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Functional Graphene-Based Nanobioimaging Platforms: New Powered Real-Time Interfaces

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In the recent years, graphene (G)-based nanomaterials (e.g. graphene oxide (GO), G hybrid nanocomposites) are increasingly explored for real-time imaging of biomolecules or cells [1,2].

Indeed, the remarkable intrinsic and tunable properties of G and derivatives (e.g. planar structure, high surface-volume ratio, high electrical conductivity, good chemical stability and strong mechanical strength) are attracting much attention, especially to manufacture reliable and ultra-fast biosensing platforms (e.g. label-free or fluorochrome-based nano-optical/biophotonic detection systems such as FRET or CRET).

Thereby, a number of emerging studies have reported a combination of functional, green, cost-effective and scalable approaches to constantly improve the overall properties (e.g. sensitivity, specificity/selectivity, stability, rapidity) of the G component for real-time and multiplexed imaging of biomolecules (e.g. biomarkers of disease such as BRCA1, p53, PSA, AFP, glucose, DNA alterations) or cells (e.g. cancer cells, stem cells, bacteria or viruses).

Interestingly, most recent studies have reported functionalized G and derivatives- based bio detectors (i.e. G coating with noble metals such as gold and/or silver nanoparticles (NPs) [3,4], other chemicals such as nitrogen [5,6], poly-L-lysine [7-9] or biologicals such as charged lipid bilayer or biomolecules such as chitosan [10,11], with satisfactory results and higher benefits than conventional bio-imaging systems. These include high specificity/selectivity, high rapidity (ca. 2-6 seconds), high stability, high sensitivity and low detection limit (usually in the range of nM to aM with signal-to-noise ratio: 3), great reproducibility and reliability.

For instance, Gs-field-effect transistors (Gs-FETs) have been rapidly developed, and are currently considered as an alternative for post-silicon electronics. Indeed, Gs-FETs, as conducting channels, represent promising chemical and biological sensors. In particular, large-sized chemical vapor deposition (CVD)-grown G films have been configured as FETs for real-time biomolecular sensing (e.g. glucose or glutamate molecules) [12]. The underlying mechanism relies on the fact that the conductance of the Gs-FET changed as the molecules are oxidized by the specific redox enzyme (i.e. glucose oxidase or glutamic dehydrogenase) functionalized onto the G film. Further, Gs-FETs driven by a reference-gate operating in buffer solution exhibited very good transport characteristics, allowing biomolecular recognition with high precision and sensitivity [13].

Meantime, Gs-based fluorescence resonance energy transfer (Gs-FRET) biosensors were recently developed notably for simultaneous multi-molecular detection [14,15]. Indeed, Gs-FRET combine both the unique biomolecular adsorption ("wiring") characteristics due to G, and the "nanoquenching" capacity due to FRET. Importantly, in case of GO-FRET, fine-tuning of the oxidation is required as it could strongly affect its fluorescence quenching ability and binding interactions to biomolecules such as single-stranded oligodeoxyribonucleotides (ssODNs), leading to a broad range of sensitivity [16].

Also, graphene-based chemiluminescence resonance energy transfer (G-CRETs) has aroused particular attention. Indeed, chemi luminescence is being used as an exciting light source to construct universal and efficient G or GO-based photo-electrochemical sensing platforms [17,18]. In case of molecular detection (e.g. DNA) by GO-CRET system, the underlying mechanism involves that GO greatly inhibits the peroxidatic activity of a horseradish peroxidase (HRP)-mimicking DNAzyme [18]. Also, the bi-functionality of GO that can highly adsorb ssDNA and effectively quench the emission of organic dyes-probably due to its structural defects-is reasonably utilized in a CRET system, achieving sensitive and selective detection of various types of biomolecules [18].

Future directions might include the development of combined fluorinated Gs-based bioimaging systems using carbon-fluorine (C-F) as a tag and C-F spectroscopy (CFS) [19-21], as well as functionalizations of G and derivatives with diamond or diamond- like NPs to enhance the electrochemical and catalytic activities of Gs. Eventually, the growing demand for compact point-of-care medical devices and portable instruments for on-site environmental sampling is stimulating intense research on simple, enhanced and flexible Gs-based sensors that can be miniaturized and function under considerable physical deformation. That is all to say that G is definitively having a bright future in real-time molecular imaging dynamics.

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