

Tracking Coupling Between Conformation and Mu-site addition in Antioxidants By μ SR and Theory

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Aging is associated with free radical reactions, processes known as oxidative stress and are the origins of degenerative disorders including heart disease, cancers & dementia. ~5% of inhaled oxygen escapes metabolic reactions, generating free-radical containing reactive oxygen species (ROS). Normally, natural mechanisms defend against free radicals and ROSs; if weakened, auxiliary mechanisms are required to maintain protection, most often by diets rich in free-radical scavengers. Flavenoids, antioxidant polyphenols (AntiOX), fall into this category with high quantities found in spices & bitter beverages; research on red wine & green tea have attracted a great deal of public attention, influencing health policy & personal habits.

However, the process by which flavenoids 'scavenge' free-radicals has yet to be quantitatively ascertained. The most common mechanism believed to be operative is via an AntiOX donating an H-atom to the ROS to form more stable phenoxyl radical which is more stable than the ROS, due electron delocalisation within the aromatic structures common to all AntiOX. An alternative mechanism has been proposed involving H-addition to the species acting as the AntiOX and is supported by clinical studies on blood serums using electron spin resonance for peptides. Therein, both product systems are stabilised, with the AntiOX forming stable H-added radicals of unknown structure and conformation.

We therefore set out to characterise the radical scavenging ability via H-addition of the green-tea polyphenol epigallocatechin gallate (EGCG) and its D-ring analogue 3,4,5-(OH)₃-benzoate (**Fig.1**) through characterisation of their neutral and radical forms, by inelastic neutron scattering (INS) on the TOSCA instrument (ISIS, RAL, UK) and Mu-spin resonance (μ SR) spectroscopy on the HiFi (ISIS, RAL, UK) & ALC (PSI, CH) instruments, respectively. Melatonin, another natural antioxidant, was likewise analysed, with similar success, providing the foundations of an emerging synergy between INS & MuSR experiments and theory.

The talk touches on the evolution, pitfalls and successful application of quantum theory (DFT and post-HF) in the design of INS and μ SR experiments.

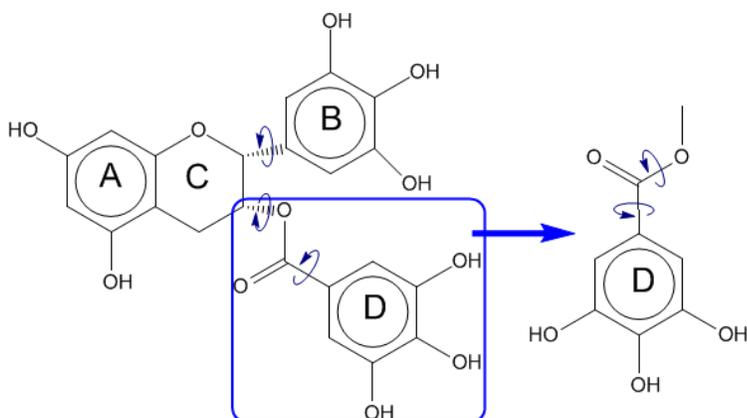


Fig.1 Structures of neutral EGCG parent compound including IUPAC ring nomenclature (left) and the D-ring analogue 3,4,5-trihydroxy-benzoate. The D-ring component is reported to be the portion responsible for the heightened radical-scavenging ability of EGCG with respect to other tea catechol anti-oxidant molecules.