

Preparation from the solvothermal aqueous alkoxide route of multifunctional, highly-optimized metal-oxide nanoparticles based on LiNbO₃ or LiTaO₃

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Non-centrosymmetric metal oxide nanoparticles are known to exhibit functional properties that are similar to their bulk counterpart when their size is typically above 20 nm. Among them, lithium niobate (LiNbO₃) nanocrystals have for instance gained significant attention as promising candidates for applications in biomedical imaging, nonlinear optics, and sensing owing to their high nonlinear optical coefficients [1,2]. Our research focus now extends towards the development of multimodal agents capable of generating diverse contrasts through various physical mechanisms, encompassing MRI [3], computed tomography (CT), photoluminescence, and nonlinear optical processes from the same core nanomaterial.

In this presentation, our deep understanding of the reaction pathways and formation mechanisms of LiNbO₃ nanoparticles will be first addressed [4]. The synthesis is based on the aqueous sol-gel route after simple mixing and hydrolysis of the metal alkoxide precursors. Interestingly, new octanuclear complexes with a {Li₄Nb₄O₁₀} core were identified at room temperature through ESI-MS, NMR, and SAXS experiments before the formation of a colloidal gel which is subsequently heat-treated under solvothermal conditions. An aggregation-mediated crystallization process then results in LiNbO₃ nanocrystals whose mean size can be adjusted in the 20-100 nm range according to the co-solvent used.

We also provide insights that this sol-gel route can be easily extended to the preparation of highly-doped LiNbO₃ nanocrystals with, for example, optimal concentrations in lanthanide ions such as Er and Yb [5]. The simultaneous emission of second harmonic and up-conversion signals is then easily assessed under femtosecond excitation and from colloidal suspensions.

Finally, synthesis of other metal oxides, including lithium tantalate (LiTaO₃), is investigated since substitution of niobium with tantalum ions may result in increasing the CT contrast. The K-edge at 67.4 keV of Ta indeed fits with contemporary CT scanners. All together, these results indicate that the aqueous alkoxide route here developed under solvothermal conditions could also be appropriate for the preparation of other multifunctional binary, and even ternary, oxide nanocrystals.

References

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