DESIGN CONSIDERATIONS FOR AN ELECTRON PARAMAGNETIC RESONANCE SPECTROMETER FOR SIMULTANEOUS EPR AND NEUTRON DIFFRACTION

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Addressing the challenges of next-generation energy technologies requires advanced tools capable of probing both the electronic and structural evolution of materials in real time. This project was initiated in response to such demands, with battery research as a key motivator, to develop a dual-modality in situ instrumentation platform that integrates Electron Paramagnetic Resonance (EPR) spectroscopy and Neutron Powder Diffraction (NPD). EPR provides insights into spin states, oxidation states, local environments, spin dynamics, and material conductivity, while neutron scattering provides structural information on light elements within dense matrices. Its high penetration depth allows in situ studies without compromising structural resolution. This work synergizes the two powerful spectroscopy techniques.
Key to the success of this project is the development of a microwave cavity capable of performing EPR without hindering the Neutron diffraction studies. This work details the design of an Elliptical EPR microwave cavity (9.5 GHz) and additional hardware for insertion on a neutron beam line. COMSOL Multiphysics is used to design a high Q-factor Aluminium cavity with thinned walls, which retains high conductivity and offers a low neutron scattering cross section, thus minimising background scattering to optimise acquisition time and diffraction signal quality. Owning to Aluminium's high strength the walls are machined to 0.1 mm thickness, minimising un-wanted background neutron scattering. The novel use of modulation posts for the EPR (as opposed to Helmholtz coils) is proposed. The posts are strategically positioned inside the cavity, so as not to short circuit the electric field of the TM_{110} mode used for EPR. The modes are shown to be un-affected by the presence of the posts and a high Q-factor (>2500) is retained; critical for high sensitivity EPR measurements. The field uniformity and amplitude at sample position is comparable with conventional Helmholtz coils and demonstrates the efficacy of this topology. A low loss (<0.5 dB) waveguide feed with an iris and tuning puck end is designed and manufactured, enabling critical coupling (-50 dB, S_{11}) of the microwave source and detector to the EPR cavity. This hardware is integrated with microwave circuitry, a lock-in amplifier and electromagnet, forming a complete EPR spectrometer suitable for transport and configuration at a neutron beam line. The systems efficacy is shown by detection of EPR spectra on DPPH (2,2-diphenyl-1-picrylhydrazyl) and is subsequently used to measure EPR spectra of DPPH in-situ on the Polaris Powder Neutron Diffractometer (ISIS). These results are the first of its kind and demonstrates the potential for developing unique instrumentation that combines different spectroscopy techniques into a signal in-situ measurement and establishes the foundation for future correlative studies across a wide range of functional materials.

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Battery Research. Advanced Materials, 31(28), 1806620.