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# Evaluation on the Performance of a Mobile Biogas Upgrading Plant

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Limiting the global warming to less than 2 °C is a major challenge for which a sharp increase of the renewable and sustainable energy production including fuels for transportation and agriculture is crucial. In these regards, biogas obtained from organic waste like agricultural residuals can be one promising solution. In order to use biogas as vehicle fuel, $CO_2$ and other hazardous impurities must be removed in the so-called upgrading process. At present, only a small share of European biogas plants and nearly none in Developing and Emerging Countries use those upgrading plants due to high investment costs. Mobile biogas upgrading plants traveling from one plant to the next for upgrading the biogas on site can reduce these investment costs for single farmers and thus, make it feasible for them. Therefore, a novel concept of a membrane-based upgrading process adapted to the restrictions of a mobile plant was developed and evaluated in Aspen Plus®. Although placing the whole upgrading plant on a truck led to strong restrictions in process design and thus, limited achievable product quality, $CH_4$ purity above 96 – 97 mol%, a common limit in national standards, was achieved.

## 1. Introduction

In 2015, a maximum temperature increase of below 2 °C was agreed within the Paris Agreement (United Nations, 2015). Therefore, a sharp increase of renewable and sustainable energy production is mandatory, including fluctuating energy sources as well as constant energy supplier and suitable energy storage systems (Kirchbacher et al., 2016). Especially, a strong increase of renewable and sustainable fuels (Liemberger et al., 2016) for transportation and agriculture will be crucial to fulfill this goal. Therefore, biogas obtained from organic waste can be one promising solution. Emerging and Developing Countries can benefit strongly from processing organic waste, which is usually rotting uncontrolled in these countries and, thereby, emitting the greenhouse gas $CH_4$. Using agricultural waste for biogas production sharply reduces the climate impact of agriculture by decreasing the amount of uncontrolled rotting waste as well as by supplementing fossil fuels needed at the farm with their own biogas. Thereby, also farmers themselves profit due to reduced fuel costs.
Raw biogas cannot be used directly as vehicle fuel due to its huge amount of adverse or hazardous impurities, like $CO_2$, $H_2S$ or $NH_3$. Especially, enhancing the heating value by reducing the incombustible $CO_2$, which accounts for up to 50 % of the raw biogas, is important. By removing $CO_2$ as well as these other impurities in the so-called upgrading process a gas comparable to natural gas is produced.
Presently, most European biogas plants use their biogas in CHP plants. Due to expiring subsidies for thereby produced electricity, new distribution pathways must be found. One possibility is upgrading biogas to high quality biomethane for substituting natural gas, which is presently done by less than 3 % of all European biogas plants (Deremince and Königsberger, 2017). In Developing and Emerging Countries, upgrading plants are even rarer, due to high investment costs. Usually, biogas in these countries is produced by small farmers using their agricultural residuals. Thus, making upgrading plants affordable to them by reducing investment costs drastically is the only way to increase the share of biogas plants upgrading their raw biogas to $CH_4$ in these countries.
In this work, a novel concept for a mobile biogas upgrading plant was developed. Until yet no concepts for biogas upgrading plants transported and operated on a truck have been investigated. Such a mobile plant can travel from one biogas plant to the next for upgrading biogas to biomethane directly on site. Investment costs for the system are reduced drastically for single farmers and thus, are easier affordable to them. This solution can be also a promising way for small European biogas producers, as stationary systems are also often too

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expensive for them. Design of the novel upgrading plant was investigated by process simulation in Aspen Plus®. Therefore, an experimentally validated membrane model developed at TU Wien was used (Makaruk and Harasek, 2009).

## 2. Materials and methods

### 2.1 Data collection

Focus of the work was led on biogas produced by small farmers in Developing and Emerging Countries and thus, Paraná in Brazil was chosen as model region as it shows various characteristics typical for these countries: in Paraná agriculture, with a big share of livestock breeding, is far the biggest industry. Thus, manure is often the major or only substrate for biogas plants resulting in high contents of $H_2S$ in the raw biogas. Further, biogas plants are mainly simple fixed-dome digesters and thus, biogas production rate and raw biogas composition are strongly dependent on surrounding conditions. Additionally, they are very sensible to solids due to the lack of stirring systems and thus, internal $H_2S$ removal by precipitation is not suitable.
In order to develop a system suitable for these conditions, raw biogas quality of existing biogas plant was investigated. Data were provided by CIBiogás, which is responsible for sample collection at the biogas plants in Paraná. Table 1 shows the broad range of typical biogas compositions in Brazilian biogas plants. These differences in the composition occur not only between different plants but also over time at the same plant. In the further investigation $O_2$ in the raw biogas was not taken into consideration as the high $O_2$ contents reported could be led back to poor sample collection. Improved sample collection led to a significantly reduced $O_2$ content, while other biogas components remained in the reported range.

*Table 1: Typical raw biogas compositions of Brazilian biogas plants and needed Biomethane quality according to Brazilian and Austrian standards. (ÖVGW, 2001, 2006; ABiogás, 2015)*

| Gas component | Unit | Brazilian raw biogas | Brazilian biomethane standard | ÖNORM G 31 and G 33 |
|---|---|---|---|---|
| $CH_4$ | mol% | 47 – 70 | ≥ 96.5 | ≥ 97.0 |
| $CO_2$ | mol% | 28 – 52 | ≤ 3 | < 2.0 |
| $O_2$ | mol% | < 1 | ≤ 0.8 | < 0.5 |
| $H_2S$ | $ppm_{mol}$ | 10 – 4,000 | ≤ 6.7 | < 3.3 |
| $H_2$ | $ppm_{mol}$ | 0 – 2,000 | - | < 4,000 |
| $H_2O$ | - | saturated at ambient temperature | max. -45 °C at 1 bar $H_2O$ dew point | max. 8 °C at 40 bar $H_2O$ dew point |

### 2.2 Process development

Today, several stationary upgrading systems are available, whereby, $CO_2$ separation is the core element of the upgrading process due to the high amount of $CO_2$ to be removed. Therefore, mainly PSA (Pressure Swing Adsorption), scrubbers and membrane separation are used in Europe. Until yet no mobile upgrading plant has been developed and thus, stationary concepts must be adapted to the requirements of mobile ones. Especially, strong restrictions regarding space and weight led to challenges in the process design. Capability of dealing with changes in feed quality and quantity, short start-up time, low energy demand and a design with low costs were other important criteria taken into consideration. In order to operate the mobile plant also by personal with lower educational levels easy operation mode with no need of chemicals is important. As membrane separation is the only process among currently used upgrading processes showing low space and weight demand, membrane separation was chosen for $CO_2$ removal (Beil and Beyrich, 2013). Further, membranes show quite low costs at small scale and meet the other parameters mentioned above in a satisfying way (Miltner, 2017).
Membrane separation was designed as a two-stage step using polyimide membranes with high permeability for $CO_2$ and low one for $CH_4$. Thereby, enriched $CH_4$ remains on the pressurized side of the membrane and thus, is already precompressed for high-pressure compression. Due to high $CH_4$ content in the permeate of stage 2 PERM2, this stream is recycled and added to the raw biogas stream. Depending on the $CH_4$ content in the $CO_2$-rich permeate of stage 1 (off-gas) a lean gas burner can be included to avoid emitting $CH_4$ into the atmosphere. Final process design of the mobile upgrading system, including two piston compressors and pre-treatment steps for removing especially water and $H_2S$, is shown in Figure 1. First compressor compresses the raw biogas to operational pressure needed for membrane separation, while the second one compresses the product gas to 210 $bar_a$ for storing the biomethane in gas cylinders. Water is removed by condensation in a cool-drying step by cooling the gas down to 7 °C. In this step also potential $NH_3$ with high solubility in water is removed from the gas stream. $H_2S$ is removed in a two-stage adsorption comprising of iron oxide as adsorbent followed by activated carbon. Thereby, also other potential impurities like siloxanes or $O_2$ can be removed. Due to the high

amount of $H_2S$ in most raw biogases in Brazil, stationary pre-desulphurization reducing $H_2S$ to a maximum of 100 $ppm_{mol}$ is crucial. Otherwise, very short intervals for exchanging the adsorbents have to be implemented, which is unsuitable for a mobile process and quite expensive due to the high amount of fresh adsorbents.

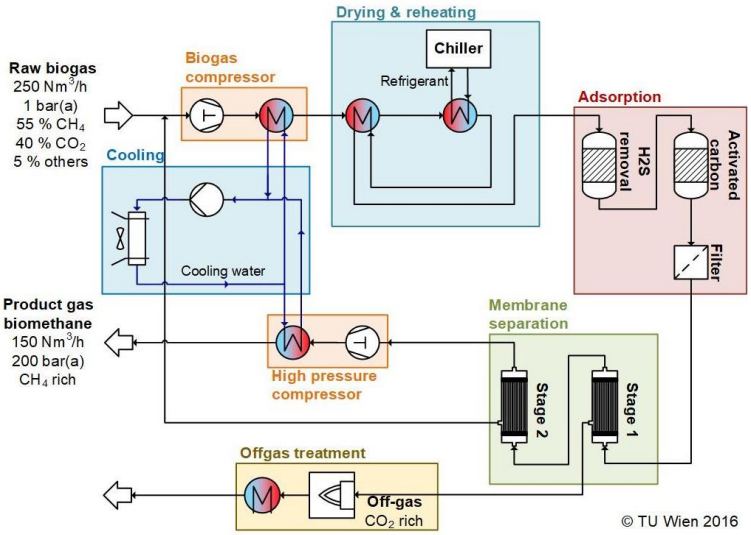


*Figure 1: Process design of the mobile membrane-based upgrading process.*

### 2.3 Process simulation

Whole upgrading process with focus on membrane separation, but also including pre- and post-treatment, was simulated in Aspen Plus®. For simplicity reasons, off-gas treatment was not included and adsorption step was implemented as simple split. A discretized, one-dimensional cell model based on the solution-diffusion model was used for the membrane separation step. This model was developed at TU Wien and validated in various applications, e.g. $CO_2/H_2$ separation (Rodrigues et al., 2010), $H_2$ purification (Lassmann, 2016) and biogas upgrading (Makaruk, 2009). In the solution-diffusion model, permeants dissolve in the membrane material and diffuse through the membrane along a concentration gradient. It is assumed that the fluids on both sides of the membrane are in equilibrium with the membrane interface and that the pressure applied across a dense membrane is constant. Separation is achieved due to specific diffusion rates and solubility of each permeant in the membrane material. $J_i$, the transmembrane flux of component $i$ is given by the equation

$$J_i = \frac{P_i(p_{i,H} - p_{i,L})}{\delta} \quad (1)$$

where $P_i$ is the permeability describing the ability of a membrane material to permeate component $i$, $\delta$ is the membrane thickness, $p_{i,H}$ is the partial pressure of component $i$ on the high pressure side and $p_{i,L}$ the partial pressure of component $i$ on the low pressure side.

*Table 2: Conditions and data used for modelling the mobile biogas upgrading system.*

| Parameter | Unit | Value |
|---|---|---|
| Biogas flow | $m^3_{stp}/h$ | 250 |
| $CH_4$ content in product gas | mol% | ≥ 96.5 |
| $CH_4$ content in the off-gas (permeate stage 1) | mol% | ≤ 10 |
| Raw biogas pressure | $bar_a$ | 1 |
| Product pressure | $bar_a$ | 210 |
| Compression efficiency (low- and high-pressure compressors) | % | 80 |
| Compressor mechanical efficiency (low- and high-pressure compressors) | % | 80 |

Process simulation was carried out to evaluate the influence of the raw biogas composition on the achievable biomethane quality and composition of the $CO_2$-rich permeate of the first stage PERM1 led into the atmosphere as so-called off-gas and to evaluate the energy demand for the process. Conditions and data used for modelling the mobile upgrading system can be found in Table 2. According to Table 1, three typical raw biogas compositions were chosen for process simulation (Table 3). Optimum ratio of membrane area in the first to the

area in the second stage was evaluated by investigating a ratio of 1:3 (Case 1), 1:4 (Case 2) and 1:5 (Case 3) while keeping the overall membrane area fixed. Additionally, two levels of operational pressure – 9 and 10 $bar_a$ – were investigated. Energy available as fuel was calculated, as the total energy content in the raw biogas is reduced due to $CH_4$ contained in the off-gas, but also due to the energy demand of the upgrading system.

*Table 3: Raw biogas compositions used for process simulation.*

| Biogas component | Unit | $CH_4$ low | $CH_4$ medium | $CH_4$ high |
|---|---|---|---|---|
| $CH_4$ | mol% | 48.07 | 57.97 | 68.68 |
| $CO_2$ | mol% | 48.07 | 37.99 | 27.47 |
| $H_2O$ | mol% | 3.85 | 3.89 | 3.85 |
| $H_2S$ | $ppm_{mol}$ | 89.7 | 358.6 | 11.4 |
| $H_2$ | $ppm_{mol}$ | 9.5 | 1075.8 | 0 |

## 3. Results and discussion

Achieved biomethane purity ranged from 96.20 to 98.05 mol% at 9 $bar_a$ operational pressure and from 97.88 to 99.16 mol% at 10 $bar_a$. Calculated methane recovery ranged from 93.90 % to 97.17 % at 9 $bar_a$ and from 92.69 % to 96.41 % at 10 $bar_a$. Figure 2 and Figure 3A show that the biomethane recovery goes down with increasing biomethane purity in the product stream due to increasing share of $CH_4$ in PERM1. $CH_4$ recovery considers only $CH_4$ losses due to the off-gas, as energy needed for operation can be delivered by various sources. Figure 3B shows the efficiency of the process expressed as ratio of finally available energy content in the product to the original energy content in the raw biogas. In contrast to $CH_4$ recovery, calculation of the process efficiency included also the energy needed for operation.

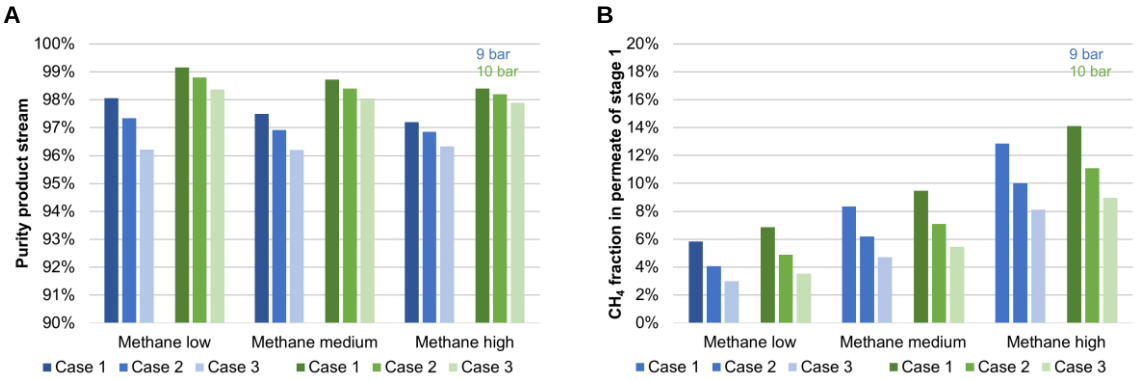


*Figure 2: (A) Biomethane purity (mol%) in product and (B) methane fraction (mol%) in PERM1 for three raw biogas compositions at 9 and 10 $bar_a$ for membrane area ratios of 1:3 (Case1), 1:4 (Case2) and 1:5 (Case3).*

Designs with a membrane area ratio of 1:3 and 1:4 were able to upgrade raw biogas with a broad range of $CH_4$ content (48 – 68 mol%) to a purity above the Brazilian biomethane limit of 96.5 mol% at both pressure levels. Same applies for the design with a membrane area ratio of 1:5 at 10 $bar_a$; at 9 $bar_a$ purity remained slightly below the limit with 96.3 mol% $CH_4$ in the product stream. Austrian standards were fulfilled for all designs at 10 $bar_a$, while at 9 $bar_a$ only a membrane area ratio of 1:3 was sufficient, as Figure 2A shows.
Purity increased with decreasing membrane area ratio, as a high membrane area in the first stage removed most of the $CO_2$ in the raw biogas and thus, smaller membrane area in stage 2 was sufficient to achieve a high biomethane purity. Membrane area ratio of 1:3 at 10 $bar_a$ operational pressure led to a purity of 99.16 mol% for a raw biogas with low $CH_4$ content. Unfortunately, high membrane area in stage 1 also led to low recoveries, as the share of $CH_4$ in PERM1 increased with reduced membrane area ratio (Figure 2B) – for raw biogas with high $CH_4$ content membrane area ratio of 1:3 led to 14,12 mol% $CH_4$ in PERM1 at 10 $bar_a$. In comparison, membrane area ratio of 1:5 reduced $CH_4$ significantly due to the smaller membrane area in the first stage. Using this ratio at 9 $bar_a$ led to a minimum $CH_4$ content in PERM1 of only 2.98 mol% for a raw biogas with low $CH_4$ content. Thereby, also the amount of $CO_2$ removed in this stage went down and so, high purity could be achieved only by increasing the membrane area in the second stage. This led to a higher permeate stream with increased content of $CH_4$ in the second stage, but unlike PERM1, PERM2 is recirculated and thus, no $CH_4$ loss is obtained.

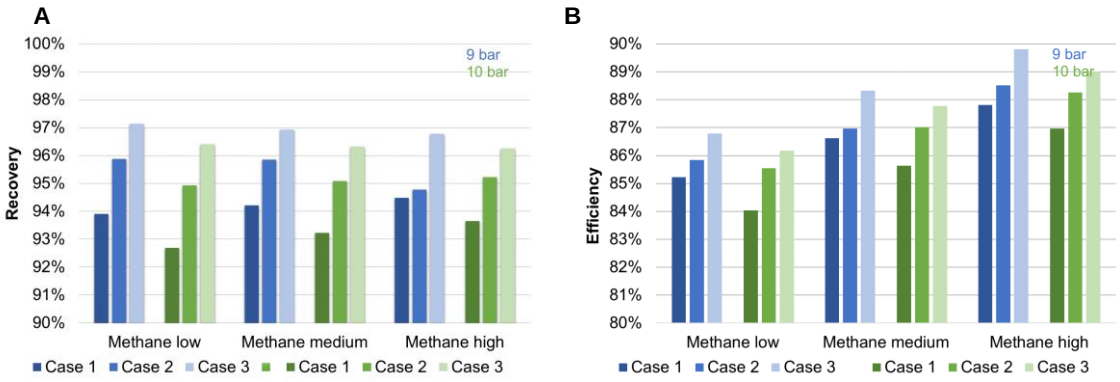


*Figure 3: (A) Methane recovery and (B) process efficiency for three raw biogas compositions at 9 and 10 $bar_a$ for the membrane area ratios of 1:3 (Case1), 1:4 (Case2) and 1:5 (Case3).*

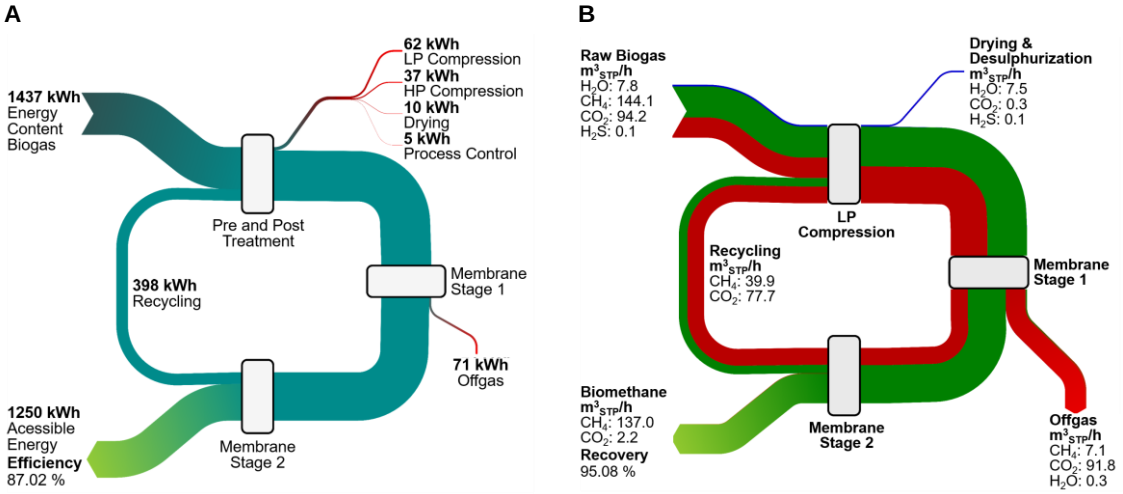


*Figure 4 (A) Energy and (B) mass balance for upgrading a raw biogas containing 58 mol% $CH_4$ with a two-stage membrane step (ratio of 1:4) at 10 $bar_a$.*

In summary, recovery was highest in designs with higher membrane area ratio (Figure 3 A), while the product purity was highest at a low membrane area ratio. Thus, a medium membrane area ratio of 1:4 at 10 $bar_a$ seems most fitting, as both the Brazilian and Austrian biomethane limit were met, while a $CH_4$ recovery of approximately 95 % was achieved. If used only in Brazil, also a lower pressure level of 9 $bar_a$ is sufficient. Still, achieved recoveries and purities remain below values produced at stationary plants (Miltner et al., 2017), as strong space restrictions limited the membrane separation to a two-stage design at maximum, as multi-stage design would need significantly more space. Due to the higher $CH_4$ content in PERM1 in comparison to stationary plants, implementation of a lean-gas burner is suggested to avoid $CH_4$ emission into the atmosphere.

Total efficiency of the process increased with increasing $CH_4$ content in the raw biogas and higher membrane area ratio, while it decreased with increasing operational pressure. Figure 3B shows that the $CH_4$ content in the raw biogas had the biggest influence on the achievable efficiency due to the reduced flow of PERM2. As this stream is recycled, lower flow led to lower amount of gas needed to be compressed to operational pressure and thus, lower energy demand of the low-pressure compressor. Also, higher membrane area ratio increased the overall efficiency, although it increased the flow of PERM2. Here, higher efficiency was caused by strongly reduced $CH_4$ content in the non-recycled PERM1 stream. Pressure level influenced the total efficiency only slightly by decreasing the $CH_4$ content in PERM1 when decreasing the operational pressure.

Energy and mass balances including energy needed for the operation of the plant were calculated for all cases at both pressure levels. Figure 4A and B show exemplary energy and mass balance for upgrading a raw biogas containing 58 mol% $CH_4$ by using a membrane separation step with a membrane area ratio of 1:4 at 10 $bar_a$.

In total, 84 – 90 % of the original energy content in the raw biogas was finally accessible as vehicle fuel. Highest efficiency of 89.81 % was achieved by using a membrane separation step with a membrane area ratio of 1:5 at 9 $bar_a$ for upgrading a biogas with high $CH_4$ content, while lowest efficiency of only 84.04 % was achieved for a biogas with low $CH_4$ content upgraded with a membrane area ratio of 1:3 at 10 $bar_a$. Pre- and post-processing, including compression steps, heat exchanger and process control, required 6.97 % and 8.67 % of the energy content in the raw biogas for the cases with highest and lowest efficiency, respectively. Biggest energy demand resulted from compressing the gas to operational pressure due to the high amount of gas that had to be compressed in this step, followed by the high-pressure compressor. With increasing $CH_4$ content in the raw biogas, energy needed for compressing the raw biogas went down, while the high-pressure compressor needed more energy as more product gas was attained. Same trend was obtained for decreasing operational pressure, as less energy was needed for reaching 9 $bar_a$ operational pressure and more energy for high-pressure compression. This was caused not only by the reduced inlet pressure but also by reduced membrane separation efficiency leading to higher amount of product gas with lower quality. Additionally, as described above, final accessible energy was reduced in the membrane separation step due to the $CH_4$ fraction in PERM1 (Figure 2B), in the case shown in Figure 4 this $CH_4$ fraction in PERM1 reduced the final accessible energy by 3.74 %.

## 4. Conclusions

It was shown that the investigated upgrading system based on a two-stage membrane separation step was suitable to produce a high quality product gas with $CH_4$ purities exceeding the Brazilian and Austrian $CH_4$ limits for biomethane. Thus, it can be used directly as substitute in natural gas-driven cars without any adaptions of the car. Additionally, also injection into the natural gas grid would be possible. Best performance was achieved with a membrane area ratio of 1:4 (stage 1 : stage 2) at 10 $bar_a$ operating pressure. This design provided a good balance between high biomethane purity and high recovery with > 98 mol% and about 95 %, respectively, and high amount of finally accessible energy of up to 89.81 % of the original energy content in the raw biogas.

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