

# ELECTROCHEMISTRY OF taC:N FILMS PREPARED BY A SINGLE ARC METHOD

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Recent work<sup>1-4</sup> has shown that thin films of tetrahedral amorphous carbon incorporating nitrogen (taC:N), prepared with dual carbon and nitrogen ion beams at ambient temperatures, have attractive electrochemical properties. These diamond-like electrodes have high stability and marked hydrogen and oxygen overpotentials, yet are more catalytic than boron-doped diamond with respect to halogen/halide and quinone/hydroquinone systems<sup>1</sup>. The dual beam ion synthesis of taC:N with filtered cathodic carbon arc and nitrogen gun was pioneered at Nanyang University, Singapore, which supplied samples deposited on silicon wafers to us for the earlier electrochemical experiments<sup>1-4</sup>. In an accompanying paper<sup>5</sup> for this meeting, a method employing a filtered arc carbon beam ion operating at variable pressures of nitrogen gas has been shown to produce comparable films at CWRU. Either the single or dual beam method is suitable for deposition on virtually any surface without the high temperatures and nucleation problems of microcrystalline diamond deposition. This paper describes preliminary experiments, which demonstrate that the single beam produces<sup>5</sup> films equivalent electrochemically to those<sup>1-4</sup> evaluated earlier.

We have prepared films by the single beam method on a number of substrates but will restrict our discussion here to Ta which has passive properties that eliminate the ambiguities of possible porosity of taC:N at the few nm thicknesses involved here. After deposition as described<sup>5</sup>, the samples on Ta were assembled in an O-ring defined Teflon cell holder with 0.28 cm<sup>2</sup> working area. Saturated calomel electrodes(SCE) and carbon rods were used as reference and counter electrodes, respectively. Potential scans of the pure Ta surface in the same electrolytes accompany those of the taC:N coated electrodes to show that none of the electrochemical activity can be ascribed to pore exposure. The behaviors of the electrodes to the Ru(NH<sub>3</sub>)<sub>6</sub><sup>+3/+2</sup> couple in 1M KCl and to the positive potential extremes in 1M HCl were selected for comparison to previous results<sup>1</sup>.

In Figure 1, cyclic voltammograms with the reversible Ru couple clearly demonstrate the electron transfer properties of the taC:N film where the Ta substrate is completely blank. Plots of peak-to-peak potential separation and peak current vs. square root of scan rate of these data in Figure 2 show the expected linearity of the latter and near reversibility of the former (72 mV extrapolation to zero scan rate) for this couple. In Figure 3, a cyclic voltammogram at 50 mV/s in 1M HCl, held at the positive extreme of each scan for 10 s, shows the reduction of Cl<sub>2</sub> in the reverse scan at about 0.5 V vs. SCE at taC:N and no activity at Ta, even when taken about a volt more positive. This behavior is similar to the selectivity for chlorine evolution at the taC:N surface earlier shown<sup>1</sup>.

More extensive characterization of these films and correlation of physical and spectroscopic examination with electrochemical and substrate junction behavior is continuing.

## REFERENCES

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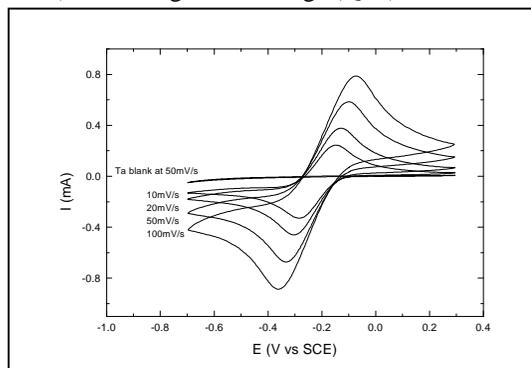


Figure 1. Cyclic voltammograms of taC:N on Ta in 10 mM Ru(NH<sub>3</sub>)<sub>6</sub>Cl<sub>3</sub> in 1M KCl at different scan rates, compared with blank Ta at 50mV/s.

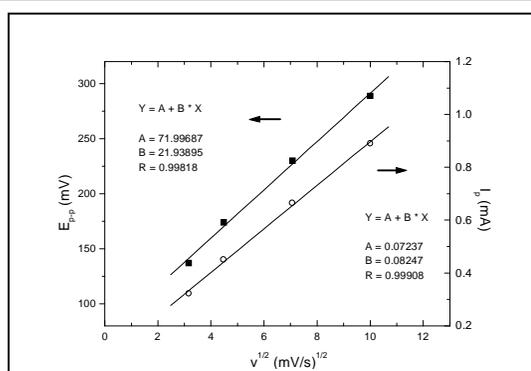


Figure 2. Plots of peak-to-peak separation potential and peak current vs. square root of scan rates.

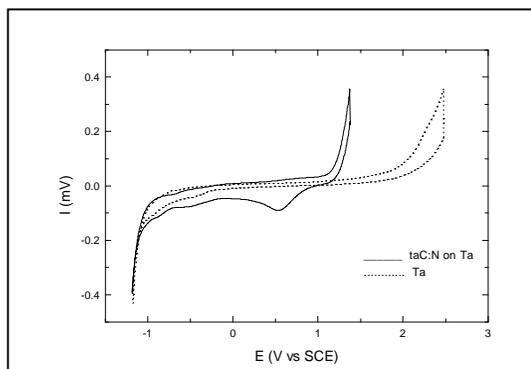


Fig. 3. Cyclic voltammograms of Ta and taC:N on Ta in 1M HCl at 50 mV/s. Both traces held at positive limit for 10 s before reverse scan.