学 位 論 文 内 容 の 要 旨

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学 位 論 文 題 名

Synthesis of Novel TiN-based Hard Films by Thermal CVD for Metal Cutting Applications
（熱ＣＶＤ法による新規ＴｉＮ系硬質膜の合成と切削工具への応用）

The present dissertation focuses on the development of new ceramic hard coatings to enhance the performance of cemented carbide tools used in metal cutting applications, so as to meet the demands for lower production cost, lower environmental impact and resource conservation in the industry. In order to achieve the increase in cutting performance, novel ceramic hard coatings, including TiC_xN_{1-x} and $Al_xTi_{1-x}N$ films, have been synthesised via a moderated and low-pressure chemical vapour deposition (LP-CVD) process using $TiCl_4$–CH_3CN–C_2H_4–H_2 and $AlCl_3$–$TiCl_4$–NH_3–Ar–H_2 precursor system, respectively. The effect of the composition on microstructure, hardness, adhesion, tribological property, phase stability and cutting evaluations are investigated.

In case of TiC_xN_{1-x} coatings, the results show that an increase of x in TiC_xN_{1-x} will increase the hardness due to a decrease in crystallite size. The maximum hardness can be achieved at 28.6 GPa for x values ranging from 0.66 to 0.69, which coincides with the minimum crystallite size of 39 nm. Contrarily, the adhesion of the TiC_xN_{1-x} coatings to carbide substrates decreased with the increase of x, due to a change in the morphology and the formation of a discontinuous bonding interphase. The decrease in adhesion of TiC_xN_{1-x} coatings caused by differences in the chemical composition of the bonding layer and C-rich TiC_xN_{1-x} coatings can be suppressed by optimizing x (x=0.63) value in the MT-TiC_xN_{1-x} coating. Considerable improvements in grain structure, micro-orientation, mechanical properties, and friction coefficients of modified MT-TiC_xN_{1-x}(x=0.63) coatings in comparison with those of a conventional MT-TiC_xN_{1-x} (x=0.45) coating can increase the tool life as high as 30%–60% in steel and cast iron turning applications.

Furthermore, synthesis of Al-rich cubic (c-) AlTiN coatings via LP-CVD have been conducted in order to develop a higher performance hard coating and promote higher cutting speeds and dry cutting. The c-$Al_xTi_{1-x}N$ coatings with (100) and (111) preferred orientations and average x values of 0.82 and 0.73 can be deposited. Moreover, these species of c-$Al_xTi_{1-x}N$ coatings comprise of Al(Ti)N/c-Ti(Al)N nano-lamellae with average lamellar periods of ca. 4.5–7.7 nm in an as-deposited state, having Al content fluctuates along the <100> direction. The unique and self-organised nano-structures are considered to occur via spinodal decomposition which decreases the free energy of formation of the system, leading to higher phase stability at elevated temperatures. The phase stabilities and mechanical properties of the Al-rich cubic c-$Al_xTi_{1-x}N$ coatings were investigated using annealing and compared with a state-of-the-art arc physical vapour

deposition (PVD) c-$Al_{0.6}Ti_{0.4}N$ coating, demonstrated good thermal stability, and superior hardness above 1000°C. These considerable improvements in the thermal stability and mechanical properties of the Al-rich c-$Al_xTi_{1-x}N$ coating resulted in a significant increase in the cutting tool life, by factors of 2 and 10 compared with state-of-the-art CVD-TiCN/Al_2O_3 and PVD-AlTiCrN coated inserts, respectively, in cast iron milling operations.

In summary, the work reveals the effectiveness of CVD process on developing advanced hard coatings, the importance of understanding the relationships between composition, microstructure on their physical properties, and how the knowledge can be adapted to develop higher performance coating materials for the future.