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Response to Comment on "Methane Pyrolysis for Zero-Emission Hydrogen Production: A Potential Bridge Technology from Fossil Fuels to a Renewable and Sustainable Hydrogen Economy"

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In his comment, Mr. Keller[1] has elaborated in detail that the overall process chain of methane pyrolysis might not be a zero-emission technology as we stated in the title of our paper.[2] We thank Mr. Keller for his valuable and detailed comment on our review, as it highlights that hydrogen generation via methane pyrolysis is an important topic nowadays. The wording in the title was chosen from the viewpoint of catalysis, underlining that methane decomposition itself is a reaction without any greenhouse gas (GHG) emissions, at least theoretically. The main intention of our review was to work out the open scientific questions, which have to be tackled for this reaction to enable the application of this technology as a bridging building block to a sustainable future hydrogen supply. However, we agree with Mr. Keller that for its later application one can not only take into account the emissions of the methane pyrolysis alone but also has to consider the complete process chain in terms of its environmental impact.

In general, methane pyrolysis as a net reaction provides a good perspective for a transitional hydrogen supply without the emissions of GHGs until enough sustainable hydrogen by, for example, electrolysis applying renewable energies is available. However, as there is no industrial process available yet, the intrinsic questions must be solved before implementation. These questions are related to a better understanding of the reaction, especially in terms of mechanism, and determining reasonable process conditions in combination with a suitable catalyst for these operating conditions. Without further development in this field, deployment during the transition period to a sustainable future hydrogen supply is not foreseeable. Therefore, a complete life cycle assessment (LCA) was out of the scope of our review but, of course, has to be considered if the application of methane pyrolysis becomes feasible.

We also agree with Mr. Keller's statement that global warming induced by GHGs should not be reduced just to CO_2 emissions as actual debates may indicate. As the emissions of CO_2 are far higher compared to other GHGs worldwide,[3] the focus is usually set on the most prominent perpetrator to simplify the discussion. Still, due to their higher global warming potential (GWP), the impact of other GHGs like CH_4 is becoming more severe with increasing emissions and cannot be neglected. As natural gas will still play a role in the transition from a fossil-based to a renewable and sustainable economy, natural gas production should be optimized in parallel to the development of methane pyrolysis to reduce the accompanied methane emissions. According to estimations from International Energy Agency (IEA), already today 40% of the actual methane emissions in the oil and gas sector can economically be avoided, and roughly a similar value can be avoided with existing technologies.[4] Implementing such measures would make use of natural gas and subsequent methane pyrolysis for hydrogen production much more efficient in terms of environmental impact. However, these are tasks more for industrial engineers and politics rather than science.[5]

Besides the required optimization of natural gas production and development of methane pyrolysis to industrial scale, an additional point to consider is that the envisaged transition can only take place gradually as it is unrealistic to replace all existing systems in parallel on the short term. Therefore, fossil-based processes are required in between to compensate for the lack of economic renewable and sustainable concepts and infrastructure. In this context, detailed cost estimation of methane pyrolysis and an overall LCA can give important information for where the implementation of methane pyrolysis provides the highest benefit regarding economic use and environmental impact.

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