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Modification of graphene films in the flowing afterglow of microwave plasmas at reduced-pressure

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Graphene films were exposed to the late afterglow of a reduced-pressure N2 plasma sustained by microwave electromagnetic fields. X-ray photoelectron spectroscopy (XPS) shows that plasma-generated N atoms are incorporated into both pyridinic and pyrrolic groups, without excessive reduction of sp2 bonding. Nitrogen incorporation was found to be preceded by N adsorption, where N adatom density increased linearly with treatment time while aromatic nitrogen saturated. This finding was confirmed by Raman spectra showing a linear increase of the D:G ratio attributed to constant surface flux of plasma generated species. Combined Density Functional Theory calculations with a Nudged Elastic Band (DFT-NEB) approach indicate that incorporation reactions taking place at point vacancies in the graphene lattice requires an activation energy in the 2-6 eV range, but the energy required for the reverse reaction exceeds 8 eV. Stable nitrogen incorporation is therefore judged to be defect-localized and dependent on the energy transfer (6 eV) provided by N2(A)-to-N2(X) metastable-to-ground de-excitation reactions occurring at the late-afterglow-graphene interface. This represents one of the first experimental evidence of the role of metastables during materials and nanomaterials processing in non-thermal plasmas.

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