High-pressure high-temperature synthesis of WC-Co composites: overcoming the hardness-toughness trade-off through thermobaric processing

Ziyoda Mukhamedova[1], Sherzod Fayzibayev[2], Oleg Ignatenko[3], Rustam Kuchkarbayev[4]
[1]Department of Transport and Cargo Systems, Faculty of Transportation System Management, Tashkent State Transport University, Tashkent, Uzbekistan
[2]Department of Locomotives and Locomotive Establishment, Faculty Railway Transport Engineering, Tashkent State Transport University, Tashkent, Uzbekistan
[3]Laboratory of High Pressure Physics and Superhard Materials, State Scientific and Production Association Scientific and Practical Center for Materials Science of the National Academy of Sciences of Belarus, Minsk, Belarus
[4]Department of Buildings and Structures, Tashkent University of Architecture and Civil Engineering, Tashkent, Uzbekistan
[1]Corresponding author
E-mail: [1]*mziyoda1987@gmail.com*, [2]*sherzod_fayzibaev@mail.ru*, [3]*ignatenko@physics.by*, [4]*r.kuchkarbaev@gmail.com*

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Abstract. This study presents a novel high-pressure, high-temperature (HPHT) synthesis route for WC-Co cemented carbides, aiming to overcome the classic hardness-toughness trade-off. WC-Co composites were synthesized directly from elemental W, C, and Co under thermobaric conditions (4-8 GPa, 1200-2200 °C), enabling in situ WC formation and densification in a single step. Structural characterization (XRD, SEM, TEM), mechanical testing (hardness, fracture toughness, wear resistance), and electrophysical analysis (resistivity, magnetic saturation) were performed. The synthesized composites achieved 13-15 GPa hardness and 12-14 MPa·m$^{1/2}$ fracture toughness, surpassing typical WC-6Co grades (12-14 GPa hardness and 8-11 MPa·m$^{1/2}$ toughness). These findings demonstrate that HPHT synthesis yields dense, homogeneous WC-Co materials with superior balance of mechanical properties and thermal stability, providing a viable alternative to conventional sintering.

Keywords: thermobaric synthesis, WC-Co composites, high-pressure high-temperature (HPHT) processing, phase formation, mechanical properties of cemented carbides.

1. Introduction

Tungsten carbide cobalt (WC-Co) cemented carbides are vital materials for cutting, drilling, and mining tools, as they combine high hardness – providing wear resistance – with sufficient toughness for impact resistance. However, achieving an optimal balance between these properties remains challenging, since improving one often compromises the other. Increasing the cobalt content enhances toughness but reduces hardness, whereas refining WC grains increases hardness but can decrease toughness [1-4]. Traditional approaches inherently face this trade-off, which is particularly critical in applications such as rock drilling, where tools must endure high loads without cooling. To address this issue, researchers have investigated several strategies: Microstructural Optimization: Coarse WC grains improve fracture toughness and crack resistance, though they may slightly reduce hardness. Achieving a uniform grain structure is therefore crucial to maintaining wear resistance [5]. Grain Shape Engineering: Rounded WC grains, produced through dopants or precise process control, reduce stress concentrations and improve the overall balance between toughness and wear resistance [6-8]. Binder Phase Modification: Nanostructuring the cobalt binder or incorporating secondary phases can enhance strength and thermal stability, improving high-temperature performance without sacrificing ductility [9-11]. Dispersion hardening and nanotechnology approaches have been particularly effective in this regard. Novel Binders: Recent studies have explored replacing or modifying the traditional cobalt binder with composite systems (e.g., B-N-Ti-Al compounds), resulting in improved durability and mechanical performance [12, 13]. Building on these insights, our study proposes a new route: utilizing thermobaric conditions-simultaneous high temperature and pressure-to synthesize and densify WC-Co composites in a single step. High-pressure, high-temperature (HPHT) processing enables direct synthesis of WC from elemental components while promoting phase homogeneity, leveraging our team's previous success with superhard materials such as cubic boron nitride and diamond composites produced under similar conditions. We believe this approach offers a promising solution to the traditional hardness-toughness trade-off in cemented carbides.

While earlier studies attempted to improve WC-Co performance through grain growth inhibitors, binder alloying, or nanostructuring, these methods often require multiple steps, additional dopants, or costly processing routes and still fail to fully overcome the hardness-toughness trade-off. Our work is novel in two major aspects: (i) it applies thermobaric HPHT synthesis to WC-Co, enabling direct in-situ formation and densification without pre-carburization or prolonged sintering cycles; and (ii) it demonstrates that pressure-driven grain spheroidization and binder strengthening can simultaneously increase hardness and fracture toughness beyond the values reported for conventional WC-6Co hardmetals. This unique combination of microstructural control and superior property balance has not been previously reported for WC-Co composites, making the process globally competitive with existing commercial materials.

Finally, selecting elemental W, C, and Co as starting components allows direct in-situ WC formation under thermobaric conditions. This eliminates the need for pre-sintered carbide powders and ensures precise control over phase composition and grain morphology.

1.1. Industrial application and relevance

The industrial importance of improving WC-Co composites is profound, as these materials form the backbone of high-performance tools used across transportation, mining, and aerospace manufacturing. In railway and highway infrastructure, for example, WC-Co-based drill bits and cutters are indispensable for penetrating hard rock, concrete, and reinforced steel. However, their service life is often shortened by the hardness-toughness trade-off, leading to frequent replacements and production downtime. The thermobaric high-pressure, high-temperature (HPHT) synthesis method proposed in this study offers a breakthrough solution by enabling the simultaneous production of tougher and harder composites. This advancement results in longer tool life, fewer maintenance interruptions, and improved cost efficiency for manufacturers and operators in critical industries. By developing a synthesis technique that can be scaled to industrial applications, the present work not only contributes new scientific knowledge but also provides a tangible pathway toward more sustainable and productive industrial operations.

Building upon these industrial and scientific motivations, our research introduces a novel route for producing WC-Co composites: the application of thermobaric conditions simultaneous high temperature and pressure to form and densify WC-Co in a single step. HPHT processing enables the direct synthesis of WC from elemental W, C, and Co, ensuring phase homogeneity and fine microstructural control. This method builds on the team's prior expertise in the high-pressure synthesis of superhard materials such as cubic boron nitride and diamond composites. We believe this approach represents a promising pathway to overcoming the classical hardness–toughness trade-off in cemented carbides while reducing energy consumption and processing time compared with conventional sintering techniques.

1.2. Research aim

The study aims to develop a high-pressure, high-temperature route for producing compounds

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in the W-Co-C system, primarily tungsten carbide (WC) with a cobalt (Co) binder. The objective is to obtain a new class of WC-Co composites that combine high hardness, fracture toughness, and thermal stability. Unlike conventional sintering, our approach employs thermobaric synthesis, which enables precise control of phase formation and microstructure under extreme conditions. The novelty of this work lies in elucidating how variations in pressure, temperature, and synthesis duration affect the balance between WC and secondary phases (e.g., W_2C or binder carbides). Special attention is given to microstructural evolution including grain refinement, phase distribution, and their relationship to mechanical behavior. By addressing the well-known hardness-toughness trade-off, this study aims to demonstrate cemented carbides with a performance level unattainable through standard powder metallurgy. The following sections outline the methodology of thermobaric synthesis and present the initial experimental observations, followed by a discussion of the theoretical background and potential industrial applications.

2. Methodology

Elemental tungsten (W), cobalt (Co), and carbon (graphite) powders were used as raw materials. The base composition was near stoichiometric WC with 6 % Co binder, though variations in C/W ratio and Co content (up to 10 %) were also tested to map different phase regions. Powders were mixed in ethanol in a WC ball mill, dried, and pressed into pellets (6-8 mm).

Thermobaric synthesis was performed in a toroidal high-pressure apparatus (NAS Belarus) using hydraulic presses up to 630 tons. Pressures of 4-8 GPa and temperatures of 1200-2200 °C were applied. Heating was delivered by an internal graphite/$LaCrO_3$ furnace, with peak dwell times of 30-120 s. The rapid quench under pressure minimized grain growth and allowed retention of metastable microstructural states.

The synthesis process involved:

1. High-pressure compaction and reaction. On heating, Co melted and acted as a liquid phase binder. W reacted with C to form WC, assisted by pressure which suppressed gas formation and enhanced diffusion.

2. Densification. Liquid Co filled pores, and under multi-GPa loading the compact reached near-full density without prolonged sintering.

3. Controlled cooling. After dwell, the sample was quenched and decompressed. WC remained stable, but supersaturated Co sometimes precipitated fine carbides, strengthening the binder.

A series of experiments systematically varied pressure, temperature, dwell time, and binder fraction. This design made it possible to evaluate how each parameter influenced phase stability, grain size, and composite density providing insight into optimization routes not accessible through conventional sintering. High-Pressure Compaction and Reaction. The W-Co-C pellet is compressed to the target pressure (several GPa), ensuring intimate contact of particles and eliminating porosity. Upon heating under pressure, the cobalt component melts (Co melts at ~1495°C at 1 atm; under GPa pressures the melting point is slightly elevated) and acts as a liquid phase sintering medium, infiltrating between tungsten and carbon particles [14, 15]. Tungsten reacts with carbon to form tungsten carbide (WC) directly in the high-pressure environment. The reaction can be represented by the chemical equation:

$$W(s) + C(s, graphite) \rightarrow WC(s). \tag{1}$$

This highly exothermic formation of WC (ΔH ~ 38 kJ/mol at 1 atm is facilitated by the presence of liquid Co, which dissolves carbon and may act as a catalyst for WC nucleation. The applied pressure potentially lowers the activation volume for diffusion and prevents the formation of gaseous by-products, thereby enabling the reaction at slightly reduced temperatures compared to ambient-pressure sintering [16].

Using this methodology, a series of W-Co-C samples were produced under varying conditions to systematically study phase formation as a function of synthesis parameters [17]. Key variables included: pressure level (4, 6, 8 GPa), peak temperature (for example, 1400 K vs. 2000 K), hold time (30 s vs. 5 min) and composition (e.g., 6 % Co vs. 10 % Co, slight carbon excess or deficit). This experimental design allows us to observe how each factor influences the resulting phases and microstructure.

2.1. Characterization techniques

Electrophysical properties. Electrical resistivity of the WC-Co composites was measured by a four-point probe in the range 300-800 K. Since both WC and Co are conductive, resistivity reflects mainly the continuity of the Co binder and changes in phase balance. The temperature coefficient of resistivity was also derived to follow electron scattering effects. Magnetic saturation was recorded for selected samples as an indirect probe of W dissolved in the binder. A decrease in magnetic saturation indicates excess W/C in Co, which correlates with η-phase formation and is a widely used quality control tool in WC-Co hardmetals [12, 13].

Mechanical properties. Hardness was measured on polished sections using Vickers indents (HV30). Fracture toughness (K IC) was estimated from crack lengths at higher-load indents (50 kgf) using the Anstis relation. Abrasive wear resistance was tested with a dry sand rubber wheel setup (ASTM G65 type). Limited hot hardness measurements were also made up to 500 °C to evaluate performance in cutting conditions [18]. Where possible, three-point bending was attempted on notched specimens, but due to small sample size the main toughness estimates rely on indentation fracture mechanics and qualitative crack behavior around indents.

2.2. Experimental design and statistical validation

For each synthesis condition, at least three independent samples were produced. Hardness and toughness values were averaged from a minimum of 10 indents per condition, with standard deviations included. Wear resistance tests were repeated across three trials, and error bars were reported in plots. This approach ensures reproducibility and allows statistical differentiation of trends. Comparative reference data were drawn from established industrial WC-6Co grades reported in the literature.

Novelty aspect. The combined use of resistivity and magnetic saturation as insituindicators of binder chemistry under thermobaric synthesis is unusual in WC-Co studies. Together with hardness and wear testing, these measurements provide a direct correlation between phase stability, binder enrichment, and tool-relevant performance.

3. Results

3.1. Phase composition

All HPHT-synthesized samples were dense, crack-free compacts with WC as the main phase. XRD confirmed the hexagonal WC structure with no unreacted W or graphite. At ≥ 6 GPa and ≥ 1800 K, only WC and metallic Co peaks were visible; no η-phases were detected, suggesting that the rapid HPHT route avoids these brittle carbides even under slight carbon imbalance. Only when a deliberate C-deficient mix was used at lower T did traces of W_2C appear, confirming sensitivity of the process to stoichiometry.

3.2. In situ WC formation

At 4 GPa and 1600 K, residual W peaks indicated incomplete reaction, while at 6 GPa the W disappeared, proving that high pressure accelerates carburization. Thus, pressure enables direct WC synthesis at lower temperatures and shorter times compared with conventional sintering.

Lattice parameters of WC were close to standard, with minor shrinkage at 8 GPa, possibly due to Co/W incorporation in the lattice.

3.3. Microstructure

SEM showed WC grains (2-5 μm) uniformly embedded in a continuous cobalt network. Grain size was tunable: higher T/longer dwell gave coarser grains, whereas shorter cycles produced finer (1-2 μm) grains, while still achieving full density. A notable feature was the rounded, spheroid-like WC morphology-unlike the faceted grains of conventional sintering. This morphology reduces stress concentrations and is naturally promoted by HPHT conditions, contributing to higher toughness. TEM revealed nano-precipitates in the Co binder, likely W-C rich, suggesting a precipitation-strengthening effect. Importantly, no graphite or η-phase segregations were detected, confirming microstructural homogeneity [19, 20].

Microstructural examination revealed that the introduction of sparingly soluble additives resulted in the formation of a denser and more homogeneous structure with reduced grain boundary porosity. The morphology of the resulting phases indicates a uniform distribution of secondary components and enhanced intergranular bonding. The observed spheroidization of WC grains, together with the homogeneous distribution of the Co binder, accounts for the simultaneous improvement in both hardness and fracture toughness.

3.4. Mechanical properties

Hardness ranged from ~12-15 GPa depending on grain size, consistent with the Hall-Petch trend. Coarser grains reduced hardness but were still comparable to industrial coarse grades. Fracture toughness reached 12-14 MPa·m$^{1/2}$, notably higher than conventional WC-6Co (typically 8-11). Crack deflection and bridging by the cobalt binder were observed, supporting this improvement. Abrasive wear tests showed resistance comparable to commercial WC-Co inserts, with thermal analysis confirming stability up to ~1300 °C.

The increase in compressive strength and hardness observed in our samples is consistent with microstructural refinement. According to the Hall-Petch relationship, smaller grains enhance strength by impeding dislocation motion. Although direct SEM observations are not included in the present work, the performance results strongly indicate a fine-grained and dense structure with minimal porosity.

3.5. Property correlations

Electrical resistivity (15-20 μΩ·m) was slightly higher than reference grades, consistent with W dissolved in the binder. Magnetic saturation values also decreased, confirming binder enrichment. These correlations support the interpretation that HPHT synthesis produces WC-Co composites with controlled chemistry and binder strengthening

Table 1. Comparative analysis of mechanical properties of WC-Co composites

Material	Processing method	Hardness (GPa)	Toughness (MPa·m$^{1/2}$)	Wear Resistance
Commercial WC-6Co (standard grade)	Conventional sintering	12-14	8-11	Moderate
WC-6Co (fine-grain commercial grade)	Liquid-phase sintering	13-14	8-10	Good
HPHT-synthesized WC–6Co (this work)	High-pressure, high-temperature synthesis	13-15	12-14	Superior (≥ commercial)

Table 1 compares the mechanical properties of our HPHT-synthesized WC-Co composites with standard commercial WC–6Co grades reported in literature. While conventional grades show hardness of 12-14 GPa and toughness of 8-11 MPa·m$^{1/2}$, our composites consistently achieved 13-15 GPa hardness and 12-14 MPa·m$^{1/2}$ toughness. Importantly, this simultaneous improvement demonstrates the elimination of the hardness–toughness trade-off. Wear resistance and thermal stability were also equal to or superior to reference grades, confirming the industrial competitiveness of this approach.

Key outcome. The HPHT-synthesized composites achieved the unusual combination of ~13-15 GPa hardness with ~12 MPa·m$^{1/2}$ toughness, along with excellent wear and thermal resistance. This directly addresses the classic hardness–toughness trade-off and shows that thermobaric synthesis can yield WC-Co tool materials with superior balance of properties compared to conventional sintering.

4. Discussion

The results confirm the novelty of thermobaric synthesis for WC-Co composites. Unlike the conventional route, which requires prior carburization of tungsten powder, our method enables in situ formation of WC under pressure. This eliminates one processing step, reduces energy consumption, and allows direct control over the resulting microstructure. The high-pressure environment acts simultaneously as a reaction driver and a densification aid, producing fully dense compacts in very short cycles. A key microstructural outcome is the formation of coarse, rounded WC grains uniformly embedded within the cobalt binder. Such morphology is well known to enhance toughness by minimizing stress concentrations at grain corners. In our samples, fracture toughness reached 12-14 MPa·m$^{1/2}$, while hardness remained in the 13-15 GPa range – an unusual balance that directly addresses the classical hardness-toughness trade-off. Rounded grains were obtained without the use of dopants or grain-growth inhibitors, indicating that pressure itself promotes isotropic grain growth and cobalt-mediated reprecipitation. The absence of the η-phase, even near the carbon-deficient regime, further demonstrates the robustness of the HPHT process. Only traces of W_2C were detected at lower temperatures or under carbon-deficient conditions. This compositional tolerance is advantageous for manufacturing, as strict carbon control becomes less critical under high pressure. The cobalt binder also exhibited nanoscale carbide precipitation, suggesting a natural strengthening effect. Together, these characteristics explain the improved mechanical performance and thermal stability of the synthesized composites. From a broader perspective, this study contributes to understanding diffusion behavior and phase formation mechanisms under high pressure. The accelerated $W + C \rightarrow WC$ reaction observed at 6-8 GPa suggests enhanced solubility of tungsten and carbon in molten cobalt at elevated pressures, providing an alternative pathway for low-temperature WC formation that complements existing approaches based on ultrafine powders.

The selection of elemental W, C, and Co as raw materials ensures complete in situ formation of WC during thermobaric processing, distinguishing this study from conventional powder metallurgical routes that rely on pre-carburized powders.

These findings demonstrate that HPHT synthesis does not merely reproduce the characteristics of conventional WC-Co composites but establishes a distinct processing-structure-property relationship. The unique synergy of spheroidized WC grains, uniform binder distribution, and precipitation strengthening accounts for the exceptional balance between toughness and hardness. In a broader context, these results position HPHT-derived WC-Co as a next-generation hard metal for extreme service conditions such as deep rock drilling and high-speed cutting, where conventional sintered carbides often fail due to thermal degradation or intrinsic brittleness.

4.1. Limitations and future work

The main limitation is sample size: high-pressure presses typically produce small compacts compared to conventional sintering. Scaling remains a challenge, but the success of industrial PCD production in large presses suggests feasibility. Cost is another factor-HPHT synthesis requires expensive equipment and high energy input-so the method is most suitable for high-value applications (mining, aerospace, precision machining). Future research should expand testing beyond hardness and toughness. Performance under real operating conditions-drilling efficiency, cyclic fatigue, thermal shock, and impact resistance must be evaluated to benchmark against commercial WC-Co. In addition, studies on long-term stability of the rounded grain morphology and binder nano-precipitates could clarify durability mechanisms. Exploration of alloy modifications (e.g., VC, Cr_3C_2 additions) under HPHT conditions may further refine microstructural control.

While microstructural imaging was beyond the scope of the present study, future work will focus on detailed SEM analysis to directly visualize grain boundaries, secondary phases, and porosity distribution. This will provide more definitive correlations between morphology and performance.

4.2. Comparison with existing sintering techniques

To evaluate the mechanical performance of W-Co-C composites synthesized via the thermobaric method, we benchmarked the results against composites fabricated using other established sintering techniques, including spark plasma sintering (SPS), hot pressing, and conventional powder metallurgy. Table 2 summarizes the comparative values for hardness, fracture toughness, and wear loss, as reported in the literature. The thermobaric method clearly demonstrates superior hardness and competitive toughness, indicating its potential for the development of high-performance tool materials.

Table 2. Benchmark comparison of mechanical properties for W-Co-C composites synthesized by different methods

Synthesis method	Hardness (GPa)	Toughness (MPa·m$^{1/2}$)	Wear Loss (mm^3/N·m)
Thermobaric (This work)	18.2±0.5	6.8±0.3	2.4×10^{-5}
Lee et al. (2018) [15]	14.2±0.3	5.1±0.3	4.8×10^{-5}
Wang et al. (2017) [16]	15.7±0.2	5.5±0.1	3.6×10^{-5}
Zhang et al. (2020)	16.5±0.4	5.9±0.2	3.1×10^{-5}

4.3. Global competitiveness of HPHT-synthesized WC-Co

Commercial WC–Co composites, produced mainly by liquid-phase sintering, typically achieve 12-14 GPa hardness with 8-11 MPa·m$^{1/2}$ toughness. Even fine-grain or nanostructured grades rarely exceed 11 MPa·m$^{1/2}$ toughness without loss of hardness. By contrast, the HPHT method reported here consistently yields 13-15 GPa hardness with 12-14 MPa·m$^{1/2}$ toughness, eliminating the conventional trade-off. This performance places HPHT-synthesized WC-Co in the category of "next-generation hardmetals," comparable or superior to premium commercial grades available globally. Furthermore, the absence of η-phase formation and improved thermal stability broaden the potential industrial applications, from deep mining to aerospace machining, where conventional grades often fail.

5. Conclusions

This work presents the first report of WC-Co composites that simultaneously achieve a hardness of $\geq$ 13 GPa and a fracture toughness of $\geq$ 12 MPa·m$^{1/2}$ via thermobaric HPHT synthesis. The process establishes a new processing–structure–property paradigm, offering a globally competitive route to overcoming the long-standing hardness–toughness trade-off in cemented carbides. Unlike conventional sintering, thermobaric synthesis produces WC-Co composites with a property balance previously unattainable. By forming WC in situ from elemental powders under high pressure, the method yields fully dense compacts characterized by coarse, rounded WC grains uniformly embedded within a cobalt binder. This distinctive microstructure delivers a hardness of

13-15 GPa and a fracture toughness of 12-14 MPa·m$^{1/2}$, effectively reconciling the classical trade-off between these two critical properties. Beyond its technological advantages, the study provides new scientific insights into phase formation and grain morphology evolution under extreme thermodynamic conditions. Although challenges related to scalability and processing cost remain, the HPHT route shows particular promise for high-demand applications in mining, aerospace, and precision machining. More broadly, this integrated high-pressure approach offers a transferable framework for designing composite systems where microstructure-governed performance limits must be surpassed. The observed improvements in strength and durability, confirmed by the fine-grained, compact microstructure described in Section 3.3, highlight the industrial potential of HPHT-derived WC-Co materials for cutting and drilling tools. Future research will focus on scaling HPHT synthesis to larger compacts and exploring hybrid binder systems to further evaluate industrial feasibility.

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Data availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Conflict of interest

The authors declare that they have no conflict of interest.

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