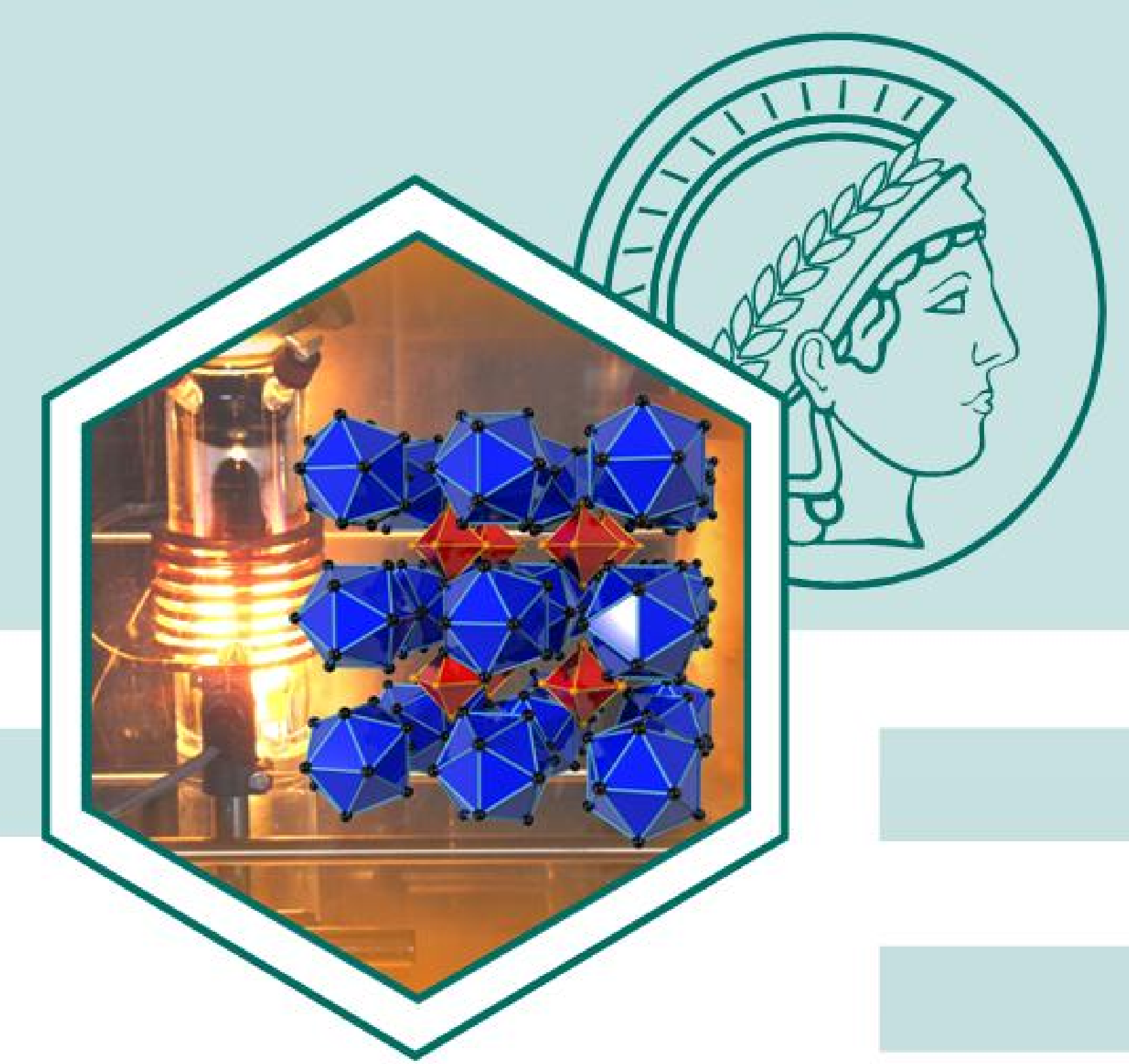


Synthesis of Carbon Nanostructures by the High Frequency Furnace Method

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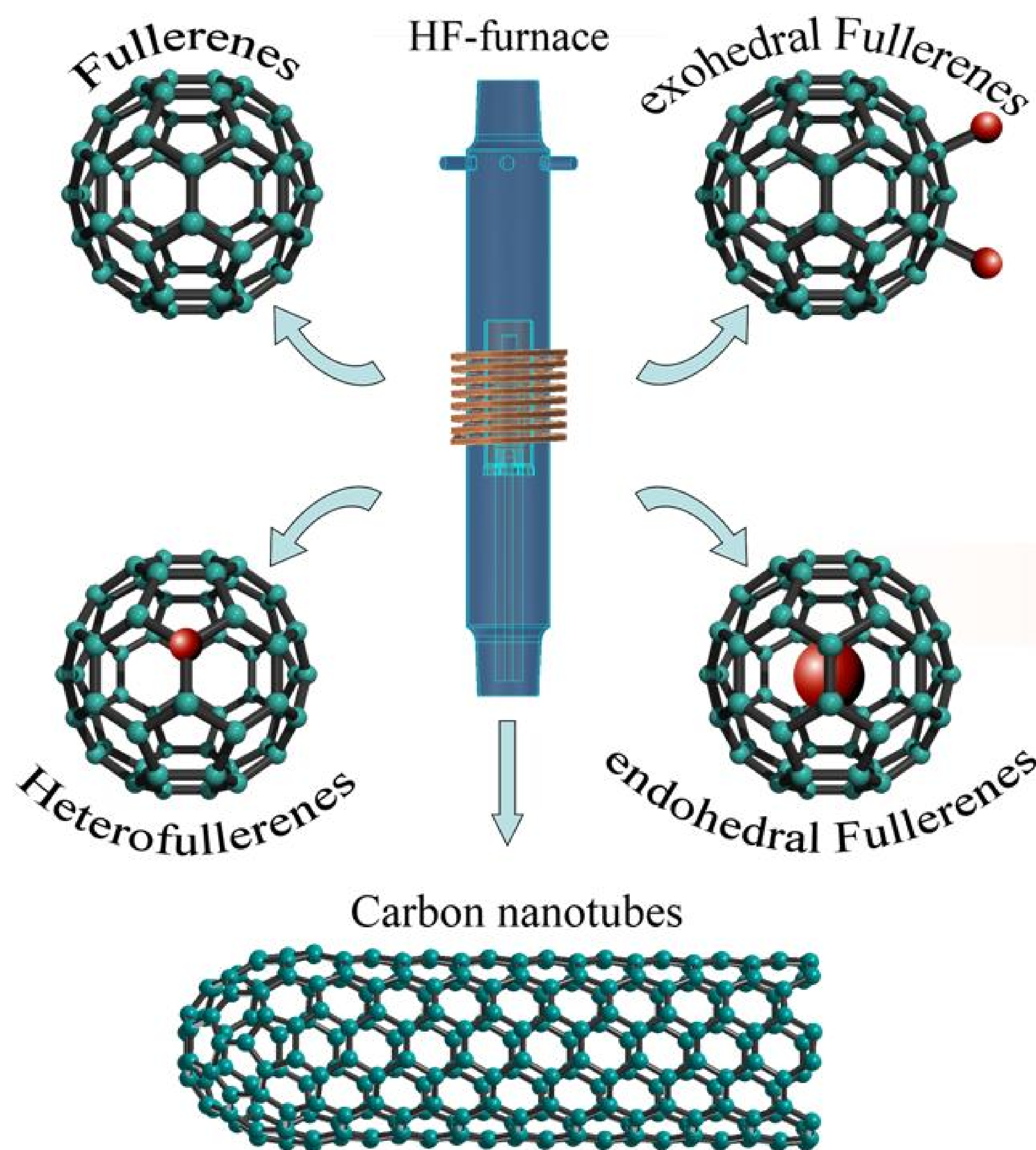
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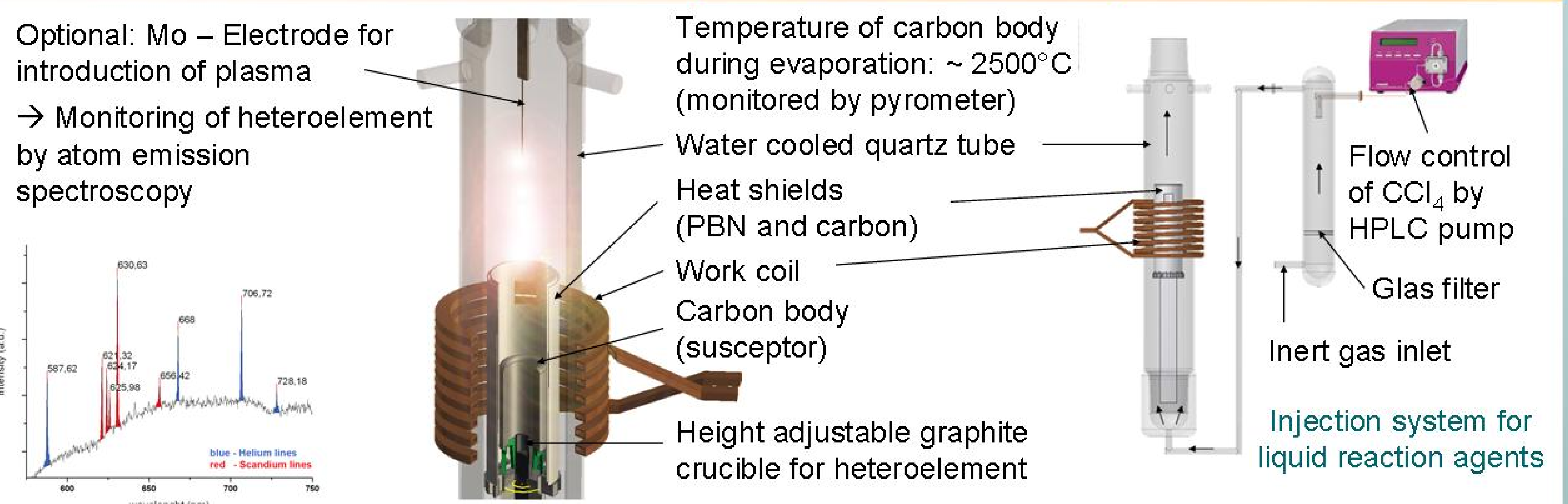
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Objectives / Introduction



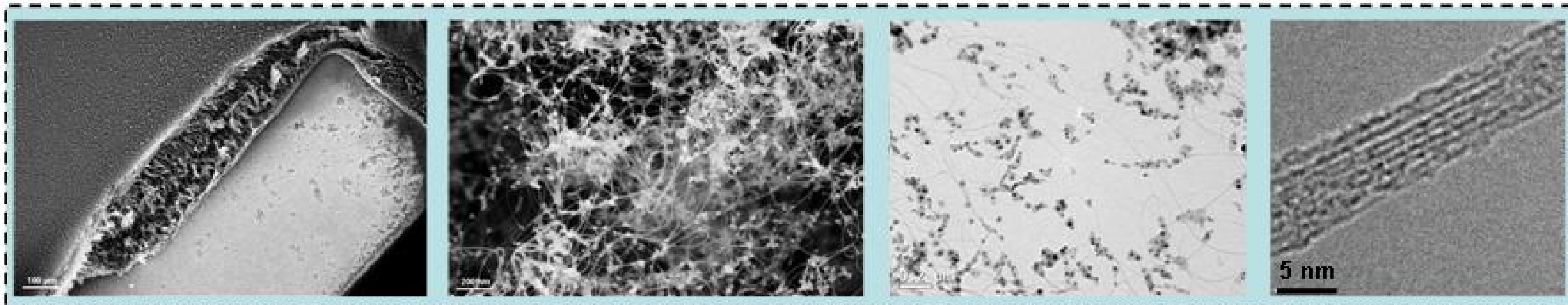
During the last two decades, nanostructures of carbon, among them fullerenes and carbon nanotubes, have attracted keen interest by the scientific community due to their structural richness and potential applications. The high – frequency furnace (HF-furnace) is an alternative synthesis approach which is based on the inductive heating of a graphite body as the carbon source.[1, 2] The possibility of independently co-evaporizing hetero-elements gives access to fullerene species, i.e. barium endohedral fullerenes and phosphor heterofullerenes which are not formed with the alternative methods.[3, 4] Exohedrally chlorinated fullerene species can be obtained directly by evaporating graphite in an atmosphere of carbon tetrachloride. This procedure gives access to new members of the fullerene family not obeying the IPR-rule. Moreover both single- as well as multi-walled carbon nanotubes can be synthesized effectively using the HF-furnace.[5]

Experimental setup: HF - Furnace



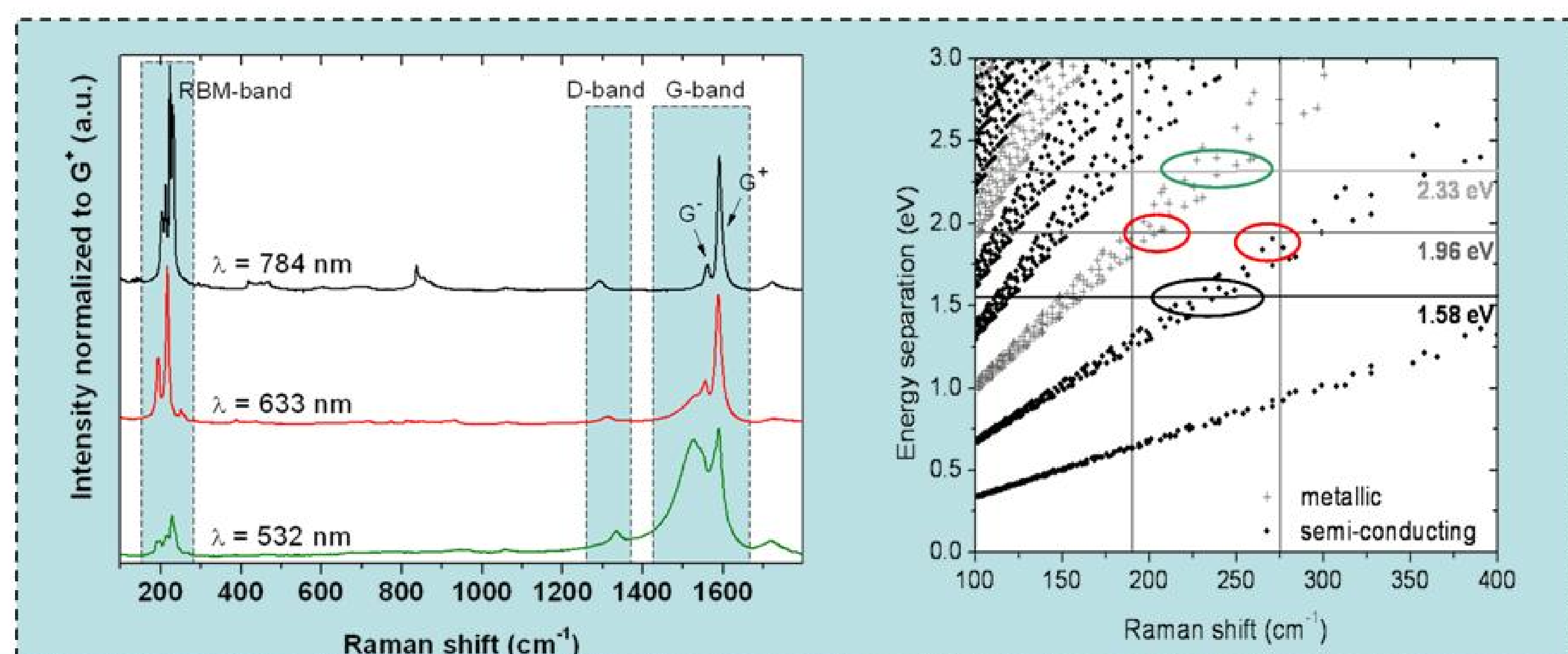
Results

Synthesis of single walled carbon nanotubes



SEM/TEM analysis: The black synthesis deposit consists of highly entangled bundles of SWCNTs and synthesis byproducts like carbon onions and carbon encapsulated metal nanoparticles. No MWCNTs were observed. The deposit is very uniform over the whole substrate regarding thickness and morphology. The bundle diameter ranges from about 14 nm down to individual SWCNTs in rare cases.

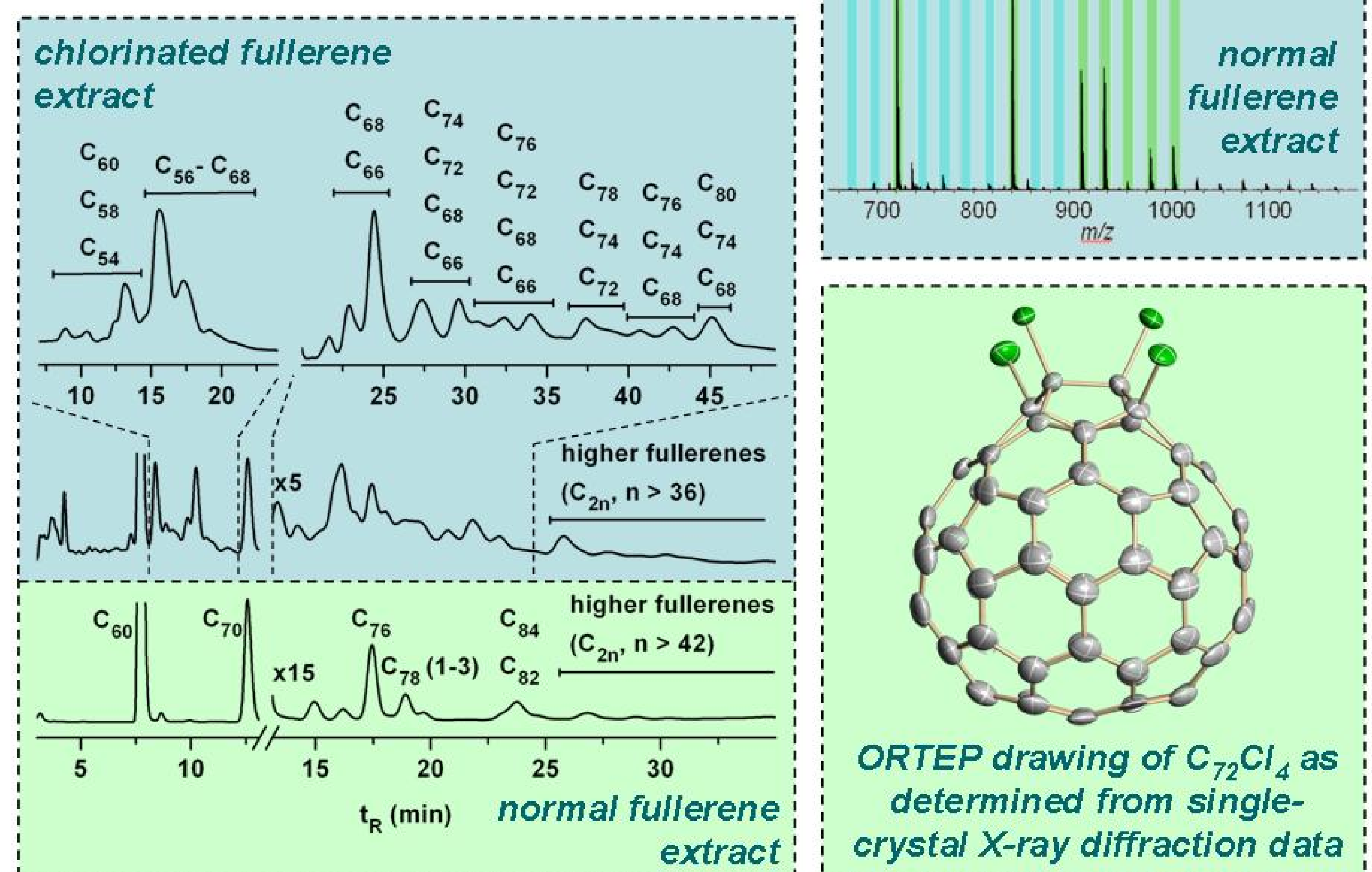
Raman analysis: SWCNT diameter lies between 1.26 – 0.84 nm (estimation from equation $\omega_{\text{RBM}} = 224 \text{ cm}^{-1}/d + 12 \text{ cm}^{-1}$). Both metallic as well as semi-conducting SWCNTs form during synthesis indicated by the G⁻-peak shape which shows typical shape of semi-conducting SWCNTs for the spectrum taken with 1.58 eV excitation energy and a Breit-Wigner-Fano peak shape (typical for metallic SWCNTs) for the spectrum taken with 2.33 eV excitation energy. The 1.96 eV spectrum shows contribution from both SWCNT types. For all three excitation energies a small D-band is observed which indicates "good quality" SWCNT material with low amount of defects.



Synthesis of chlorinated non-IPR fullerenes

By decomposition of carbon tetrachloride as reactive agent during the fullerene synthesis, various soluble fullerene cages not available without chlorine stabilization are obtained as shown by MS analysis of toluene extract. Especially, chlorinated species of non – IPR cages with C_{2n} , $n > 33$ are observed and can be synthesized in yields suitable for HPLC separation.

Chlorinated C_{68} , C_{72} and C_{74} cages were isolated and the molecular structure of $C_{72}Cl_4$ was determined by single crystal X-ray analysis from a co – crystal with CS_2 . Two adjacent pentagons are stabilized by 4 chlorine atoms.



Conclusions

- SWCNTs have been synthesized by the high frequency furnace method co-evaporating graphite and a metal catalyst
- The SWCNTs, both metallic and semi-conducting species, exhibit a diameter distribution between 1.26 – 0.84 nm and were found in a state of several μm long bundles
- No MWCNTs were observed in the substrate deposit

- New non-IPR fullerene species, with C_{2n} , $n > 33$, have been synthesized by introducing CCl_4 as reactive agent during the synthesis process
- HPLC/offline MS analysis indicates the presence of either different isomers of some species or different chlorination degrees for the same isomer
- Significant yields are obtained suitable for further isolation by HPLC, crystallization and X-ray characterization as demonstrated for the first example $C_{72}Cl_4$

References

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