Formation of intermediate decomposition products (PAHs) during methane pyrolysis in a liquid metal bubble column reactor

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To counteract the progress of climate change, it is necessary to minimize CO_2 emissions by further developing the energy system. Besides established hydrogen consumers in metallurgy such as the production of molybdenum and tungsten powder from their oxides, the iron and steel industry among others is now also considering the increased use of hydrogen as an energy carrier and reducing agent. With this background, the production of hydrogen must be massively expanded and further developed to meet the increasing demand in metallurgy and other energy-intensive industries. Nowadays, nearly 96 % of the global hydrogen production comes from Steam Methane Reforming (SMR) and other processes based on fossil raw materials at an environmental burden of about 10 t CO_2 per t H_2, accelerating the consequences of global warming. A promising path to produce clean hydrogen is via methane pyrolysis using molten metals. Compared to SMR, significant less CO_2 is produced due to conversion of methane into hydrogen and solid carbon, making this route more sustainable to generate hydrogen. Solid carbon can be further used for different applications, like for agricultural purposes, where the carbon must be very pure without contaminants (e.g., polycyclic aromatic hydrocarbons (PAHs)). Within the scope of this work, a liquid metal bubble column reactor at the Chair of Nonferrous Metallurgy of the Montanuniversität Leoben is improved with the aim to define the process engineering challenges and to find solutions for them. During methane pyrolysis a number of different hydrocarbons, including C_2-, C_6- hydrocarbons or PAHs, are formed next to H_2 and undecomposed CH_4. The different hydrocarbons are found both in the exhaust gas system and in the produced carbon, which is undesirable mainly because of their toxic properties. The condensates produced in the test set-up as well as the solid fractions are analysed for these formed products in order to build up a better understanding in the process of pyrolysis and to force the prevention of the formation of these PAHs. Temperature and process duration in particular influence the formation of the gaseous main components H_2 and CH_4 and the intermediate products of methane pyrolysis. The most abundant compounds of PAHs in all samples are built up of 4 aromatic rings (e.g., pyrene) followed by compounds consisting of 3 aromatic rings (e.g., anthracene). Interestingly, the amounts of PAHs are much lower if the molten metal is covered with a carbon layer. The development of a suitable process and reactor design in laboratory scale as well as the stepwise upscale to pilot scale is crucial for a full understanding of the interaction of all process parameters at different reactor sizes, gas injection systems and continuous product discharge.