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SYNTHESIS GAS PRODUCTION BY THE CO-GASIFICATION OF RDF-BASED PYROLYSIS CHAR AND TAR

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Abstract

We aimed to study the possibilities of synthesis gas production using char and tar originating from a Refuse-Derived Fuel (RDF) pyrolysis pilot plant. The main product of the thermal waste treatment plant is pyrolysis gas, while currently, the char is not utilized. During the pyrolysis process, a large amount of tar is also generated. Our goal was to find a more ecologically friendly alternative than landfilling in the case of both by-products. Steam gasification experiments were conducted at 900 °C in a small tubular reactor. The carbon conversion of the char-tar blend reached 95.3%.

Keywords: *pyrolysis, gasification, RDF, syngas*

1. Introduction

The global solid waste – including municipal solid waste (MSW) – generation exceeds 10 billion tons annually and by 2025, it may reach 14.8 billion based on the statistics (Hasan et al., 2021). The environmentally friendly and economically feasible management of this large amount of waste requires the continuous development of waste management systems. The Waste Framework Directive (2008/98/EC) of the European Union incorporates the basic principles of the ideal waste management and establishes a priority order among the different waste management methods (**Figure 1**).

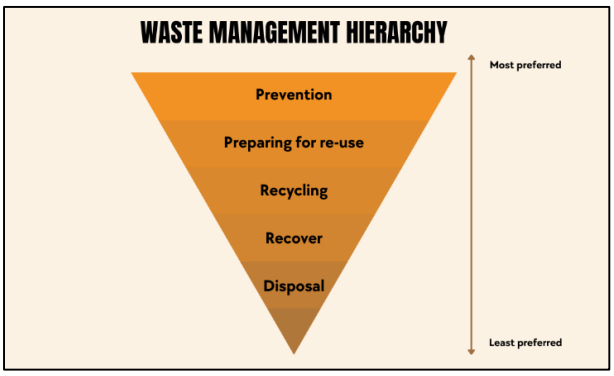

Figure 1. *Waste management hierarchy*

According to this, landfilling is considered the least preferred way to treat waste. However, in Hungary landfilling is still the leading method in the case of municipal solid waste (KSH). Since recycling has limitations (finite cycles, contaminated material), energy recovery is crucial in preventing waste from ending in landfills. Among the different Waste-to-Energy technologies, combustion is still the leading and most developed process. However, other thermochemical methods, like pyrolysis and gasification, have become more common in the recent decades. These technologies are not only able to reduce the volume of waste going to landfills but provide the opportunity to produce value-added products (e.g., syngas, activated carbon) (Gopu et al., 2018; He et al., 2010).

Recently, a newly developed mechanical-physical processing plant started operating in Zalaegerszeg (Hungary), where MSW is separated into Refuse Derived Fuel (RDF), organic fraction, and recyclable materials (Faitli et al., 2018). The separation of recyclables from residual municipal solid waste (RMSW) supports the waste hierarchy and in terms of energy recovery also beneficial, because RDF – or with standardized term: Solid Recovered Fuel (SRF) – represents a high-quality fuel compared to unsorted MSW (Directive 2008/98/EC; Szentannai and Szücs, 2018). Furthermore, using RDF instead of MSW can reduce harmful emissions and improve the efficiency of the technology. In this study, a pyrolysis char and tar blend gasification was investigated. The samples were generated in an RDF pyrolysis pilot plant in Zalaegerszeg. The char produced generally has high carbon content (~38-44 wt.%) and can be further utilized to generate syngas through gasification (Sebe et al., 2021). Without utilization, the tar is a hazardous waste (Abdel-Shafy and Mansour, 2016), but blended with the char can be suitable for synthesis gas production. The characteristics of the syngas are strongly affected by the gasification agent. In this study, steam was used, because it has many advantages over other agents; e.g., it allows syngas production with low tar and high H_2 content (Yang and Chen, 2015).

2. Materials and methods

2.1. Preparation of char and oil blends

The examined char and tar samples are the by-products of RDF pyrolysis. The RDF was produced using non-selectively collected RMSW in a mechanical-physical processing plant. **Figure 2**. shows the mechanical-physical waste processing technology.

The key step of this process is the separation of PVC from the waste stream using NIR (near infrared) separators; thereby, it is possible to produce RDF with low chlorine content. The chlorine content is one of the most important characteristics in terms of the quality of the waste fuel as it can cause corrosion and harmful emissions during thermal utilization (Ma et al., 2020). After processing the waste, the produced RDF was pelletized.

A pilot-scale auger pyrolysis reactor with 20-60 kg/h capacity was used to produce mainly syngas, but a considerable amount of char and tar was also generated. The maximum temperature in the reactor during pyrolysis was 700 °C. Before the gasification experiments, char samples were ground in a mortar. In this study, char was blended with 25 wt.% tar. **Table 1**. shows the results of the elemental analysis of the feedstock material, which was measured with a Carlo Erba EA 1108 elemental analyzer.

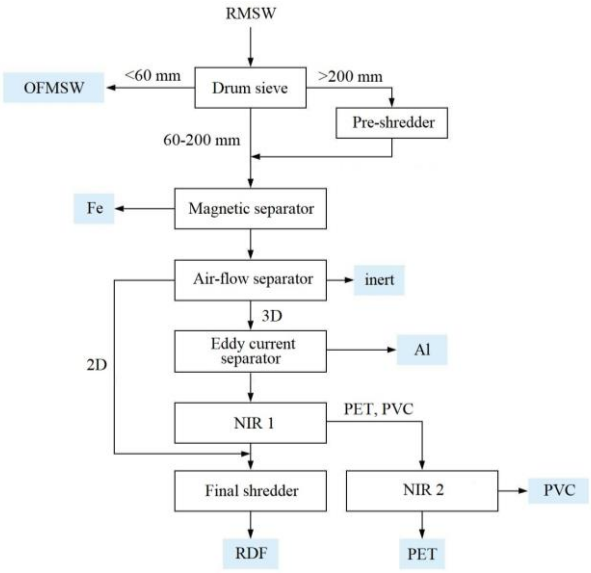

Figure 2. *Mechanical-physical RMSW processing technology (Faitli et al., 2018)*

Table 1. *Elemental analysis of the examined char-tar blend*

Parameter	Mean values wt.%
Carbon	44.29
Hydrogen	1.52
Nitrogen	0.80
Sulfur	0.09

2.2. Experimental apparatus

The experimental work was carried out using a laboratory-scale reactor (***Figure 3***.), which was operated at atmospheric pressure. The reactor consisted of a horizontal stainless-steel tube with an outer diameter of 30 mm. The tube was placed inside 2 electrically heated furnaces, and the surface temperature was measured using K-type thermocouples.

Approximately 30 g of char-tar blend was placed inside the reactor tube for each experiment. The sample was heated from ambient temperature to 900 °C at a heating rate of 10 °C/min under N_2 atmosphere. Once the target temperature was achieved, water was introduced into the reactor with a flow rate of 10 mL/h. The water flow rate was controlled using a Spritzenpumpe Medfusion 2010-type infusion pump connected to the reactor tube. When the water reached the high-temperature zone of the reactor and evaporated, the generated steam reacted with the char-tar sample. The gasification process lasted for 3.5 hours until the produced syngas flow rate fell below the lower detection limit of the variable-area flow meter. The experiment was performed three times and the mean value is reported in this work. The flow rate of the syngas was measured by a Medingen-type rotameter with a 3-30 L/h measuring range.

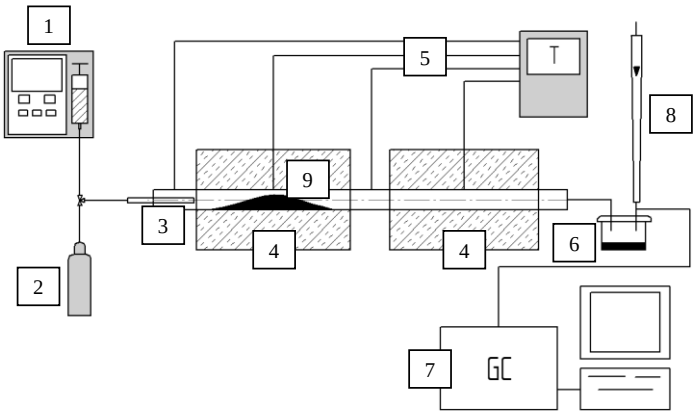

Figure 3. *Laboratory-scale gasification system (1. Infusion pump, 2. N_2 cylinder, 3. Stainless steel tube, 4. Electric furnaces, 5. K-type thermocouples, 6. The liquid collecting vessel, 7. Agilent 490 Micro GC gas chromatograph, 8. Rotameter, 9. Sample)*

The flow rate values were corrected using the following formula (Rollmann):

$$q_G = q_A \sqrt{\frac{\rho_A}{\rho_G}} \tag{1}$$

where: q_G – corrected flow rate [L/h], q_A – measured flow rate [L/h], ρ_A – density of air on 20 °C [kg/m^3], ρ_G – density of the generated gas [kg/m^3].

The density of the generated gas was calculated by the following equation:

$$\rho_G = 0{,}01 \cdot \left(H_2[\%] \cdot \rho_{H_2} + CO[\%] \cdot \rho_{CO} + CO_2[\%] \cdot \rho_{CO_2} + \cdots\right), kg/m^3 \tag{2}$$

The specific flow rate was computed using the equation:

$$q_{G_{sp}} = (q_G \cdot 1000)/(60 \cdot m_k)\,, dm^3/kg \tag{3}$$

where m_k is the initial weight of the feedstock [g].

The gas composition (H_2, CO, CO_2, hydrocarbons C_1-C_3 and H_2S) was measured by an Agilent 490 Micro GC gas chromatograph with a TCD detector and two columns (PoraPLOT U, CP-CO_x). The lower heating values of the gases were calculated using the following formula based on the measured concentrations:

$$LHV_{gas} = \sum_{i=0}^{n} c_i \cdot LHV_{c_i} \tag{4}$$

Furthermore, carbon conversion was calculated as well using the equation:

$$\eta_C = 100 \cdot \left(1 - \frac{m_s \cdot c_{c,s}}{m_f \cdot c_{c,f}}\right) \tag{5}$$

where m_f and m_s are the weights of the feedstock material and the solid residue, while $c_{c,f}$ and $c_{c,s}$ are the initial and final carbon concentrations.

3. Results and discussion

Figure 4. shows the specific syngas yield and gas composition during the heating period. Based on the gas chromatography results, the pyrolysis started at a relatively low temperature. Although below 700 °C, the intensity of gas formation was minimal, and it did not reach the lower detection limit of the rotameter (3 L/h). Above 700 °C – the maximum temperature of the pilot-scale RDF pyrolysis reactor – the gas yield increased significantly and contained mostly H_2, CO_2, CO, and hydrocarbons in small quantities.

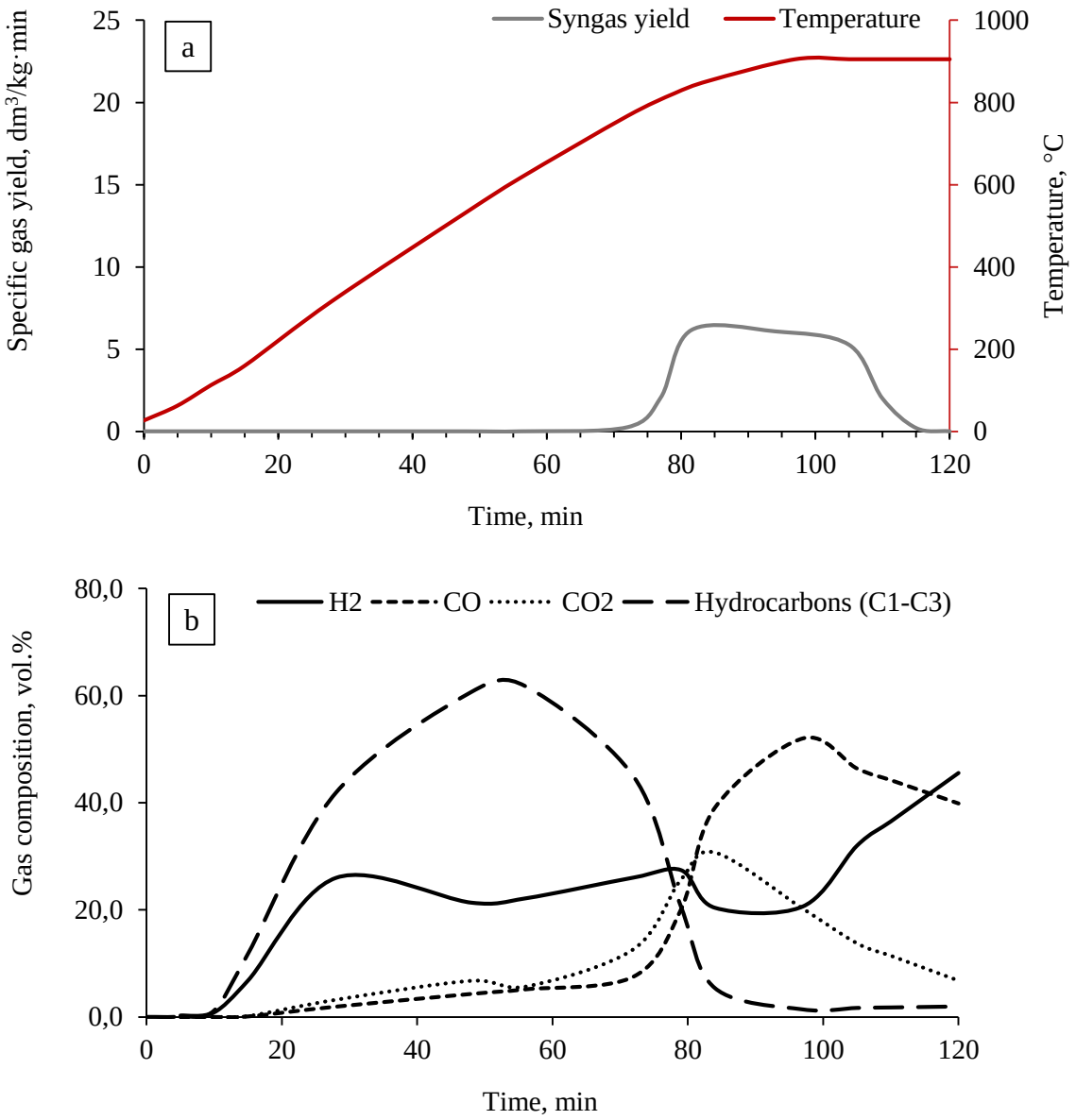

Figure 4. *Syngas yield (a) and composition (b) during pyrolysis.*

In the gasification stage (**Figure 5**.), gas production was more intensive than during the pyrolysis stage, due to the material's low volatile and high carbon content. The H_2 is one of the main components of the synthesis gas. The main component of the produced gas was H_2, and its concentration showed an increasing tendency until the end of the experiment. In contrast, the CO concentration continuously decreased in the syngas. Furthermore, 5-20 vol.% CO_2 and <2 vol.% hydrocarbons (C_1-C_3) were also detected. Although methanation ($2\ C + 2\ H_2O \leftrightarrow CH_4 + CO_2$) is one of the main reactions of gasification, methane formation favors higher pressure (Nagy and Dobó, 2020; Nanou, 2013).

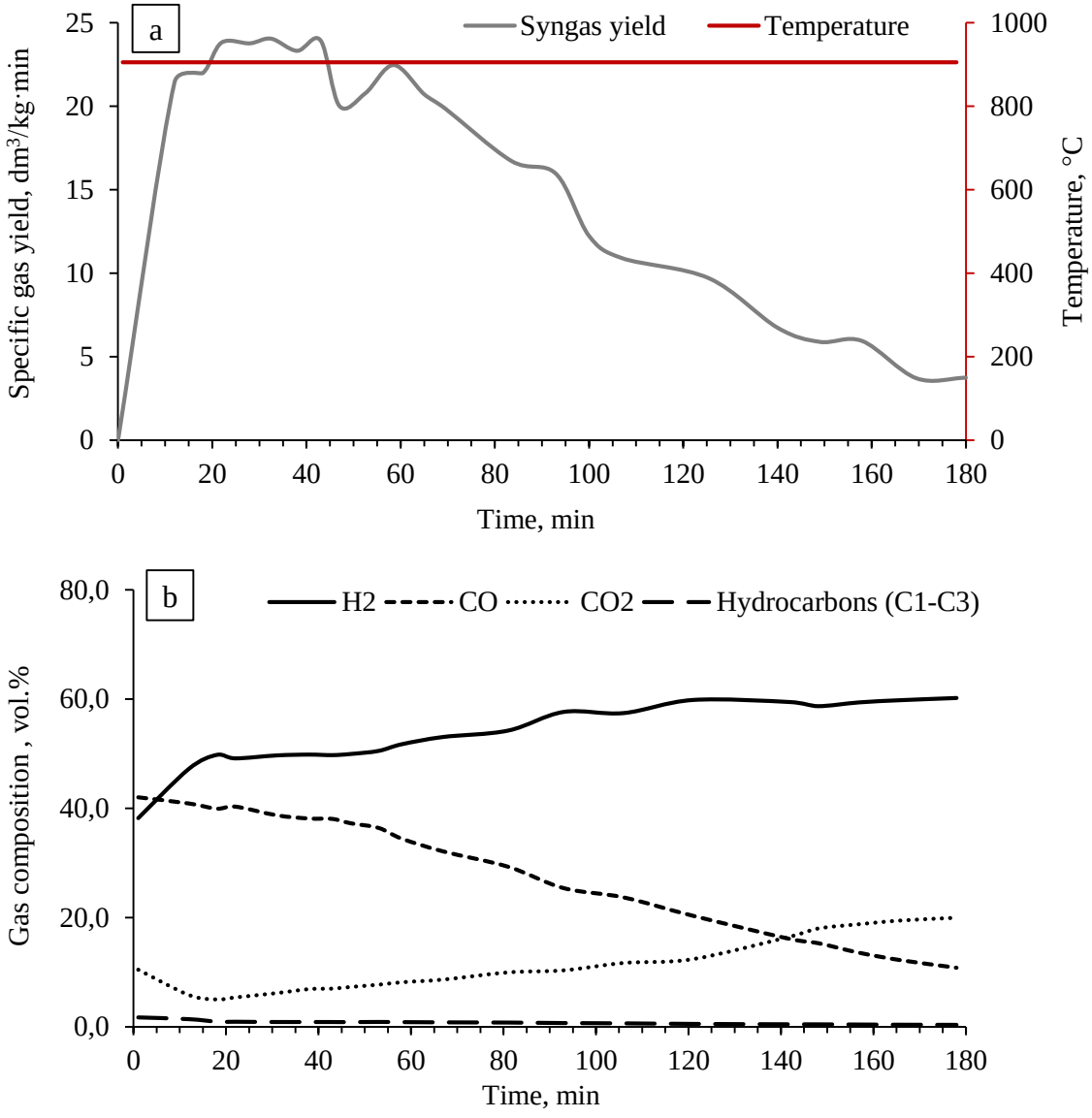

Figure 5. *Syngas yield (a) and composition (b) during steam gasification*

The H_2S concentration in the syngas was also determined. At the pyrolysis stage, it was only detected before the intensification of gas formation and probably originated from the volatile compounds of the liquid phase. H_2S was found in the syngas from the beginning of gasification, and its concentration continuously increased to a final 0,67 vol.%.

Figure 6 shows the H_2/CO ratio and LHV of the syngas during pyrolysis, gasification, and the whole experiment. Regarding chemical utilization, H_2/CO ratio is an essential characteristic of syngas. The optimal H_2/CO depends on the targeted product (e.g., Fischer-Tropsch fuels: ~2, methanol: ~2, synthetic natural gas: ~3) (Litvinenko and Meyer, 2018). This ratio was lower during pyrolysis, but the syngas yield also was significantly lower in this stage. Therefore, it just slightly affected the characteristics of the total syngas composition.

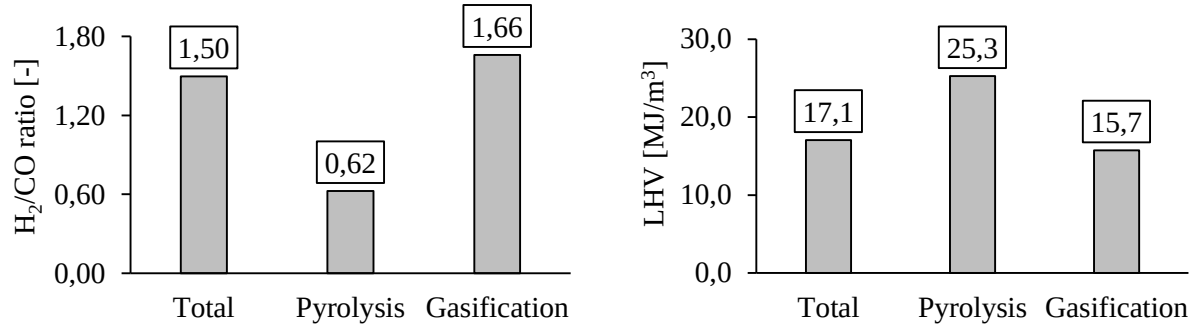

Figure 6. *H_2/CO (left) ratio and LHV (right) of the syngas*

From energetic point of view, LHV is a crucial parameter. In the pyrolysis stage, the formation of hydrocarbons is higher than in the case of gasification, so the LHV of the gas was higher at the beginning of the experiment.

Table 2 shows the carbon content and dry weight of the initial char-oil blend and the solid residue of the experiment, which were the basis of further calculations.

Table 2. *Data for carbon conversion calculation*

Material	Carbon content, wt.%	Weight, g
Feedstock	44,3	23,8
Residue	4,1	12,1

The weight of the char-tar blend dropped by 49,2% by the end of the experiment, and the carbon conversion reached 95,3%.

4. Summary

Managing the continuously increasing waste of humanity is challenging, and the high global landfilling rate is a great concern. According to the waste management hierarchy, non-recyclable materials should be considered for energy recovery to solve this problem. Gasification and pyrolysis provide an opportunity to reduce the volume of the MSW. At the same time, these technologies also allow the generation of valuable products for the chemical industry or the energy sector.

Due to the heterogeneity and variable composition of the feedstock, residual MSW-based thermal treatment technologies are challenging to control. However, MSW can be turned into waste-based fuels, so-called RDFs, which have several advantages over the original raw material, like higher HHV, lower moisture content, and with modern separation technologies, lower chlorine content as well. When RDF is used for pyrolysis gas generation, the remaining char generally has high carbon content, which makes it suitable for further gas production by gasification. The tar is also worth utilizing. Otherwise, it must be disposed because it contains several hazardous components.

In this study, our goal was to investigate the gasification behavior of RDF pyrolysis char and oil to find a way to reduce waste and produce syngas. The gasification was carried out in a laboratory-scale reactor. Approximately 30 g char-tar blend was placed into the reactor and heated from ambient temperature to 900 °C at a heating rate of 10 °C/min under N_2 atmosphere. Once the temperature reached 900 °C, 10 mL/h H_2O was inserted into the reactor.

During the heat-up (pyrolysis) period, the gas formation was minimal compared to the gasification. Throughout the experiment, H_2, CO, and CO_2 were the main gas components. Nonetheless, at the beginning of pyrolysis, hydrocarbon gases were present in significant concentration also, which caused a growth in the LHV of the syngas. The H_2/CO ratio increased during gasification. By the end of the experiment, the weight of the material was reduced by 49.2%, and the carbon conversion reached 95.3%.

Our further plan is to investigate the pyrolysis of RDF in a larger (2-3 kg/h) system and in-situ gasification of the pyrolysis products. Thereby our goal is to produce more syngas with less tar content and reduce the amount of the waste/by-products of the technology.

5. Acknowledgment

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