Formation of Mo-Ta Solid-Solution Nitrides under Extreme Conditions

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Abstract

Many transition metal (TM) nitrides are refractory ceramics. They are typically characterized by high melting temperature, oxidation resistance, chemical inertness, high electrical conductivity and remarkable catalytic properties. The binary nitrides based on either Ta or Mo have some remarkable properties, beyond applications just exploiting their high melting temperatures and oxidation resistance. For example, TaN exhibits exceptionally high thermal conductivity and therefore is widely used in the semiconductor industry (as a critical diffusion barrier and adhesion layer in copper interconnects), while Mo_2N is increasingly recognized for its catalytic properties.

It is interesting to study Mo-Ta-N ternary nitride compounds, as broader structure-property tunability is expected. Within this concept, either solid solutions of known nitrides or novel ternary phases can be of interest for fundamental and applied sciences. However synthesis of nitride compounds remains a challenging task, as typically high pressure conditions are required.

In this project, we explore reactions of Mo-Ta alloy with nitrogen and the resulting phases within the ternary Mo-Ta-N system, in the range of pressures and temperatures up to 50 GPa and 2000-3000 K. We employed laser heated diamond anvil cell (LH-DAC) was to achieve high pressure high temperature conditions, and synchrotron radiation of PETRA III, DESY allowed to study the crystalline products of the reaction.

An example of a diamond anvil cell (DAC) is shown in Figure 1. It is a Sym70 cell equipped with Boehler-Almax diamonds (300 μm diameter). The DAC was loaded with MoTa alloy (1:1 composition), ruby chips and gold powder that served as pressure markers, and filled with N_2 gas. We performed laser heating at the pressure 47 GPa by slowly raising temperatures up to 2600(200) K. The simultaneous collection of diffraction patterns showed the formation of new crystalline phases during heating. After temperature quenching, the reaction products were studied by X-ray diffraction imaging, which allowed to identify positions of the sample suitable for multigrain XRD data collection using ω-scans with Δω = 0.5°. Within our study we solved and refined crystal structures of several previously unknown nitride phases, including a phase crystallizing in the orthorhombic space group Pnma with a unit cell volume V~174 Å^3 as well as other phases (their crystal structures are shown in the Figure 1).

The most important observation is the presence of occupational disorder on the crystallographic metal sites, including both Mo and Ta. The new phases feature high incompressibility, and remain stable at the ambient conditions after pressure release.

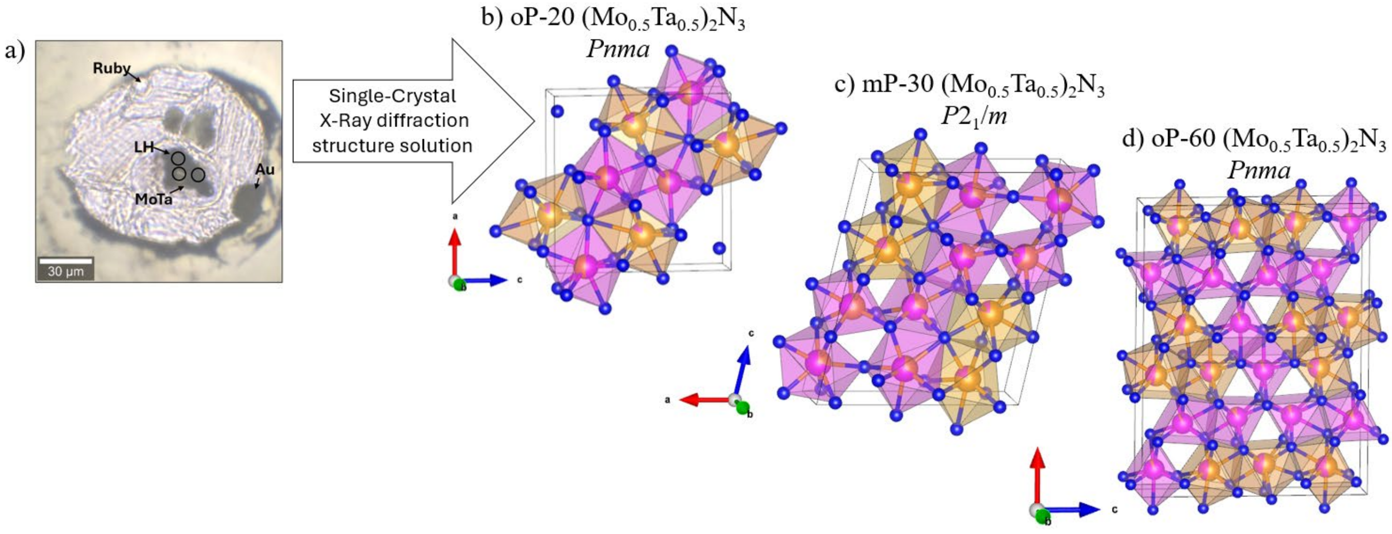

Fig 1. a) Photo of DAC chamber with laser-heated MoTa alloy. b) oP20-MoTaN3 Pnma phase. c) mP30-MoTaN3 phase. d) oP-MoTaN3 phase. Colors: orange – Ta, pink – Mo, blue – N; orange and pink polyhedra are bicapped and capped prisms with different volumes around 13 Å^3 and 16 Å^3 respectively.