Combinatorial Reactive Sputtering with Auger Parameter Analysis enables Synthesis of Wurtzite Zn_2TaN_3

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The discovery of new functional materials is one of the key challenges in materials science [1]. Combinatorial high-throughput approaches using reactive sputtering are commonly employed to screen unexplored phase spaces [2]. During reactive combinatorial deposition the process conditions are rarely optimized, which can lead to poor crystallinity of the thin films. In addition, sputtering at shallow deposition angles can lead to off-axis preferential orientation of the grains. This can make the results from a conventional structural phase screening ambiguous. Here we perform a combinatorial screening of the Zn-Ta-N phase space with the aim to synthesize the novel semiconductor Zn_2TaN_3. While the results of the XRD phase screening are inconclusive, including Auger parameter analysis in our workflow allows us to see a very clear discontinuity in the evolution of the Ta binding environment [3,4]. This is indicative of the formation of a new ternary phase. In additional experiments, we isolate the material and perform a detailed characterization confirming the formation of single-phase wurtzite Zn_2TaN_3. Besides the formation of the new ternary nitride, we map the functional properties of $Zn_xTa_{1-x}N$ and report previously unreported clean chemical state analysis for Zn_3N_2, TaN and Zn_2TaN_3 [4]. Overall, the results of this study showcase common challenges in high-throughput materials screening and highlight the merit of employing characterization techniques sensitive towards changes in the materials' short-range order and chemical state.

[1] W. Sun, C. J. Bartel, E. Arca, S. R. Bauers, B. Matthews, B. Orvañanos, B.-R. Chen, M. F. Toney, L. T. Schelhas, W. Tumas, J. Tate, A. Zakutayev, S. Lany, A. M. Holder, G. Ceder, Nat. Mater. 18 (2019) 732–739.

[2] S. Zhuk, A. Kistanov, S. Boehme, N. Ott, F. La Mattina, M. Stiefel, M. V. Kovalenko, S. Siol, Chem. Mater. 33 (2021) 9306–9316.

[3] S. Zhuk, S. Siol, Appl. Surf. Sci. 601 (2022) 154172.

[4] S. Zhuk, A. Wieczorek, A. Sharma, J. Patidar, K. Thorwarth, J. Michler, S. Siol, Chem. Mater. 35 (2023) 7069–7078.