Unraveling the crucial role of Cu(I) intermediates in self-reducing MOD inks for flexible electronics

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The integration of electronic components in flexible materials such as paper, textiles and plastics is identified as a key challenge for the development of future smart applications including sensors, wireless communication and wearables. In this perspective, the fast and scalable low-temperature deposition of nanoscale metallic features is of utmost importance. Recently, a new class of copper inks, consisting out of self-reducing amine-based formato complexes was developed.[1,2] These novel inks yield metallic depositions with an outstanding electrical conductivity (± 10 μΩ cm) at temperatures of 150 °C or less, well below the reduction temperature of neat copper formate (around 225 °C). However, a fundamental explanation for this reaction mechanism and the corresponding temperature shift upon coordination is currently missing. In this work, a wide variety of self-reducing copper complex inks were synthesized and analyzed in detail using single-crystal XRD, EXAFS, FTIR and Raman spectroscopy. In addition, the reduction mechanism was monitored using in-situ XRD and XANES, in combination with cutting-edge statistical data treatment methods such as principal component analysis (PCA), revealing the crucial role of the Cu(I) intermediate and its complex structure.[3] Eventually, recommendations for the design of effective inks can be formulated, contributing to a future of flexible printed electronics.

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1 A.Yabuki, S. Tanaka, Materials Research Bulletin, **2012**, *47*, 4107.
2 Y. Farraj, M. Grouchko, S. Magdassi, Chemical Communications, **2015**, 51, 1578.
3 W.Marchal, PhD thesis, **2018**.