

A-192

## Recrystallisation of Pyrolysis Carbon into Graphitic Materials

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Methane pyrolysis is a promising route for hydrogen production without direct CO<sub>2</sub> emissions [1]. However, the economic viability of methane pyrolysis strongly depends on the utilization of the solid carbon co-product [2]. Although pyrolysis carbon contains only minor impurities, its morphology and particle structure generally do not fulfil the requirements for established carbon black applications [3]. The transformation of pyrolysis carbon into graphitic materials therefore represents an important step toward closed carbon cycles.

In this work, a molten-metal recrystallisation process for the valorisation of pyrolysis carbon is presented [4]. The process concept originates from catalyst systems originally developed for the formation of carbon nanotubes and graphitic nanostructures from hydrocarbons [5]. Amorphous carbon is first dissolved in molten iron and subsequently precipitated through controlled cooling of the melt. Under oxygen-free conditions, graphitic carbon structures form on the melt surface, analogous to previously reported carbon precipitation phenomena in metal–carbon systems [6]. After proof of concept, an experimental reactor was developed to investigate process scalability. Inductive heating enables precise thermal control of the molten metal, while inert gas injection through a sparger separates graphitic particles from the melt surface and transports them pneumatically to a cyclone separator. A cyclic setup additionally allows continuous recharging of pyrolysis carbon.

SEM investigations reveal graphitic crystals with diameters of approximately 100 µm and thicknesses below 5 µm (Figure 1). Raman spectroscopy and XRD analysis confirm the formation of highly ordered graphitic structures with only minor deviations from a pristine hexagonal lattice. Ongoing process optimization focuses on reducing crystal thickness and the number of atomic layers toward graphene nanoplatelets.

The results demonstrate the successful transformation of a low-value carbon co-product into high-value graphitic materials. Potential applications include polymers, conductive inks, specialty cement, and batteries, thereby linking methane pyrolysis with advanced carbon-material value chains.

### References

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**Figure 1:** Concept of the molten-metal recrystallisation process for transforming amorphous pyrolysis carbon into graphitic carbon materials.