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Techno-economic and environmental survey of an industrial gasification and methanation plant for the production of synthetic methane from tyre crumb

P.-A. Maitre¹, J. Cren¹, S. Valin¹, E. Goudal², E. Philippe², J.-P. Faure³

Abstract: A new potential industrial pathway to valorise rubber from worn tyres into synthetic methane is assessed via a techno-economic and environmental analysis. In this process, tyre crumb is firstly gasified in an Entrained Flow Reactor (EFR), which produces syngas that is further adjusted to a H₂/CO ratio of 3, cleaned and finally reacted in a methanation reactor to produce synthetic methane. An industrial-scale gasifier (2.5 t/h) was modelled and all other elementary steps of the plant scaled accordingly. Two different pathways were considered: the first one, whereby the syngas adjustment is conducted by a Water Gas Shift (WGS) reactor, and a second one in which H₂ produced by an electrolyser is added to the gas stream. Both pathways were shown to have a similar global energy yield, whilst the electrified process demonstrated a significant improvement of carbon use (89 % vs. 35 %). The specific CO₂ content of the synthetic methane reached 400 kg/MWh_{LHV} and 227 kg/MWh_{LHV} for the pathway with WGS and the electrolyser, respectively. Finally the Levelised Cost Of methane (LCO_{CH₄}) was estimated to be equal to 105 €/MWh_{HHV} for the pathway with WGS and 118 €/MWh_{HHV} for the pathway with the electrolyser.

Keywords: synthetic fuel, waste valorisation, electrolysis, carbon assessment

1 Introduction

Contrary to paper and most plastics, rubber from tyres can hardly be recycled to other uses due to the various nature of inseparable elements it contains (silica, sulphur, carbon, zinc). For this reason, 16.5 % of worn tyres are retreated when possible (especially in the heavy-transportation industry), whilst 41 % are repurposed in the shape of tyre crumb for various uses (e.g. backfills, retention ponds, playground coatings). Importantly, 42.5 % of worn tyres are utilised for heat generation in heavy industries such as cement/steel production or directly in incinerators for district heating. The production of waste tyres is very unlikely to decrease in the future as the whole industry of road transportation (heavy and individual) depend on the use of tyres. Actors from the field therefore seek alternative source of valorisation with higher remuneration potential.

In this frame, Aliapur, a waste tyre specialist (376,000 tons of recycled / recovered tyres in 2019)

in collaboration with GRDF (main distribution system operator for gas in France) have commissioned the CEA to assess the feasibility of an industrial process that would allow the conversion of tyre crumb into synthetic methane. To this end, laboratory tests of gasification of tyre crumb into syngas (mixture of CO and H₂) were carried out at the CEA on a pilot-scale entrained flow gasifier [1]. The experimental results showed that the gasification of tyre crumb can be performed with a very high conversion rate of carbon from solid to gas (>90 %), which confirmed the technical feasibility of the gasification of such resources in this type of gasifier. Starting from the results obtained on the pilot gasifier and extrapolating them to the industrial scale (20 kt/y), a techno-economical study encompassing other utilities and all the elementary bricks of a plant was performed to assess the economic viability of such process at the industrial scale. The environmental assessment of the produced synthetic methane was performed

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in terms of CO₂ emitted to compare it with other sources of gas and heat.

2 Gasification and methanation plant: considered pathways

The gasification process is very similar to the so-called coal to gas process for which an overall block-diagram is presented in Figure 1, pathway 1.

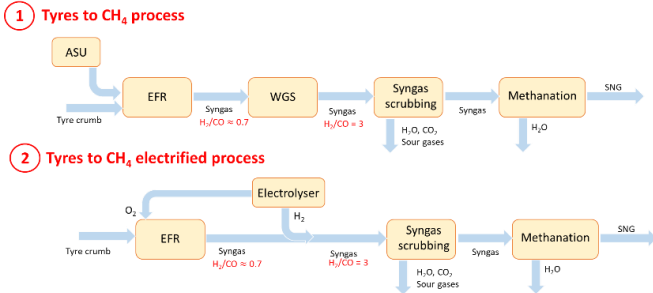


Figure 1: Block scheme of the two cases considered

The very fine tyre crumb (<1 mm; 80 % mass. C, 35 MJ_{LHV}/kg) is injected into an entrained flow reactor (EFR), operating at high temperature (>1400 °C) and medium pressure (10 bar) with pure oxygen (provided by an Air Separation Unit) and steam. This reactor is considered to operate in Auto-Thermal mode (heat is provided by the combustion of a fraction of the rubber resource). The syngas obtained at the reactor outlet contains H₂O, CO₂, COS, H₂S and in larger proportions CO and H₂ with an H₂ /CO ratio close to 0.7. This gas is then injected into a Water Gas Shift (WGS) reactor, where the CO is converted by water following the reaction: $CO + H_2O \rightarrow CO_2 + H_2$. The latter produces dihydrogen whilst consuming CO, thus making it possible to adjust the H₂/CO ratio to a desired value of 3 to maximize the final methanation synthesis. The adjusted syngas is first scrubbed with amines to remove water, CO₂ and the sour gases it contains. It is then injected into the methanation reactor to produce synthetic methane according to the reaction: $CO + 3H_2 \rightarrow CH_4 + H_2O$. The methanation reaction takes place with a CO conversion rate close to 100% and the water is condensed at the reactor outlet so that the synthetic produced methane is very pure. Note here that the choice was made not to consider the methanation of CO₂ according to $CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$, as this reaction, although manageable, has not yet reached the same level of industrial maturity as the methanation of CO.

A secondary process (Figure 1, pathway 2) which includes the use of an electrolyser is also considered in the study. The aforementioned electrolyser holds a double role: first, the hydrogen it produces is used to adjust the H₂/CO ratio (instead of the WGS reactor), maximising the conversion of the CO in syngas. In parallel, the pure O₂ stream produced by the electrolyser is injected in the EFR, reducing/removing the need for an Air Separation Unit. Note here that the French grid is considered to be the source of electricity for the electrolyser.

3 Results

3.1 Gasifier performance results

The experimental results obtained on the pilot-scale EFR were extrapolated to model an industrial reactor capable of treating 2.5 t/h of tyre crumb. During the experiments, it was found that the output composition of the experimental reactor corresponded to the values of thermodynamic equilibrium at 1450 °C. Consequently, for the sake of simplicity and as a first approach, the calculation of the output compositions was performed at thermodynamic equilibrium by minimizing the Gibbs energy, and considering a complete conversion of the solid (90 % carbon conversion experimentally measured). The steam inlet flowrate was adjusted so that no solid carbon remains at thermodynamic equilibrium. The modelling results are presented in Table 1. Note here that the cold gas efficiency (CGE) is equal to 75 %, which is a good performance though slightly lower than the CGE generally obtained with coal in EFRs (≥ 80 %) [2].

Table 1: Modelled gasifier performances

Power / Tyre crumb mass rate	23.4 MW / 2.5 t/h
Operating conditions	1450°C – 10 bara
O ₂ flowrate	1800 Nm ³ /h
Water in gasifier mass rate	851 kg/h
Cold gas efficiency	75 %
Syngas power	17,5 MW (LHV)
Total syngas out flowrate (CO, CO ₂ , H ₂ , H ₂ O, N ₂)	6 956 Nm ³ /h

3.2 Mass and energy balance at the scale of the whole plant

The two pathways previously presented (Figure 1) were run with the same design and operating conditions of the EFR. The rest of the elementary steps in the plant (WGS reactor, ASU,

electrolyser...) were scaled and integrated together according to a set of industrial-inspired hypotheses. The overall balance results are presented below in Table 2.

Table 2: Energy and matter balances for the whole plant

Scenario	EFR + Water gas shift	EFR + electrolysis
CH ₄ production(t/h)	0.93	2.2
CO ₂ on-site emissions (t/h)	4.2	0.68
CO ₂ /CH ₄ ratio	4.5 (-)	0.31 (-)
O ₂ need gasifier (t/h)	2.6	2.6
O ₂ production (ASU / electrolysis) (t/h)	2.6	5.2
H ₂ production by electrolysis (t/h)	0	0.65
O ₂ surplus (t/h)	0	2.6
Overall carbon yield	35 %	89 %

Scenario	EFR + Water gas shift	EFR + electrolysis
Tyre crumb in (MW)	23.4	23.4
Electricity consumption (MW)	1.6	36
Methane power (MW LHV)	13	30.6
Overall energy yield	52 %	52 %

From these results, several general observations can be made:

- The scenario with the electrolyser allows a significant increase of the carbon yield, going from 35 % to 89 %. Indeed, in the absence of WGS, the use of carbon in the form of CO is maximal and the only carbon missing in this yield is that of the CO₂.
- The oxygen generated by the electrolyser in the corresponding scenario is enough to supply the EFR reactor and even leave a surplus.
- As it could be expected, the total electricity consumption of the process with electrolysis is much higher (36 MW against 1.6 MW). However, at the end of the process, as more methane is produced, the overall energy yield is identical between the two scenarios considered.

3.3 Carbon assessment

The CO₂ emissions of the produced methane are various and happen at all the different stages of the preparation and consumption of the gas (Figure 2).

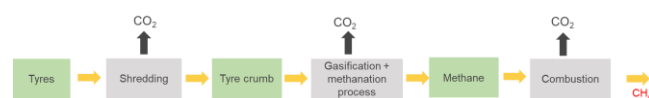


Figure 2: Different CO₂ emissions in the whole life cycle of the tyre crumb resource

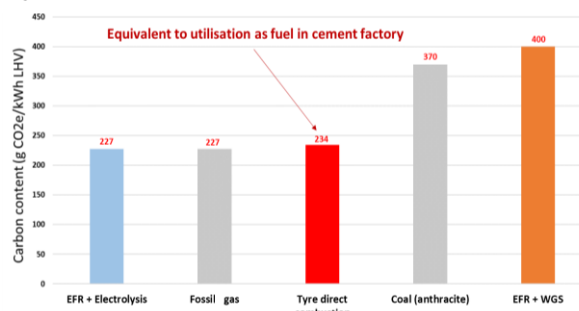
Three major emission steps are identified:

- Preparation: transportation, grinding, separation of tyre components, all of which are responsible for the emission of CO₂.
- Gasification: due to the WGS reaction, or in the gasifier. Non-direct emissions such as the electricity consumed at the plant are also considered. For the latter, a carbon content corresponding to the French average (of 52 gCO₂/kWh) was considered.
- Final combustion of methane.

The CO₂ content results per thermal kWh_{LHV} of CH₄ are gathered in

Figure 3.

Figure 3: Carbon content of the different pathways to heat production. Comparison with direct tyre combustion and fossil gas



It appears that the methane produced with the EFR + WGS process has a high carbon content of 400 gCO₂/kWh, when the EFR + Electrolysis scenario exhibits a value of 227 gCO₂/kWh, which is comparable to fossil gas combustion. This discrepancy between the scenarios is solely due to the difference in carbon yield between the two processes since the preparation of tyres and the combustion of methane are the same. To improve the overall carbon yield of the plant and thus reduce the carbon content of the synthetic methane, methanation of the CO₂ emitted at the plant should also be considered.

3.4 Economical evaluation

The production of synthetic methane was subsequently evaluated in economical terms. First

of all the direct investment costs, in terms/units of CAPEX ISBL were calculated (Figure 4).

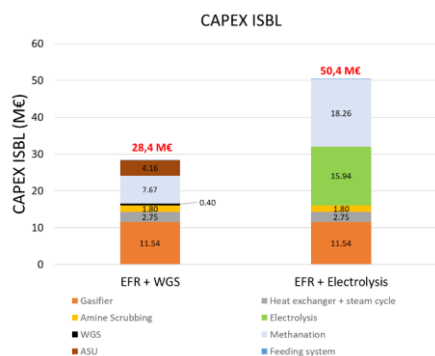


Figure 4: Direct investment for the two scenarios considered

Results showed that the pathway with the electrolyser is almost twice as expensive as the EFR + WGS. Two main reasons can be found; firstly because the electrolyser, for the power considered (34 MW) is expensive (16 M€), and secondly as the methanation reactor has to be scaled 2.5 times bigger as 2.5 times more CO gets converted to CH₄ in this scenario. Finally, indirect costs (hazards, engineering, contracting...) have been estimated at 67 % of CAPEX ISBL, which leads to a grand total investment of 48.3 M€ for the EFR+WGS case and 85.7 M€ for the EFR + Electrolysis scenario.

The total investment added to the different OPEX made it possible to estimate a cost of production for the methane produced (LCO_{CH₄} for Levelised Cost Of CH₄). This calculation was carried out considering a plant with a lifespan of 30 years, for an annual operation time of 8000 h and considering a discount rate of 8 %. In this study, the prepared tyre crumb costs 300 €/t and the electricity from the French grid costs on average 60 €/MWh. The cost breakdown on the LCO_{CH₄} for the different scenarios are shown in Figure 5.

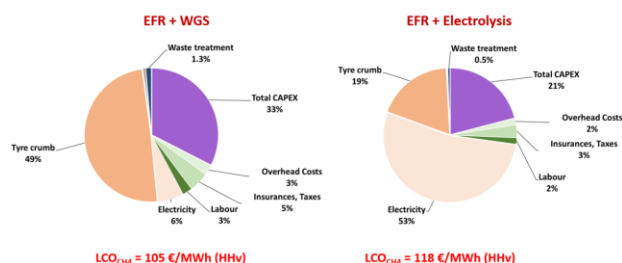


Figure 5: Levelised cost of CH₄; cost breakdown for the different scenarios considered

For the conditions considered, the EFR + WGS scenario allows to produce synthetic methane for

13 € less per MWh. The EFR + WGS case is strongly impacted by the price of the inputted tyre crumb (49 % of LCO_{CH₄}), whilst for the case of the EFR + electrolysis, the consumed electricity has the greatest impact (53 % of LCO_{CH₄}).

4 Conclusion and prospects

A new potential industrial pathway to valorise rubber from worn tyres into synthetic methane was studied comparing two main scenarios: EFR + WGS and EFR + electrolysis. The study revealed that the EFR + WGS process would more feasible economically (105 €/MWh instead of 118 €/MWh). However, the CO₂/kWh emission of the synthetic methane produced is high, hence the interest in maximizing the conversion of carbon into gas using the scenario with an electrolyser. The latter scenario however relies on inexpensive (≤60 €/MWh) and low-carbon (≤60 gCO₂/kWh) electricity for the process to become economically viable and environmentally beneficial compared to fossil gas.

With the current fluctuations in the gas prices and the willingness from many countries to increase their energy independence, the conversion of tyre crumb into synthetic methane could find its place in the energy market, determining the operational and economical conditions to make it viable are still to be sought.

Building on results from this first study, the work carried out at the CEA will continue and focus on ways to further decarbonise the synthetic methane by implementing CO₂ methanation and the use of local renewable electricity in the scenarios involving electrolysis. Investigations on tire gasification are also still needed and ongoing, especially focusing on the design of EFR adapted to tyre crumb and on ash management.

5 References

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6 Acknowledgments

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