Ultrafast low-temperature reduction of graphene oxide based on atmospheric pressure plasma

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Reduction of graphene oxide (GO) is a promising technique for large-scale production of graphene. The conventional way of removal of oxygen functional groups is based on either high-temperature vacuum annealing (above 500°C)\textsuperscript{[1]} or chemical processing\textsuperscript{[2]} using strong reducing agents like hydrazine. A major drawback of the vacuum-annealing reduction technique is its inapplicability to temperature-sensitive substrates including polymers having melting temperatures close to 100°C. Chemical reduction is a reliable highly-efficient procedure, however, a several hours lasting wet chemical processing is required to completely reduce GO. As an alternative method, cold plasma-based reduction is a promising candidate [3]. Here, the vacuum-generated plasma such as RF or microwave plasma (working at pressure of approximately 1 mbar) has been applied. On the other hand, the atmospheric arc-discharge reduction of GO has been demonstrated recently [4] but due to its inherently high processing temperature is inappropriate for temperature-sensitive substrates and is hardly applicable to large-area reduction. In this regard, even though the low-temperature plasma generation at atmospheric pressure is a challenge, it would be an ideal solution for the fast GO reduction.

In this work, we employed Diffuse Coplanar Surface Barrier Discharge (DCSBD) to generate the non-equilibrium low-temperature plasma at atmospheric pressure which was applied to reduce GO in pure hydrogen and methane atmospheres as well as in the mixture of these two gases. We show that the results are equivalent with those for the GO reduction by vacuum annealing at 850°C. In addition to avoiding vacuum during the entire procedure, the high volume power density of DCSBD plasma (\textasciitilde 100 W/cm\textsuperscript{3}) guarantees extremely short reduction times. We show that merely 1 second in hydrogen plasma removes approximately 50% of the oxygen-containing groups from GO as evidenced by X-ray photoelectron spectroscopy. The longest treatment time studied was 10 seconds. Such exceptionally short reduction times along with no vacuum requirements renders the DCSBD plasma-based GO reduction a promising candidate for large-scale graphene production on any type of substrate.

References