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Nucleation and growth of graphene nanosheets in gas phase by decomposition of hydrocarbons and alcohols in microwave plasma

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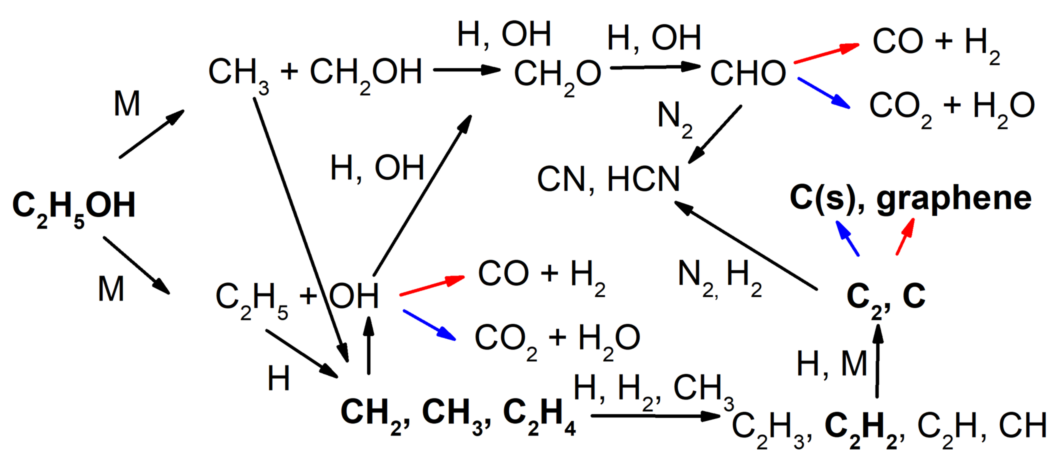
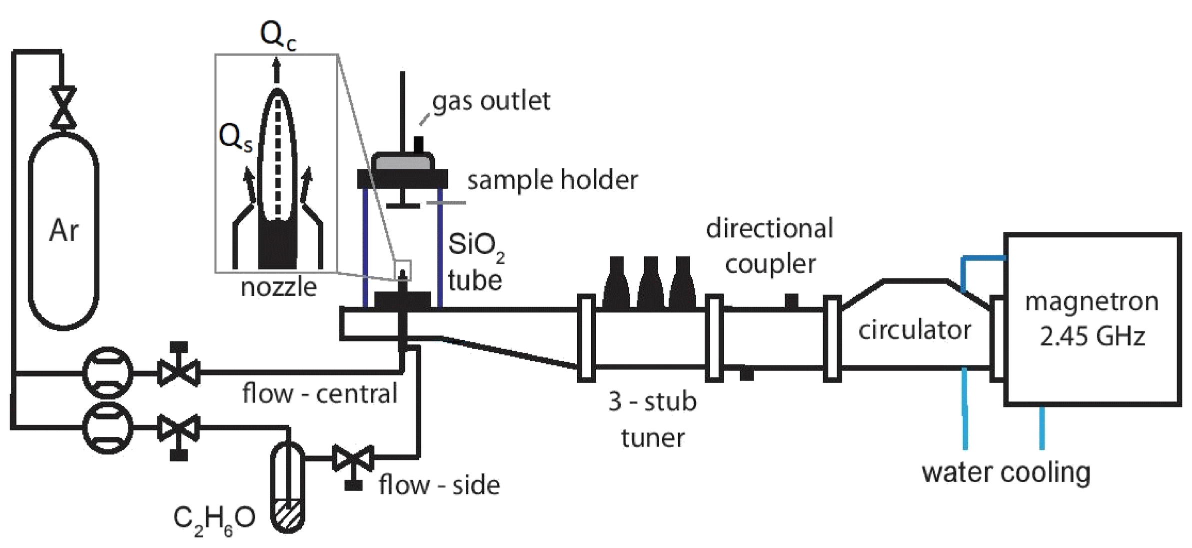
INTRODUCTION AND MOTIVATION

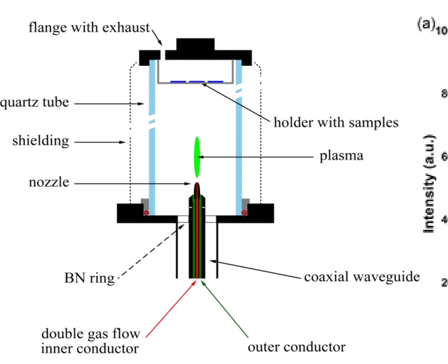
- **The decomposition of ethanol in microwave discharges** at atmospheric pressure argon atmosphere has been preferred method for the gas phase synthesis of graphene nanosheets. This method was first published and recently reviewed by A. Dato [1] and **represents simple and environmentally friendly alternative** to liquid or shear forces exfoliation of graphene.
- Application of high temperature plasma, microwave or DC plasma sources, and experimental results suggests that synthesis can be divided into two groups: a) **low temperature regime**, below 2000 K – producing CO_2, H_2O and **amorphous phase** and b) **high temperature regime**, above 4000 K, producing CO, H_2 and **graphene nanostructures**.
- This mechanism works well for ethanol, but with higher alcohols or aromatic hydrocarbons, the synthesis produces only amorphous nanomaterial. **Therefore, question remains how can we synthesize graphene in gas phase from higher alcohols or just simple hydrocarbons.**
- Recently, we showed [2,3] that instabilities in the dual-channel microwave plasma torch could be used for controllable formation of disorder in these carbon nanostructures.
- The nucleation and growth of the graphene in this environment depends on formation of C_2, CO and H_2 at high temperature, above 4 000 K. At high enough concentration of C_2 growth species produced by dehydrogenation of C_2H_y molecules, graphene **nuclei are formed and further grow on boundary between plasma (green) and neutral gas (orange thermal radiation). While oxygen and nitrogen impurities control amount of free C atoms, hydrogen controls nucleation and growth of graphene nanosheets** [4].

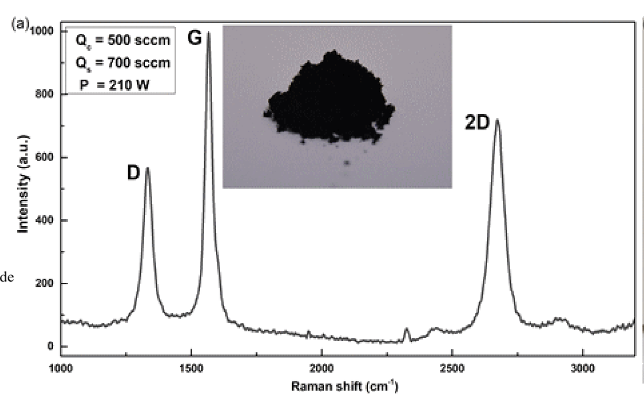
EXPERIMENTAL

- Microwave generator - 2.45 GHz, 100 -450 W, 2 kW maximum power
- Rectangular to coaxial waveguide transition terminated with dual channel carbon nozzle enclosed in the reactor chamber (ø 8 cm x 20 cm long quartz tube with aluminum flanges)
- Working gas, argon, was introduced through central channel ø 1.4 mm (Q_C 500 - 920 sccm) and argon carrying ethanol vapors was introduced into secondary channel annulus ($ø_i$ 7.7; $ø_o$ 8.4 mm)(Q_S 300–1400 sccm) corresponding to (0.1 - 5 sccm of 98 % ethanol). Methane, acetylene and hydrogen flow rate 5-100 sccm.
- **Overall efficiency of graphene synthesis for ethanol was over 8 wt%.**



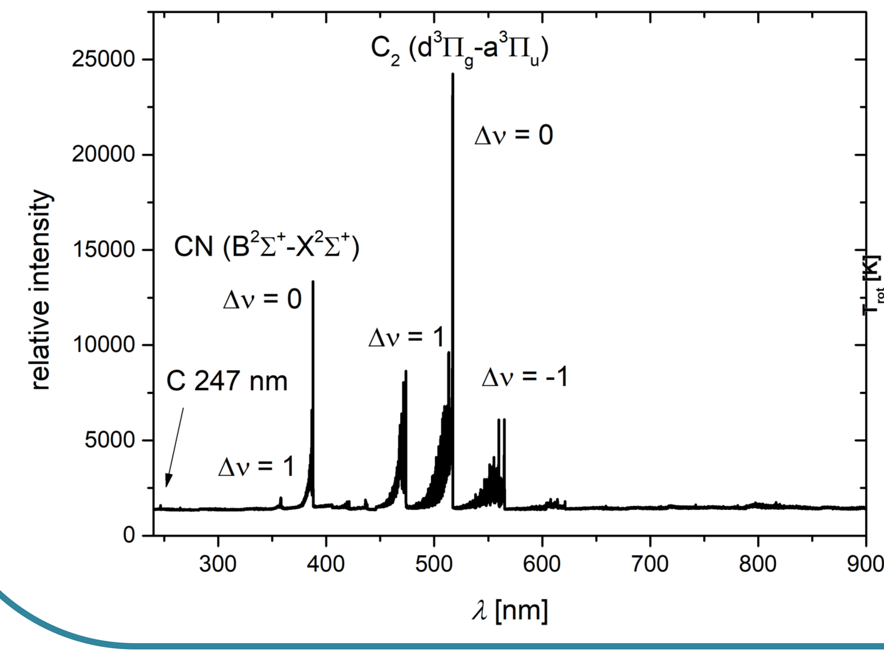


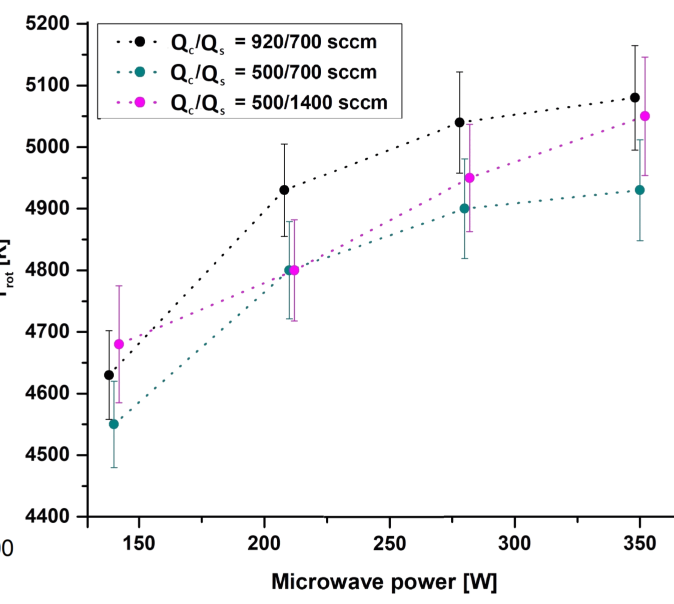




PLASMA DIAGNOSTICS OF MPT

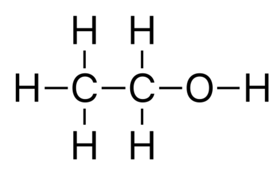
- Optical emission spectroscopy identified C_2 and C as main carbon containing species. C_2/C ratio was obtained from C_2 Swan system (516 nm) and C atomic line (247 nm).
- Rotational temperatures were obtained from CN transition (380-390 nm) and could be used to approximate neutral gas temperature T_{rot}.
- **C_2/C ratio and neutral gas temperature increased with delivered microwave power.**

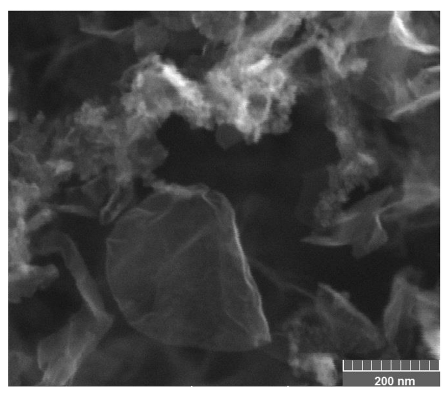


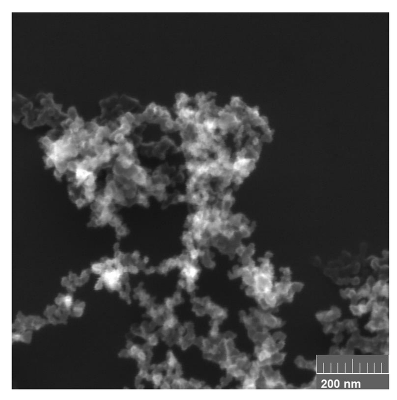


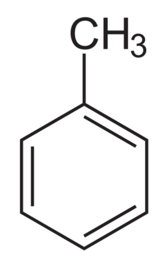
GRAPHENE NANOSHEETS SYNTHESIS

Ethanol vs Toluene. Why ?

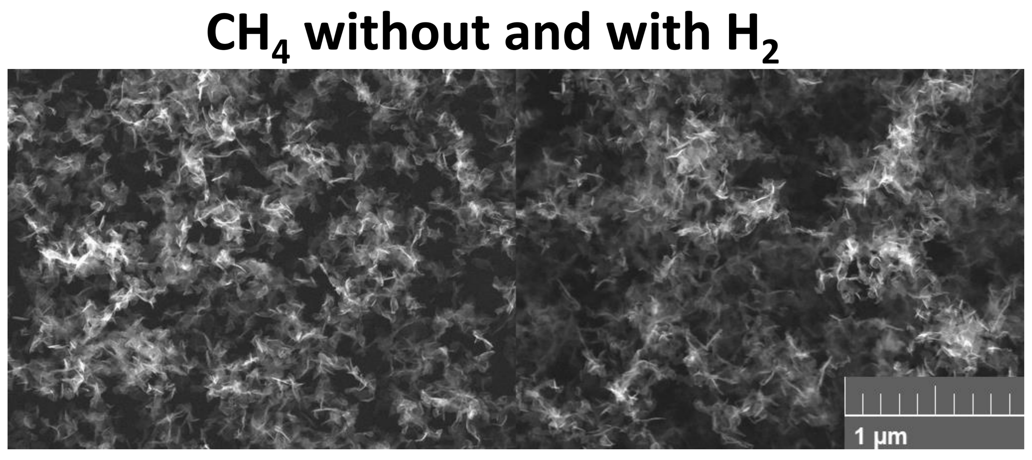


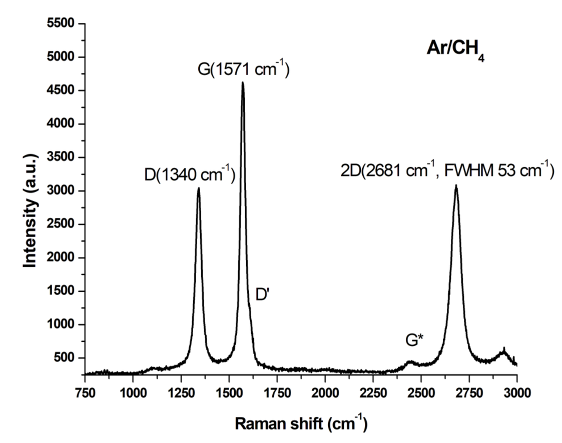




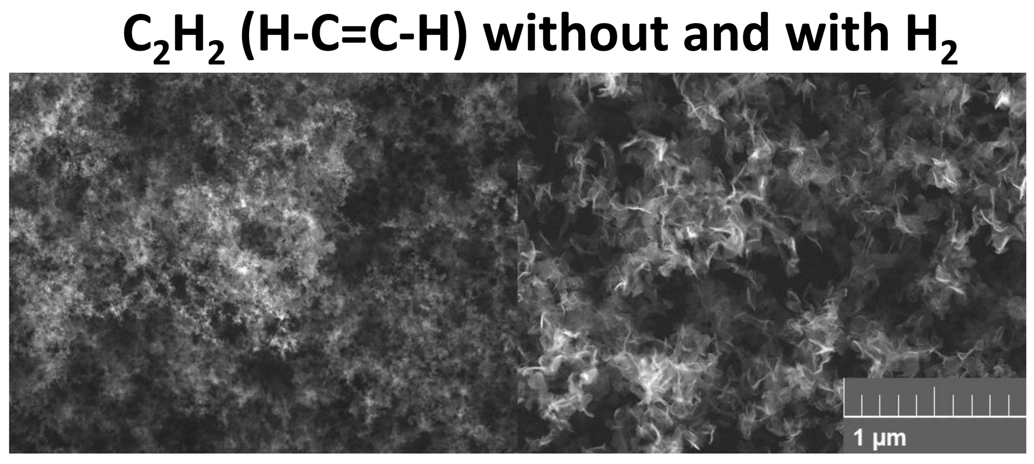


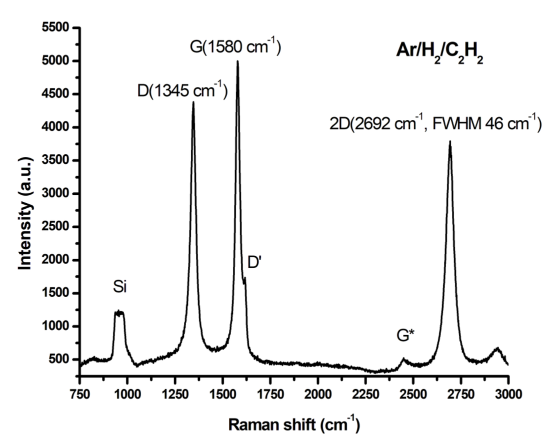
CH_4 without and with H_2





C_2H_2 (H-C=C-H) without and with H_2





Amorphous vs nanosheet structure

CONCLUSIONS

Graphene synthesis in gas phase was studied using decomposition of ethanol and simple hydrocarbons in microwave plasma torch at atmospheric pressure. Aromatic hydrocarbons do not automatically lead to formation of graphene nanosheets. **Control of Ar, H_2 and carbon precursor (ethanol, CH_4, C_2H_2) flow rates together with delivered microwave power led to synthesis of good quality graphene nanosheets in case of all precursors.**

ACKNOWLEDGEMENT

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REFERENCES

[1] A. Dato. J. Mater. Res., 34 (1), 2019, 214.
[2] Toman J, et al., Journal of Physics D: Applied Physics, 52(26), 2019, 265205.
[3] O. Jašek et al. Diamond and Related Materials, 105, 2020, 107798.
[4] O. Jašek, J. Toman et al. „Nucleation and growth of graphene nanosheets by decomposition of hydrocarbons and alcohols in gas phase“, submitted paper.

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