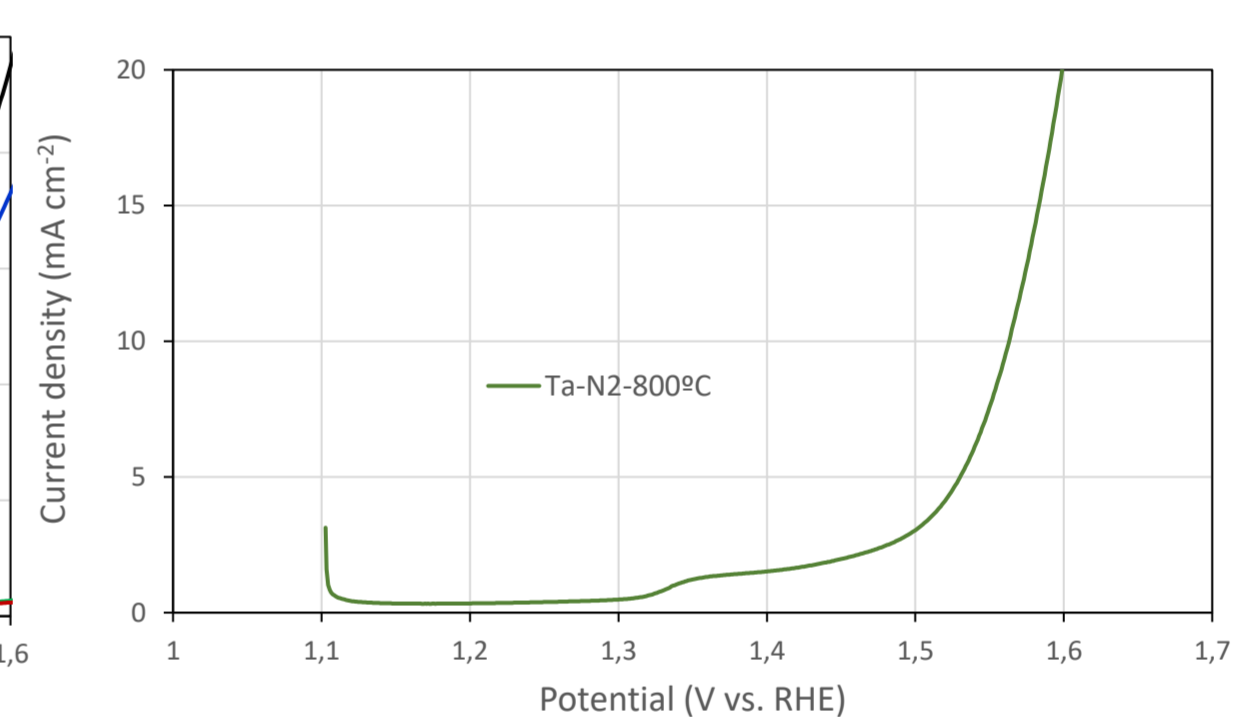
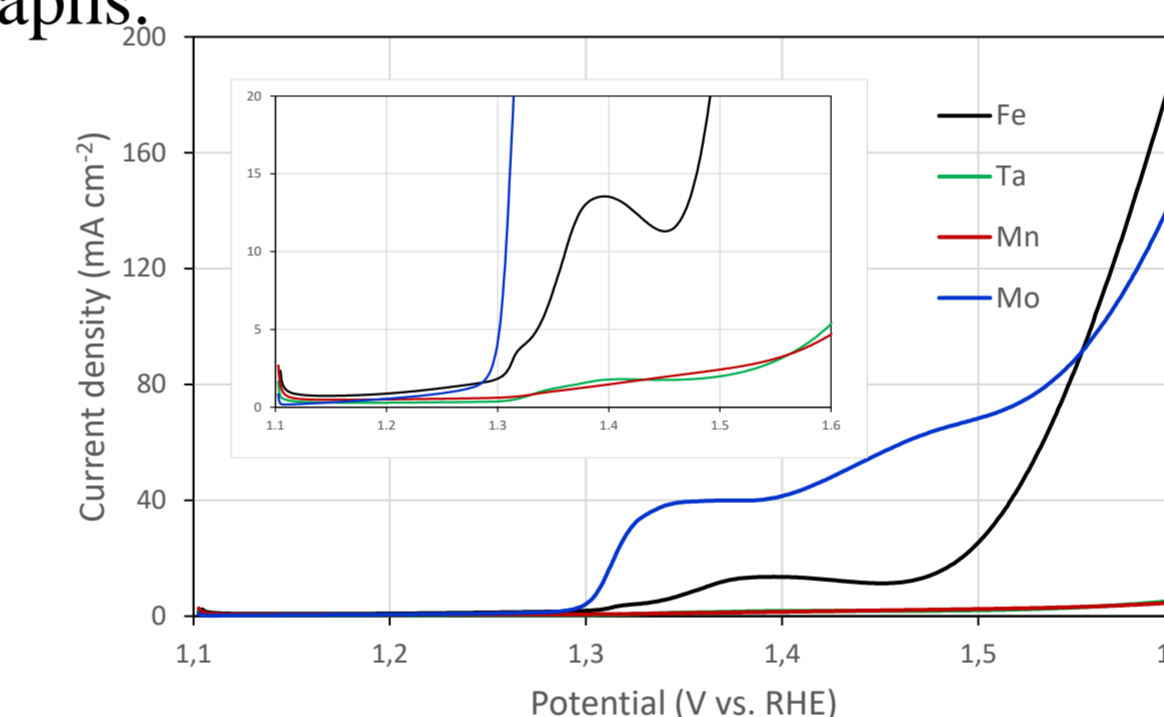
Results

It has been obtained a viscous final material capable of penetrating into the strips of nickel foam leading to a surface area of 380 square meters per gram after the thermal treatment at 450°C. This treatment originates the elimination of organic components producing a porous structure. Only with the thermal treatment at 250°C, most of the organic compounds remain in the gel, as we can see in the SEM image where a phase separation can be observed. he scanning electron microscopy it can be seen how they continue without having been degraded part of the organic compounds and how the porosity of the compound is still not high.

The qualitative analysis Energy-dispersive X-ray spectroscopy indicates the presence of Ta, N, O and Cl (probably from HCl). Others elements (Cu, Zn, Au) are from the substrate used for the SEM evaluation and gold from used to coat the sample.



A 5-fold increase in current density between the sample after treatment in nitrogen (left) and the sample after treatment at 800°C (right) can be seen in the following graphs.



It has been proven that after the application of tantalum oxynitride after treatment at 450°C, the Nickel strips have a better conductivity than the untreated Nickel strips.

A similar our superior behavior has also been observed in other metals different from tantalum such as molybdenum, iron, manganese, titanium and zirconium with ORR and OER results.

Conclusion

1. Tantalum oxynitrides have been synthesized with the necessary properties to act as an effective cathode without the presence of carbon compounds.
2. The elimination of organic compounds through heat treatment has resulted in high porosity.
3. Heat treatment at 800°C with a nitrogen flow increases the current density.

References

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