The use of aliphatic alcohol chain length to control the nitrogen type and content in nitrogen doped carbon nanotubes

George Bepete,[a] Zikhona N. Tetana,[a] Susi Lindner,[b] Mark H. Rümmeli,[b] Zivayi Chiguvare[a] and Neil J. Coville[a]

[a]DST/NRF Centre of Excellence in Strong Materials and Molecular Sciences Institute, School of Chemistry, University of the Witwatersrand, WITS 2050, Johannesburg, South Africa

[b]Leibniz Institute of Solid State and Materials Research, P. O. Box 270116, D-01171, Dresden, Germany

george.bepete@students.wits.ac.za

Abstract

The nitrogen in N-CNTs can exist in a variety of oxidation states, the most important being the pyridinic type N and the quaternary N. Optimization of each type of N is important if the respective properties brought about by each type of N species are to be utilized in applications. To this end, we have synthesized a range of N-CNT materials using an ethanol/acetonitrile reactant mixture pyrolysed at different CVD synthesis temperatures and alcohol/acetonitrile mixtures with aliphatic alcohols with different chain length (ROH; R = CH_3, C_2H_5, C_4H_9, C_5H_{11}, C_7H_{15} and C_8H_{17}) to determine if the N incorporated can be influenced by temperature or oxygen content in the precursor. The C/N ratio in the precursor mixture of each different alcohol increased with increase in alcohol chain length, for example from 7.1 for a methanol/acetonitrile mixture to 12.6 for an octanol/acetonitrile mixture. XPS analysis of the CNTs produced from an acetonitrile/ethanol mixture using different CVD temperatures (700 – 1000 ^oC), revealed that nitrogen incorporation in N-CNTs decreased with an increase in CVD temperature and that the type of nitrogen species incorporated also varied. Molecular nitrogen and a low content of pyridinic nitrogen was obtained in N-CNTs grown at 700 ^oC and 800 ^oC, while quaternary nitrogen was noted in all N-CNTs grown. The N content in the N-CNTs grown at 850 ^oC increased with the alcohol chain length and also controlled the nitrogen species incorporated, an effect related to the oxygen content of the reactant mixtures.

References

[1] Bepete G.; Tetana ZN.; Lindner S.; Rümmeli MH.; Chiguvare Z.; Coville NJ. Carbon, **52** (2013), 316-325.

Figures

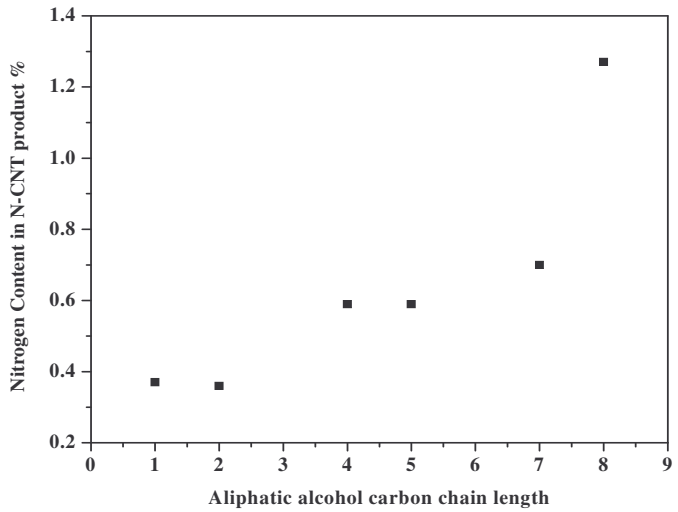

Figure 1. The N composition in N-CNTs made from the different alcohol precursor mixtures obtained from XPS analysis.

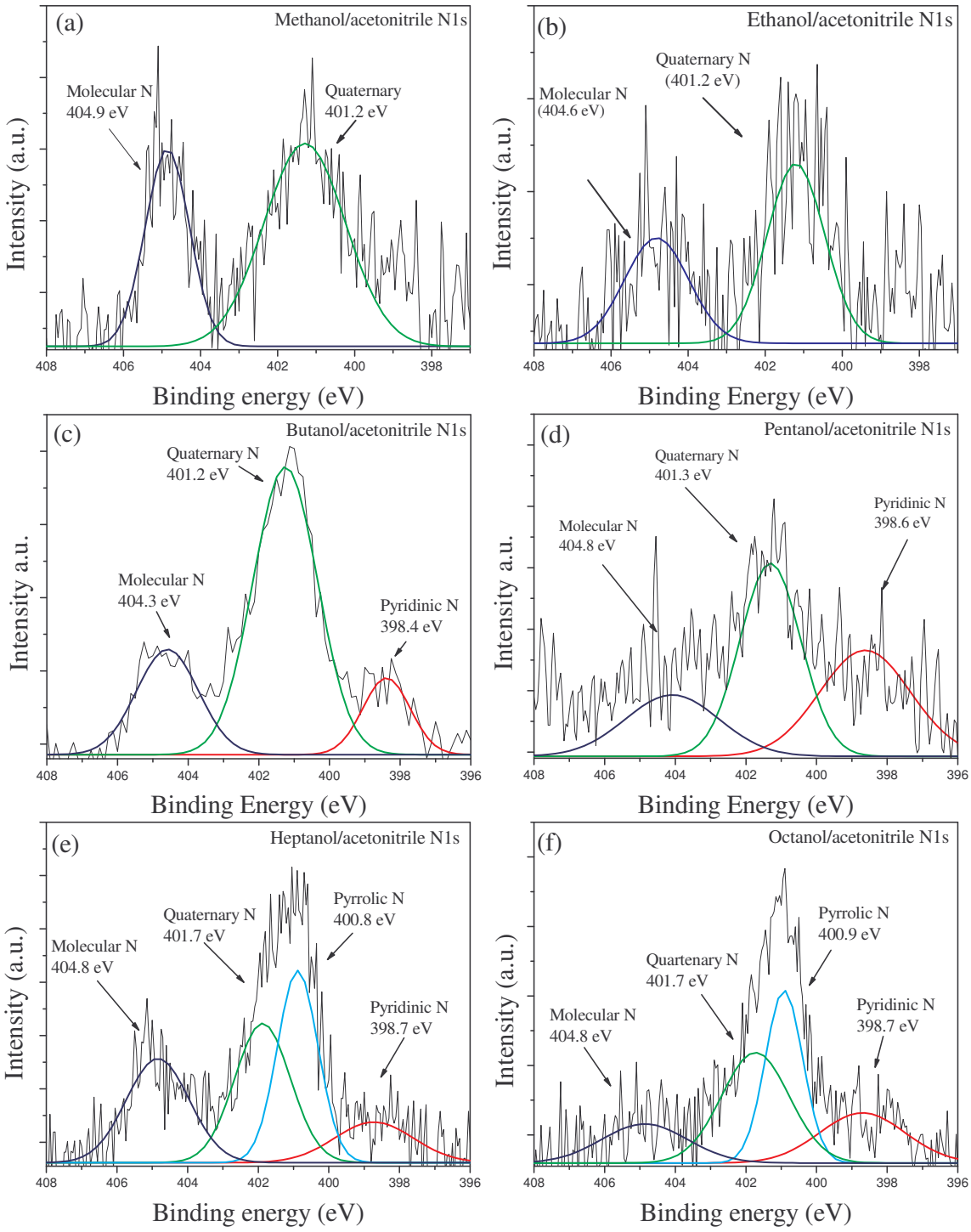

Figure 2. XPS N1s spectra of N-CNTs produced in CVD at 850 ^{o}C showing different types of nitrogen incorporated in N-CNTS when (a) methanol, (b) ethanol, (c) butanol, (d) pentanol, (e) heptanol, and (f) octanol were used in the precursor.