## An in-materio reservoir computing with single-walled carbon nanotube/ N-confused tetra-tolyl-porphyrin complex

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**[Introduction]** In-materio reservoir computing (RC)<sup>[1]</sup> is gaining popularity as it paves the way for an energy-efficient computing performance compared to the present-day software approach. One such inmatrio RC device was obtained using single-walled carbon nanotube (SWNT)/porphyrin-polyoxoxmetalate (Por-POM) previously, which showed successful RC performance due to the non-linear redox dynamics originating from POM molecules enhanced in the presence of the porphyrin molecules.<sup>[2]</sup> The synergistic charge transfer effect of porphyrin and POM with SWNT was elucidated in that case, but the sole contribution from porphyrin was not explored as an in-materio RC. In this research, we study the effect of a porphyrin isomer called N-confused tetra-tolyl-porphyrin (NCTTP)<sup>[3]</sup> with SWNT on the RC performance. NCTTP consists of a twisted pyrrole ring with the nitrogen electron donating group pointing outwards, as shown in Fig. 1(a) inset. We fabricated an SW-NCTTP random network (RNW) device and studied its RC performance towards waveform generation. We showed that the non-linearity arising from the SW-NCTTP coupling is indeed beneficial for higher RC performance and that the porphyrin derivatives play a significant role in the design of an in-materio reservoir.

[Result and discussion] The SW-NCTTP in-materio RC device was prepared using the previous procedure via sonication and direct thin film transfer using vacuum filtration.<sup>[2]</sup> The surface modification of SWNT was confirmed from the UV-Vis spectroscopy, Fig. 1(a), where the Soret band of SW-NCTTP shows a redshift (442 nm) compared to the bare NCTTP (437 nm), indicating the presence of electronic coupling between the two moieties.<sup>[4]</sup> Such coupling leads to emergent non-linear sine wave outputs relative to the input as shown in Fig. 1(b), which we believe is a result of the charge transfer from the outwardly-pointing electron donating NH group of NCTTP. In addition, a set of linear output dynamics (Fig. 1(b) inset) is also observed, indicating that the RNW contributes from conductive SWNT. To elucidate the importance of such mixed dynamics, we studied the RC performance of triangular waveform generation, as shown in Fig. 1(c). The training of 5 readouts with only non-linear dynamics showed a 2% relative increase in test accuracy (98.4%, Fig. 1(c) top panel) than the training of 6 readouts with only linear dynamics (96.5%, Fig. 1(c) bottom panel). The reason can be understood from the FFT of the outputs in Fig. 1(d), where the non-linear dynamics give rise to higher harmonic generations relative to the linear outputs (Fig. 1(d) inset), making the reconstruction of triangular wave highly accurate. The result thus validates the importance of non-linearity towards higher information processing required for RC. Also, it confirms that a fault tolerance performance is achievable even with a lower number of readouts if nonlinear dynamics are present in the system.



Fig. 1(a) UV-Visible absorbance spectra of NCTTP (black) and SW-NCTTP (red). [Inset] Structure of NCTTP. Brown circle indicates the N-confused pyrrole ring. (b) Lissajous plots of 1 Hz,  $\pm 1$  V sine wave input vs. output of one electrode. [Inset] another electrode. (c) [Top] Triangular waveform fitting obtained by training all the non-linear output dynamics. [Bottom] One obtained from training all the linear output dynamics. (d) FFT obtained from t output signal in (b). [Inset] FFT of the inset of (b).

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