

TECHNO-ECONOMIC AND ENVIRONMENTAL ASSESSMENT OF HYDROGEN PRODUCTION BASED ON NATURAL GAS STEAM REFORMING PROCESS

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ABSTRACT. Hydrogen is foreseen as a promising energy vector for the future on the road to develop a low carbon economy. In this respect, developing energy-efficient improved hydrogen production systems with low CO₂ emissions is an important aspect. This study is evaluating the most relevant economic and environmental impact elements for hydrogen production based on catalytic natural gas steam reforming. As an illustrative example, an industrial-relevant hydrogen production plant with an output of 50000 Nm³/h was investigated. In addition, a pre-combustion carbon capture feature, based on reactive gas-liquid absorption using alkanolamines (Methyl-DiEthanol-Amine - MDEA), was considered to reduce the CO₂ emissions. The overall carbon capture rate was 70%. Similar natural gas reforming plant without decarbonization feature is also discussed to quantify the efficiency and economic penalties for CO₂ capture. As assessment methods, computational tools, thermal integration analysis and an in-depth techno-economic and environmental procedure were used. For instance, to quantify the overall environmental impact, Life Cycle Assessment (LCA) was used. Various relevant technical, economic and environmental indicators are calculated and discussed in the present work.

Keywords: *Hydrogen production; Natural gas steam reforming; Carbon capture by chemical scrubbing; Techno-economic and environmental assessment.*

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INTRODUCTION

Global warming and climate change are significant challenges of the current time. In an attempt to reduce greenhouse gas emission, low carbon technologies have to be developed and deployed in various industrial sectors [1]. A wide diversity of methods and technologies can be considered to reduce CO₂ emissions e.g., large-scale usage of renewable energy sources, carbon capture, utilization and storage (CCUS) options, improving energy conversion, developing new energy carriers with reduced environmental impact [2]. An important element of all new low carbon technologies should be a promising economic impact of the process compared to current state-of-the-art technologies.

Along these important lines, hydrogen is predicted to become an important energy carrier for the future having significant technical, economic and environmental benefits (e.g., high energy conversion efficiency, no greenhouse gas emission when used, lower environmental impact etc.) [3]. Therefore, new developments are expected for the whole hydrogen value chain (production - transport - storage - utilization). Currently, hydrogen is produced from fossil sources (natural gas, oil, coal) with large CO₂ emissions and it is mostly used in chemical and petro-chemical sectors as reactant (e.g., ammonia, various hydrogenation processes etc.) as presented in Figure 1 [4].

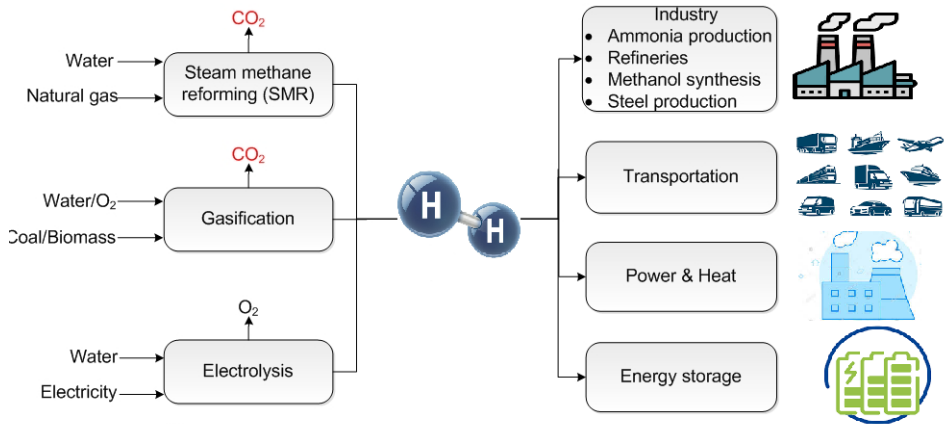
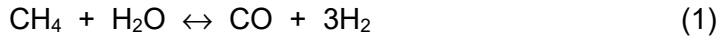


Figure 1. Main hydrogen production routes and applications

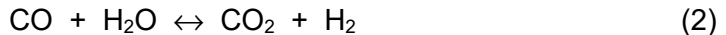
This work is aiming to combine the natural gas catalytic steam reforming process (the most used hydrogen production route) with carbon capture feature to significantly reduce the environmental impact of this process. As mentioned before, the new technological concepts must have

techno-economic and environmental advantages over current technologies. In this respect, this analysis will consider similar hydrogen production route without carbon capture as a benchmark case. As key novelty elements of this work, one can mention the usage of an integrated process modeling - thermal integration - techno-economic and environmental assessment methodology to propose improved hydrogen production systems with better performance indicators.

The hydrogen production based on catalytic steam reforming relays of the following reversible chemical reaction:



The temperature of the natural gas steam reforming reactor is high (around 800 - 900°C). In addition, the process is highly endothermic, accordingly heat has to be provided. Various configurations can be used, the most important ones are: fired-based reforming reactor (additional natural gas has to be combusted outside of the reactor) and autothermal reforming reactor (heat is generated in-situ using oxygen or air). This work considers the fired-based reforming reactor which has significant benefits than autothermal ones e.g., higher energy efficiency, lower plant complexity, better economics etc. [5-6]. The syngas is then catalytically converted with steam to increase the hydrogen yield and to concentrate the carbon species as CO₂ for subsequent capture. The water gas shift (WGS) reaction is the following:



As decarbonization technology, the chemical scrubbing using MDEA-based gas-liquid absorption process was considered. This carbon capture method is based on the following chemical reaction [7]:



The captured CO₂ is then fed to Carbon Processing Unit (CPU) for drying and compression (up to 120 bar) to satisfy the quality requirements of transport (through pipeline) and storage (e.g., saline aquifers, enhanced oil recovery, depleted oil and gas fields etc.) systems. After decarbonization, the hydrogen rich-gas is purified by Pressure Swing Adsorption (PSA) to the required purity (min. 99.95% vol. was considered in this analysis) and then compressed to 60 bar for transport. The tail gas is recycled back to the plant (as fuel to reforming reactor).

PROCESS CONFIGURATION AND MAIN DESIGN ASSUMPTIONS

The following hydrogen production systems based on natural gas catalytic reforming were investigated in term of main techno-economic and environmental performances:

Case 1: Natural gas steam reforming without carbon capture;

Case 2: Natural gas steam reforming with carbon capture.

The conceptual design of decarbonized hydrogen production route based on catalytic natural gas steam reforming process (Case 2) is presented in Figure 2. Several key technological modifications are present in comparison to the correspondent non-decarbonized design (Case 1): carbon capture unit based on chemical scrubbing, carbon processing unit for drying and compression of captured CO₂ stream.

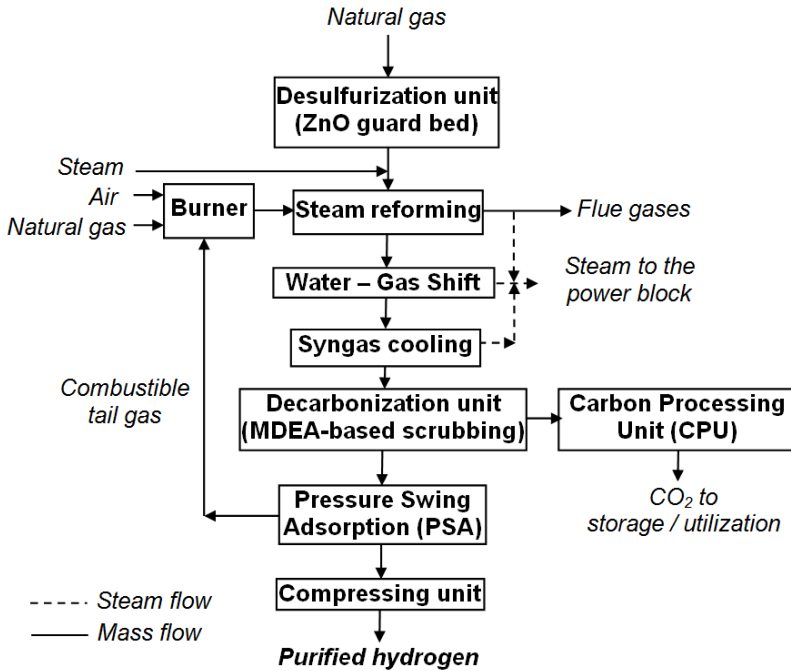


Figure 2. Decarbonized hydrogen production system based on natural gas steam reforming (Case 2)

The evaluated hydrogen production systems based on natural gas catalytic reforming were simulated using ChemCAD software. Table 1 presents the most important modeling assumptions [8-10].

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Table 1. Main design assumptions

Process unit	Design parameters
Plant capacity & purity	50000 Nm ³ /h (equivalent to 150 MW _{th}) 99.95% (vol.)
Natural gas characteristics	Composition (% vol.): 89% CH ₄ , 7% C ₂ H ₆ , 1% C ₃ H ₈ , 0.1% C ₄ H ₁₀ , 0.009% C ₅ H ₁₂ , 0.001% C ₆ H ₁₄ , 2% CO ₂ , 0.89% N ₂ Lower heating value (LHV): 46.73 MJ/kg Pressure: 30 bar
Desulfurization unit	ZnO guard bed Sulfur removal yield: >99%
Catalytic reformer unit	Outlet temperature: 900°C Gibbs reactor Thermal mode: heat exchanger Pressure drop: 1 bar
Water Gas Shift (WGS) unit	Outlet temperature: 400°C Reactor type: Equilibrium reactor Thermal mode: adiabatic Steam to CO ratio: 2 (molar) Pressure drop: 1 bar
Decarbonization unit Absorption column: Desorption column:	Solvent: Methyl-DiEthanol-Amine (MDEA) Solution concentration: 50 % wt. No. of stages: 20 Column pressure drop: 1 bar No. of stages: 15 Column pressure drop: 1 bar Solvent regeneration with LP steam Heat duty: 0.6 MJ/kg CO ₂
CO ₂ processing unit (CPU)	Drying agent: Tri-Ethylene-Glycol (TEG) 4 stages with inter-cooling Delivery pressure (at plant gate): 120 bar CO ₂ composition (vol. %): >95% CO ₂ , <2000 ppm CO, <250 ppm H ₂ O, <100 ppm H ₂ S, <4% other gases (N ₂ , Ar, H ₂)
Hydrogen compression unit	Delivery pressure: 60 bar Compressor efficiency: 80% Outlet temperature: 30-40°C
Heat recovery unit	Steam conditions: 480°C & 48 bar Steam turbine efficiency: 85% Condensing pressure: 48 mbar Cooling water temperature: 15°C
Heat exchangers	$\Delta T_{\min.} = 10^{\circ}\text{C}$; Pressure drop: 2 - 4% of inlet pressure

PROCESS ASSESSMENT METHODOLOGY

The evaluated cases were simulated using ChemCAD. The thermodynamic package (Soave-Redlich-Kwong - SRK) was selected considering the present chemical species as well as the operational parameters of the processes (e.g., pressure, temperature etc.). For the chemical scrubbing unit, the electrolyte package was used considering the ionic system present in aqueous solution (see reaction 3).

The simulation results were compared with experimental / industrial data for model validation [11-12]. No significant differences were observed. For instance, the methane conversion is about 98% comparable with values from industrial applications. Both hydrogen production designs were optimized in term of energy utilization using pinch method [13]. As an illustrative case, Figure 3 presents the hot and cold composite curves for decarbonized concept (Case 2).

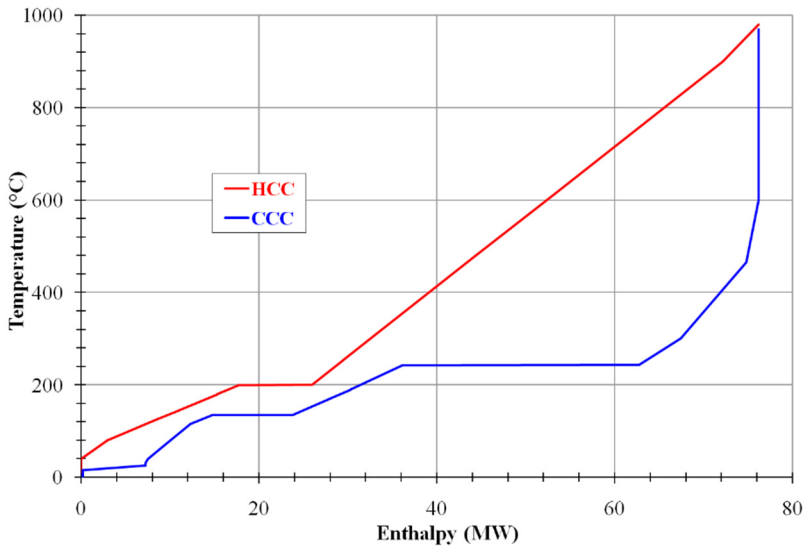


Figure 3. Composite curves for decarbonized hydrogen concept (Case 2)

One can noticed that the available hot streams within the plant (e.g., hot syngas from reforming reactor, shifted gas from WGS reactor, flue gases from reactor burner etc.) are covering the cold stream duties. Even further, an excess heat exists which it is then for steam generation. The generated steam is partly used for catalytic reformer (as reactant) and the rest is expanded in a steam turbine for power generation (to cover the ancillary power consumptions).

Following the thermal integration analysis, the overall mass and energy balances were then used for the quantification of the overall techno-economic and environmental performance indicators. The most important performance indexes are presented below:

- Hydrogen and power generation efficiencies ($\eta_{Hydrogen}$ and η_{power}) show the overall energy yield of the concepts:

$$\eta_{hydrogen} = \frac{\text{Hydrogen thermal output}}{\text{Natural gas thermal input}} * 100 \quad (4)$$

$$\eta_{power} = \frac{\text{Net power output}}{\text{Natural gas thermal input}} * 100 \quad (5)$$

- Plant decarbonization rate (R_{CO_2}) is quantified as the ratio of captured CO₂ molar flow to the input carbon molar flow of natural gas:

$$R_{CO_2} = \frac{\text{Captured CO}_2 \text{ molar flow}}{\text{Input carbon molar flow}} * 100 \quad (6)$$

- CO₂ emission factor (E_{CO_2}) is computed considering emitted CO₂ mass flow for each MW power plus hydrogen generated within the process:

$$E_{CO_2} = \frac{\text{Emitted CO}_2 \text{ mass flow}}{\text{Hydrogen output} + \text{Net power}} * 100 \quad (7)$$

- Capital cost of a specific plant sub-system (C_E) with a given capacity (Q) is calculated by scaling method using the formula which considers the base cost (C_B) corresponding to the base capacity (Q_B):

$$C_E = C_B * \left(\frac{Q}{Q_B}\right)^M \quad (8)$$

- Specific capital investment (SCI) is calculated as the ratio of total capital cost and the plant energy output (hydrogen and power):

$$SCI = \frac{\text{Total capital cost}}{\text{Hydrogen output} + \text{Net power}} \quad (9)$$

- Levelized cost of hydrogen (LCOH) is calculated as ratio of annualized capital and operational costs and the plant hydrogen output:

$$LCOH = \frac{\text{Annualized capital \& operational costs}}{\text{Hydrogen output} + \text{Net power}} \quad (10)$$

- CO₂ removal and avoided costs is calculated based on levelized cost of hydrogen and CO₂ emission factors in both non-decarbonized and decarbonized scenarios:

$$CO_2 \text{ removal cost} = \frac{LCOH_{\text{Capture}} - LCOH_{\text{No capture}}}{CO_2 \text{ removed}} \quad (11)$$

$$CO_2 \text{ avoided cost} = \frac{LCOH_{\text{Capture}} - LCOH_{\text{No capture}}}{E_{CO_2 \text{ No capture}} - E_{CO_2 \text{ Capture}}} \quad (12)$$

The main economic assumptions used in the evaluation of hydrogen production systems based on natural gas reforming with and without carbon capture are presented in Table 2 [14-16].

Table 2. Main economic assumptions

Natural gas cost, € / GJ	5.0
Boiler feed water (BFW) cost, € / t	0.10
Cooling water (CW) cost, € / t	0.01
CW treatment cost, € / m ³	0.0025
BFW treatment cost, € / month	45000
Personnel number	78
Direct labour cost, € / person / y	50000
Administrative costs, % of direct labour cost	30
Plant maintenance costs, % of CAPEX per year	2.25
Plant capacity factor, %	85.62
Internal rate of return, %	8
CO ₂ storage cost, € / t	7
Carbon emission tax, € / t	25
Construction period, years / annual CAPEX share, %	2 / 40, 60
Power plant operation life, years	25

In addition to the global plant environmental indicators (decarbonization rate and CO₂ emission factor), an in-depth Life Cycle Analysis (LCA) was performed for the evaluated cases. The LCA analysis considers not only the

plant but also up-stream (e.g., natural gas, catalysts, solvent chains) and down-stream processes (e.g., CO₂ transport and storage, solvent degradation and disposal). As indicators Global Warming Potential (GWP), Freshwater Eutrophication Potential (FEP), Ozone Depletion Potential (ODP), Fossil fuel Depletion Potential (FDP), Freshwater Ecotoxicity Potential (FETP), Human Toxicity Potential (HTP), Metal Depletion Potential (MDP), Photochemical Oxidant Formation Potential (POFP) and Terrestrial Ecotoxicity Potential (TETP) were considered according to the agreed LCA methodology. The LCA results are reported to 1 MW H₂ produced. The full description of the LCA methodology is presented in several different papers of the same authors [17].

RESULTS AND DISCUSSION

Both hydrogen production cases based on natural gas reforming with and without carbon capture were simulated using ChemCAD software. The mass and energy balances of the evaluated concepts were then used for calculation of main performance indicators. Table 3 presents the most important technical indicators.

Table 3. Key technical performance indicators

Performance indicator	UM	Case 1	Case 2
Natural gas flowrate	t/h	15.37	15.37
Natural gas LHV	MJ/kg	46.73	
Natural gas thermal energy	MW _{th}	203.63	203.63
Gross power output	MW _e	8.51	5.84
Hydrogen thermal output	MW _{th}	150.00	150.00
CO ₂ capture & compression	MW _e	0.00	2.12
Hydrogen compression	MW _e	2.09	2.09
Power island	MW _e	1.05	1.04
Ancillary power consumption	MW _e	3.14	5.25
Net power output	MW _e	5.37	0.59
Net power efficiency	%	2.63	0.28
Hydrogen thermal efficiency	%	73.66	73.66
Cumulative plant energy efficiency	%	76.29	73.94
Carbon capture rate	%	0.00	70.00
CO ₂ emissions factor	kg/MWh	267.45	82.50

As can be observed from Table 3, the introduction of pre-combustion carbon capture using a MDEA-based gas-liquid absorption unit involves an energy penalty of about 2.34 net percentage points. On the other hand, the CO₂ specific emission factor was significantly reduced by about 3.24 times (from 267.45 to 82.50 kg/MWh) corresponding to a plant decarbonization rate of 70%. The main reason that the decarbonization rate is not higher than 70% represents the unconverted methane and carbon monoxide in reformer and shift reactors [18]. These chemical compounds are not removed by the chemical scrubbing unit being then combusted in the burner to provide the heat duty required by the reforming reaction. For a higher carbon capture (90%), a post-combustion capture unit has to be used to capture CO₂ from flue gases coming from the reformer burner [19]. This work was not considered an additional post-combustion capture unit due to significant technical elements (increased heat consumption for solvent regeneration) as well as negative influence on economic costs.

Following the technical evaluation of both reforming processes, the next step was to assess the economic performance indicators (e.g., specific investment costs, operational & maintenance costs, levelized cost of hydrogen, CO₂ capture costs). The main economic indicators of hydrogen production concepts based on natural gas reforming with and without decarbonization are presented in Table 4.

Table 4. Key economic performance indicators

Performance indicator	UM	Case 1	Case 2
Total installed cost (ex. contingency)	MM €	69.30	98.91
Total investment cost	MM €	83.16	118.69
Specific investment cost per kW net	€ / kW	535.24	788.15
Total fixed O&M cost (year)	MM €	6.73	7.06
Total fixed O&M cost (MWh)	€/MWh	5.78	6.25
Total variable O&M cost (year)	MM €	28.73	28.50
Total variable O&M cost (MWh)	€/MWh	24.22	25.23
Total fixed and variable costs (year)	MM €	34.96	35.56
Total fixed and variable costs (MWh)	€/MWh	30.00	31.48
Levelized cost of hydrogen (LCOH)	€/MWh	37.57	43.03
CO ₂ removal cost	€/t	-	28.95
CO ₂ avoided cost	€/t	-	30.71

As can be observed from Table 4, the introduction of pre-combustion carbon capture involves important economic modifications as follows: specific investment cost increases by about 47.25%, the total fixed and variable costs increase by about 5% and the hydrogen production cost increases by about 14.5%. The CO₂ avoided cost is slightly higher than the current CO₂ emission tax (25 - 28 €/t [20]), but the difference is not significant showing the promising perspectives of decarbonization process.

Sensitivity studies (see Figure 4) were performed to see the influence of various key economic elements (e.g., natural gas price, capital and operational costs, internal rate of return, plant availability factor) on hydrogen production cost.

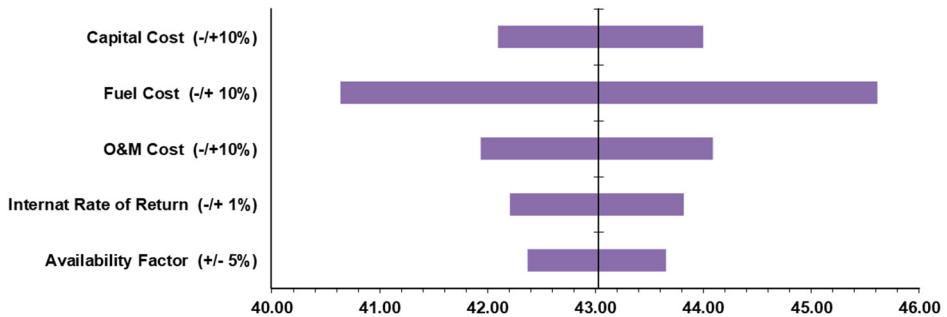


Figure 4. Sensitivity study for decarbonized hydrogen production cost

Finally, an LCA analysis was performed for the investigated cases to determine the most relevant environmental impact indicators. Table 5 presents the key environmental indicators for the evaluated cases.

Table 5. LCA environmental indicators

Performance indicator	UM	Case 1	Case 2
GWP	kg CO ₂ eq. / MWh H ₂	278.48	101.08
FEP * 10 ⁵	kg P eq. / MWh H ₂	39.29	40.28
ODP * 10 ⁹	kg CFC-11 eq. / MWh H ₂	5.12	5.27
FDP	kg oil eq. / MWh H ₂	107.98	112.56
FETP * 10 ²	kg 1,4-DB eq. / MWh H ₂	5.30	5.68
HTP	kg 1,4-DB eq. / MWh H ₂	3.74	4.25
MDP	kg Fe eq. / MWh H ₂	0.53	0.64
POFP * 10 ³	kg NMVOC / MWh H ₂	11.41	48.28
TETP * 10 ³	kg 1,4-DB eq. / MWh H ₂	0.44	0.78

It can be noticed that introduction of carbon capture feature involves a reduction of Global Warming Potential (GWP) by about 2.75 times (from 278.48 to 101.08 kg CO₂ eq./MWh H₂). The rest of environmental indicators show low to moderate increases (up to 15%). These can be explained by two main elements: reduction of overall energy efficiency due to carbon capture and usage of additional materials (e.g., chemical solvent) for plant decarbonization. Only Photochemical Oxidant Formation Potential (POFP) shows a significant increase compared to the base case (323%) due to the influence of chemical solvent (MDEA) production process.

CONCLUSIONS

This work is evaluating the main techno-economic and environmental performance indicators of decarbonization process for a hydrogen production plant based on natural gas reforming. As the decarbonization technology, the chemical gas-liquid absorption using MDEA was used. A 70% carbon capture rate was considered for a plant capacity of 50000 Nm³/h (corresponding to 150 MW hydrogen thermal output). As the results show, the decarbonization process of hydrogen production has a positive influence on key plant performance indicators e.g., low carbon capture energy penalty (about 2.3 net efficiency percentage points), moderate increase of the hydrogen production cost (by about 14.5%), attractive CO₂ avoidance cost in comparison to the current carbon tax (30.7 vs. 25 - 28 €/t) and 2.75 times reduction of global warming potential (assessed by a full LCA analysis). All these promising elements underline the potential of decarbonized hydrogen as a key energy carrier for the future low carbon economy.

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