Highlights:

- A sample case study based on valorization of hydrogen sulfide extracted from sour gas is presented.
- The potential economic favorability of modular chemical process intensification is demonstrated.
- A complementary evaluation of green solvents is also provided to further improve the sustainability of the proposed solution.
Modular and Intensified – Reimagining Manufacturing at the Energy-Chemistry Nexus and Beyond

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Graphical Abstract

Abstract

Today’s manufacturing is based on ample fossil fuel sources, large and centralized plants, and high waste intensity. Climate change, aging infrastructure, dwindling resources, increasing population, changing geopolitical landscape, and the COVID-19 pandemic have laid bare the frailties of the current global supply chain. While there is still place for centralized production, geographic variation in renewable energy sources and sustainable feedstocks calls for a flexible approach towards smaller-scale and more decentralized production. With the pressing need for decarbonization of power generation and the chemical value chain, flexible manufacturing will play a major role in redefining the energy-chemistry nexus. Intensification and modularization are identified as the key enablers for
such a transition. A sample case study based on valorization of hydrogen sulfide extracted from sour gas is presented to demonstrate the potential economic favorability of modular chemical process intensification. Our work shows that a net profit of US$97 million can be achieved over a five-year operational period when compared to a conventional process. A complementary evaluation of green solvents is also provided to further improve the sustainability of the proposed solution.

**Keywords:** process intensification, decentralization, waste-to-value, sustainability, energy-chemistry nexus

**Introduction**

For over a century, we have been heavily relying on fossil resources to meet our energy and chemical needs for various products, processes and/or services. However, because of the rising environmental concerns, further development of the sustainable alternatives to fossil resources is necessary. Although electricity production from sustainable resources is becoming economically competitive [1], increased grid penetration of variable renewable electricity critically relies on low-cost, robust, and reliable energy storage technologies [2]. Among these storage technologies, chemical storage by synthetic fuels is a promising solution because it can directly fit into the vastly developed existing infrastructure for use, transport, and storage. Apart from their importance as energy sources, synthetic fuels can also substitute fossil fuels for chemical production and industry. The substitution of fossil resources by sustainably produced synthetic fuels and chemicals using recycled carbon would help us reach net-zero carbon dioxide emissions and close the carbon cycle [3–5]. This approach is the basis for the Power-to-X (PtX) paradigm. PtX is the only known route to decarbonizing large parts of our heat, transport, and heavy industry sectors.

Unlike fossil fuels, sustainable energy resources are available locally, and fuel/chemicals are also locally consumed. Therefore, a transition toward synthetic fuels produced from captured carbon and
sustainable energy resources would significantly benefit from an increased share of decentralization in the chemical supply chain. Moreover, recent geopolitical instabilities and the COVID-19 pandemic have highlighted the vulnerabilities of a global supply chain that is based on single-sourcing and inventory models driven exclusively by cost control. Smoothly localized or regionalized supply chains enhance agility and resilience, allowing for nimble adjustments based on market demands, disruptions, and uncertainties. Transition toward more local or regional production means that units and plants operating at smaller scales will be required.

However, the chemical process industry has long relied on large plants with sprawling footprints to reduce the capital intensity based on the principle of economies of scale. To design chemical plants that meet the objectives of next-gen manufacturing, there needs to be a structural shift towards flexible production—flexibility in terms of location, capacity, feedstock, market volatility, customer demand, etc. [6]. Therefore, the challenge for the process engineers of today and tomorrow is to break the traditional reliance on the economies of scale while driving down the capital and operational costs of flexible plants. This has led to an increased interest in process intensification and modularization, especially at the policy level: European Roadmap for Process Intensification in 2007 [7], EU’s 2009-13 CoPIRIDE [8,9] and F³ Factory [10–14] projects, EU’s 2015-18 IbD project [15–17], US Department of Energy’s 2015 Quadrennial Technology Review [18] and its RAPID Advanced Manufacturing Institute [19] in early 2017, EU’s 2017-21 PROMECA project [20], EU’s 2020-24 BioSPRINT project [21], etc.

**Table 1** Few of the definitions proposed for process intensification.

| Authors (year)          | Definition                                                                 |
|-------------------------|---------------------------------------------------------------------------|
| Ramshaw and Arkley (1983) | Devising exceedingly compact plant which reduces both the main plant item |
| Authors (year) | Definition |
|---------------|------------|
| [22]          | and the installation costs. |
| Stankiewicz and Moulijn (2000) | Drastic improvements in chemical manufacturing and processing, substantially decreasing equipment volume, energy consumption, or waste formation and ultimately leading to cheaper, safer, sustainable technologies. |
| [23]          |                      |
| Touris and Porcelli (2003) | Replace large, expensive, energy-intensive equipment or process with ones that are smaller, less costly, and more efficient; combine multiple operations into fewer devices (or a single apparatus). |
| [24]          |                      |
| Arizmendi-Sanchez and Sharratt (2008) | Synergetic integration of process tasks and coupling of phenomena; targeted intensification of transport processes. |
| [25]          |                      |
| Van Gerven and Stankiewicz (2009) [26] | Maximize the effectiveness of intra- and intermolecular events; give each molecule the same processing experience which results in products with uniform properties; optimize the driving forces at every scale and maximize the specific surface area to which these forces apply; maximize the synergetic effects from partial processes which enable multitasking. |
| [26]          |                      |
| Lutze et al. (2010) [27] | Add/enhance phenomena in a process through the integration of operations, functions, phenomena or alternatively through the targeted enhancement of |
phenomena in an operation.

Baldea (2015) [28] Leads to substantially smaller, cleaner, safer, and more energy efficient technology or that combine[s] multiple operations into fewer devices (or a single apparatus).

A universal definition of process intensification (PI) has not yet been established, but all of the proposed definitions (see Table 1) overlap in their objective to reduce the cost and environmental footprint [29] of a process while increasing its productivity. Simply put, PI refers to the collection of developments that enable or significantly improve one or more physicochemical phenomena via synergistic integration of multiple phenomena or targeted enhancement of transport forces at various scales. Such developments are intended to result in smaller, cleaner, safer, more energy- and resource-efficient chemical processes.

On the other hand, modular chemical processing aims to dramatically reduce the complexity of construction, modification, replacement and/or assembly of different process units in a plant using novel technologies and manufacturing approaches in modular fabrication, design, and deployment. In such a modular world, a chemical plant is a highly customizable, small-to-medium scale engineering structure composed of standardized, modular substructures. This approach satisfies the flexibility requirements laid out above while offering profit realization in distributed production and risk minimization in both centralized and distributed production [6,11,30,31]. While PI and modularization can be used independently, it is beneficial to integrate the two approaches [32,33]. For modular processing to be cost effective, it must overcome the higher unit cost generally associated with using
the same technology as a full-scale, conventional process but at a reduced scale. One approach to achieve this is to utilize PI solutions, many of which require high surface-to-volume ratios and/or short length scales. This combination of intensification and modularization is referred to as modular chemical process intensification (MCPI) [19,32], which provides the base recipe for next-gen flexible manufacturing.

Few of the potential applications of MCPI-based distributed production include the biomass supply chain, shale or natural gas supply chain, PtX, and wastewater treatment. Biogas, syngas, and natural gas processing presents significant opportunities for modular and intensified processes—from gas upgrading and water processing to conversion technologies that could create viable and more sustainable alternatives to the large-scale centralized plants that need to improve energy efficiency and address the high capital investment required [19]. In the case of natural gas, modular small-scale gas-to-liquid or gas-to-wire technologies have recently been developed to make use of stranded, associated, or declining gas reserves that would otherwise be economically infeasible for large-scale production infrastructure [34–37]. MCPI, including the technique discussed later in this text, can also be used for integrated carbon capture and utilization (CCU) from the waste streams of such distributed energy production facilities [38–45]. These developments are critical for cost-effective realization of the PtX concept since they allow for mild and indirect hydrogenation routes to methanol [46–48]. Conventional carbon dioxide capture and storage demand high capital and energy requirements due to the expensive desorption and compression steps. In post-combustion capture processes, these two steps respectively account for about 20% and 40% of the capital costs and 60% and 30% of the energy costs [49]. Eliminating the compression and transportation of purified CO₂ while integrating desorption with absorption towards a viable utilization pathway presents an exciting MCPI solution for wide-scale deployment of CCU technology. Furthermore, many bioprocesses including wastewater treatment...
depend on highly distributed, localized feedstocks that require the development of efficient MCPI technologies for a hybrid centralized-decentralized production landscape [50–56].

In the aforementioned processes and many others, removal of impurities or hazardous pollutants from various process streams is essential and, in some cases, mandated by law. Hydrogen sulfide (H₂S) is a colorless, corrosive, highly toxic, and flammable gas that is found as an impurity or an inhibitor in many chemical and biochemical processes, such as processing or purification of natural gas, syngas, biogas, landfill gas, fuel gas, CO₂ feedstocks, natural gas liquids, etc. Environmental and/or process constraints often require the removal of sulfur content to trace amounts. Several different classes of H₂S removal technologies exist or have been reported [57]: chemical or physical absorption, adsorption, membrane separation, cryogenic distillation, direct chemical or biological oxidation, hybrid (combination of two or more) processes, etc. The choice is generally based on various factors such as the feed composition, gas volume, space and weight considerations, and location. For offshore applications, space and weight limitations are generally tighter and more crucial.

This work investigates an MCPI-derived solution to a present industrial need of an economical and sustainable process for offshore natural gas sweetening. In this regard, Section 2 will discuss the current practice of H₂S removal on Danish offshore platforms, while section 3 will present the MCPI-derived alternative, analyze its economic impact, and evaluate further improvements in terms of sustainability. Section 4 presents the concluding remarks. With this work, we also probe the following general questions. Can MCPI-based solutions be cheaper and more sustainable than their traditional counterparts? Is there a need or an advantage in investing in the development and implementation of MCPI-based solutions today?
H₂S Removal on Offshore Platforms

As shown in Figure 1a, H₂S produced on offshore platforms in the Danish North Sea is generally removed using a non-regenerative triazine-based liquid scavenger. Among triazine-based scavengers, 1,3,5-tri-(2-hydroxyethyl)-hexahydro-s-triazine (MEA triazine) is the most widely used one in the offshore oil and gas industry [58,59]. The multiphase fluid containing crude oil and associated gas extracted from the reservoir is initially put through a three-phase separator where most of the H₂S leaves with the resulting lift gas [60]. Although the scavenger is injected at multiple points along the production line, the major injection point is between the three-phase separator and the scrubber where the concentration of H₂S in the gas phase is the highest. The treated gas from the scrubber is then compressed and cooled to remove the natural gas liquids before being sent to a dehydration unit. On the other hand, the spent scavenger stream is sent through a condensate separator and a degasser before being discharged with produced water. This process converts H₂S to less acidic, water-soluble compounds in an irreversible reaction, which involves the nucleophilic substitution of sulfur into the triazine ring. It is reported that the triazine is first converted to thiadiazine and then to dithiazine where the reaction terminates [58,61].

Although this process is rather established, it comes with significant challenges. The irreversible chemical substitution reaction has a limited uptake capacity. The capacity generally varies, but most commercially available formulations have a stoichiometric uptake capacity of about 0.15 kg/L H₂S with the maximum practical capacity limit of approximately 80% of that value. Controlled contact between triazine and H₂S is critical, as either excessive contact even at low H₂S concentrations or lesser contact at high concentrations of H₂S can over-react triazine and lead to polymerization and precipitation of amorphous dithiazine when the scavenger is 60% or more spent [58,62]. These deposits can precipitate and collect in aggregates that form blockages in process equipment and cause fouling of heat.
exchangers, tubulars, tank trucks, water disposal wells, scrubbers, compressors, and so forth. The necessary remedial action typically involves taking the affected equipment offline so that manual cleaning operations can be performed. Typically, this involves jetting and can be very time-consuming and challenging [61]. To both meet the gas quality standards and circumvent the need for control and monitoring, it is common practice to use excessive amounts of triazine, resulting in prohibitive costs for treatment. In addition, the spent scavenger solvated in the aqueous phase is currently discharged with the produced water generated at the facility. Although the products of the scavenging process are generally non-toxic, the unreacted reagent is toxic to aquatic environment posing further risk during disposal. With increasing levels of H₂S and tightening environmental laws, this disposal practice will soon raise a concern and likely to become infeasible. Overall, this process faces serious questions in terms of cost and sustainability.
Figure 1 Simple illustrations of the process configurations for the (a) currently used triazine-based removal and (b) proposed PTC-based removal.

An alternative to triazine-based scavenging for offshore H$_2$S treatment is alkanolamine-based absorption which is one of the most mature and widely employed H$_2$S removal technology in the industry [59]. However, as is the case for carbon capture, alkanolamine-based absorption of H$_2$S is accompanied by an expensive desorption step after which the captured H$_2$S stream is sent to a Claus unit to recover elemental sulfur. Currently, recovered elemental sulfur is produced primarily during the processing of natural gas and crude petroleum. Due to the depletion of sweet oil and gas reserves around the world,
players in the O&G industry are increasingly looking toward sour fields to meet our energy needs. In addition, with the ever-growing adoption of waste-to-biogas and biomass gasification processes, the amount of recovered sulfur is only bound to increase. In the past 2-3 decades, sulfur production has consistently outpaced the demand leading to the large stockpiles of block sulfur waste in several countries around the world [63]. At a time when there is an increasing momentum to achieve a (near) zero-waste and circular economy, bulk sulfur disposal poses a serious barrier to such a transition. Moreover, the conventional alkanolamine-based absorption requires either the compression and transport of captured H$_2$S for further processing onshore or the installation of several additional equipment units offshore for sulfur recovery via either the multistage Claus process or the LO-CAT liquid redox process. These disadvantages make it either impractical or expensive and unsustainable. Lower hydrocarbon pricing and shifting public opinion demand either less expensive and more sustainable production or shutting down production in regions entirely. This has necessitated more innovative, economical, and sustainable H$_2$S treatment methods that can fulfil the threshold limit requirements.

**Process Intensification: Phase Transfer Catalysis**

Liquid-liquid phase transfer catalysis (PTC) is a type of multiphase catalysis that involves simultaneous reaction and extraction in a biphasic system, usually an aqueous-organic system. While several reactions simultaneously occur in the two immiscible liquid phases, a surface-active transfer catalyst (for example, tetrabutylammonium ion) continuously transfers a reacting inorganic anion from the aqueous phase to the organic phase to facilitate one or more main reactions that would otherwise not take place [64,65]. In addition to this targeted enhancement of interfacial transport, several phenomena are synergistically integrated leading to intensification: (a) the catalyst combines with the reacting anion in the aqueous
phase to form an active catalyst species, (b) the produced active catalyst species transfers from the aqueous phase to the organic phase due to a shift in phase equilibrium, (b) the transferred anion pushes the equilibrium of the main reaction set in the organic phase towards the product(s) and the regenerated catalyst species, (c) the regenerated catalyst species transfers back from the organic phase to the aqueous phase due to a shift in phase equilibrium, and (d) The countercurrent transfer of the regenerated catalyst and the active catalyst species pushes the reaction equilibrium in the aqueous phase towards the formation of more active catalyst species and in the organic phase towards the formation of more product(s). This intensified system offers numerous advantages, such as novel synthesis routes, high yields and purity of the products, operational simplicity, mild reaction conditions, easier product separation, suitability for large-scale synthesis, and an environmentally benign nature of the reaction system.

**Figure 2** Phase transfer catalysis in terms of intensification principles: synergistic integration of reaction, interfacial extraction, and phase separation along with targeted enhancement of interfacial mass transfer using a phase transfer catalyst.

To overcome the severe drawbacks associated with both the triazine-based scavenging and the conventional alkanolamine-based processes, an MCPI-inspired solution in the form of phase transfer catalysis is showcased here as the cornerstone of a modified alkanolamine-based process. Instead of
generating a considerable quantity of potentially toxic waste, a highly sustainable and relatively profitable route of converting the hazardous \( \text{H}_2\text{S} \) to value-added products is presented. The proposed PTC-based strategy replaces the expensive stripping/desorption column in a conventional alkanolamine-based process with an intensified biphasic catalytic reactor in which solvent regeneration and \( \text{H}_2\text{S} \) conversion to value-added sulfur-containing products take place simultaneously.

**Figure 3** An illustration of phase transfer catalysis taking place in the biphasic reactor.

Figure 1b shows a simple schematic of this process. In the scrubbing column, the sour gas stream is put into contact with a countercurrent stream of aqueous \( \text{N}-\text{methyl} \text{diethanolamine} \) (MDEA) solution. \( \text{H}_2\text{S} \) is dissolved into the MDEA stream in the form of inorganic sulfide ions while sweetened natural gas leaves at the top of the scrubbing column. The \( \text{H}_2\text{S} \)-rich aqueous MDEA stream, along with an organic stream, is then sent into a biphasic reactor (see Figure 3). The organic stream contains an organic reactant (benzyl...
chloride) and a phase transfer catalyst (tetrabutylammonium chloride/bromide, QCl/Br); the latter migrates to the aqueous phase and transfers the inorganic sulfide ions to the organic phase to react with the former to produce benzyl mercaptan and dibenzyl sulfide. This continuous removal of dissolved H$_2$S from the aqueous MDEA solution allows for the simultaneous regeneration of the solvent. The regenerated MDEA solution and the organic mixture (including organic solvent and sulfur-containing products) are sent to a decanter or a gravity separator to separate and recycle H$_2$S-free MDEA solution to the scrubbing column. In the final step, the products are separated from the organic solvent, which is then recycled back into the biphasic reactor. As an application case study, the Gorm field in the Danish North Sea is considered here. While the details on the specific chemical system are provided in section S2 of the supplementary text, a few important performance indicators from the Aspen Plus simulation and mathematical modeling are presented in Table 2.

| Parameter                        | Value       |
|----------------------------------|-------------|
| Sour gas H$_2$S content          | 2%          |
| Sweet gas H$_2$S content         | < 3 ppmv    |
| Lean amine MDEA content          | 30 wt%      |
| Rich amine H$_2$S loading        | 0.59 mol/mol|
| H$_2$S capture efficiency in the absorber | > 98.5%    |
| H$_2$S removal/conversion efficiency in the biphasic | 100%       |
Techno-Economic Analysis

To quantify the economic benefits of adopting this intensified solution, a technoeconomic analysis (TEA) is conducted with several assumptions (see section S4 of the supplementary text) to allow for a more complete comparison between the currently used triazine-based scavenging process (process P1) and the proposed PTC-based process (process P2) for offshore use. A few of the more important assumptions and considerations are briefly mentioned here. Since the discount rate for the net present value (NPV) analysis is usually an estimate of the rate of return expected by the investors, it is calculated as the weighted average cost of capital using the capital asset pricing model to estimate the risk associated with the investment. As a result, the discount rate for the established process P1 and the new process P2 are calculated to be 3.99% and 11.35%, respectively. The tax rate for the facility is taken to be 34.1%. Both the discount rate and the tax rate are based on information available from Total, the owner of the Danish North Sea operations. For process P1, no capital investment is considered.

**Figure 4** (a) Cumulative discounted cash flow (DCF) for the three different process P1, P2 and P3 (left to right on the legend), (b) Effect of H₂S concentration in the inlet sour gas on the NPV of both the triazine and the proposed PTC-based processes.
For the NPV analysis, in addition to the processes P1 and P2, a hybrid process scenario (P3) is considered in which 30% of the sour gas volume is treated by process P2 while the rest is continued to be treated by process P1. This percentage value was chosen in consultation with our industrial partners to allow for a smooth transition to process P2. Figure 4a shows the NPV comparison of these three scenarios. It clearly demonstrates the significant economic advantage associated with process P2 relative to process P1.

While P1 has negative NPV of US$44 million exclusively based on its operational expenditure, P2 results in an NPV of US$53 million over the same five-year period due to an average annual operating income of over US$10 million. Moreover, treating just 30% of the sour gas as in process P3 lowers the NPV of P1 by US$28 million, strongly indicating that a partial or complete switch to the proposed process P2 is not only sustainable but also economical.

Concentration of H$_2$S in the sour gas is an important parameter that influences the TEA results. As such, Figure 4b presents how NPVs of P1 and P2 vary with this parameter. While NPV of P2 is directly proportional to the amount of H$_2$S in sour gas, the relationship becomes inversely proportional in the case of P1. This trend is unsurprising since P2’s profitability is largely dependent on the amounts of products manufactured from the capture and conversion of H$_2$S present in the sour gas. Similarly, since P1 does not have a revenue stream, the operational costs of the process increase with the amount of H$_2$S that must be treated. Figure 4b also shows that the proposed process P2 would never reach a breakeven point for H$_2$S concentrations below 500 ppmv, approximately. However, it remains relatively profitable in comparison to the base process P1 regardless of the inlet H$_2$S concentration.

Figure 5 shows the sensitivity analysis of P2 with respect to several other parameters. The most influential factor is the price(s) of the product(s), which can make or break the technology application since it directly impacts the generated revenue. This necessitates diversity in the high-value, sulfur-based product portfolio so as not to influx the market with significantly high amounts of the same
product which could drive its price down. On other hand, equipment and utility costs are the least influential factors with a marginal effect on the NPV of P2. This shows that the TEA of P2 presented in this work remains largely consistent even in the absence of a completely designed process flowsheet.

![Sensitivity analysis for the NPV of the PTC-based process.](image)

**Figure 5** Sensitivity analysis for the NPV of the PTC-based process.

It is worth noting that the process P2 is clearly beneficial for onshore applications too as it is a direct improvement over the conventional alkanolamine-based process. Since PTC allows for the intensified extraction-reaction of H$_2$S at mild conditions, the biphasic reactor significantly reduces both the capital and operating costs of the desorption step when compared to the stripping column in the conventional MDEA-based process. In addition, since this process does not require the use of the modified Claus process to convert the pure H$_2$S stream to elemental sulfur or the disposal of any toxic or non-toxic waste, it presents an attractive opportunity to economically reduce the total environmental footprint of the process.
The proposed process can be further intensified and improved with the advancements in the design of reactors and gas-liquid contactors. Microchannel technology has the potential to transform the energy and chemical processing industries by greatly reducing the size of equipment modules while boosting performance. Microchannel reactors can provide 10-1000 fold acceleration in performance by reducing the heat and mass transfer resistances [66]. As a result, on top of the considerable energy savings, the reaction time can be reduced from hours to minutes, or even seconds [67]. Similarly, replacing the conventional absorption tower with a rotating packed bed would significantly lower the size and cost of the unit required to capture H$_2$S. A rotating packed bed provides greatly increases mass transfer rate and enhances micro-mixing at short contact times [68,69], making it ideal for selective capture of H$_2$S from gaseous streams containing other impurities such as carbon dioxide [70–73]. This could allow for PTC and other MCPI technologies to be deployed more economically than demonstrated here.

**Solvents and Sustainability**

Solvents are a major source of energy usage and waste generation in many chemical, biochemical, and petrochemical processes, including PTC. Due to the increasingly tight regulations and strong public opinion, a thoughtful selection for process-efficient and sustainable solvents in industrial activities has become an immediate requirement for chemists and chemical engineers alike. In the previous section, PTC has already been shown to substantially improve the economics of offshore natural gas sweetening processes by replacing the existing triazine-based H$_2$S removal. It also makes hydrogen sulfide removal more sustainable by eliminating the significant amounts of waste generated by the current process. In this section, the process P2 is further improved by employing a quantum-chemistry-based screening method [74] to find green and sustainable solvents for the PTC-based process. Since one of the solvents in the biphasic system is water, the alternatives are evaluated only for the organic solvent, toluene.
The environmental impact of the solvent candidates is evaluated based on four impact categories, namely human toxicity potential by ingestion (HTPI), human toxicity potential by dermal exposure or inhalation (HTPE), aquatic toxicity potential (ATP), and photochemical oxidation potential (PCOP). These categories are chosen based on the specific process and application. The volatility of the organic solvent permits its recovery by distillation but also creates unwanted air emissions and the risk of worker exposure. This brings both the human toxicity categories and PCOP into focus. Since the process is carried out offshore where produced water is discharged to the sea, ATP is chosen to represent the toxicity impact on the surrounding water body. It must be noted that HTPI, HTPE, and ATP are measures of local impact, while PCOP is a measure of regional impact.

However, choosing the best solvent for a specific application depends not only on the environmental impact but also on several other factors, such as process performance, ease of product recovery, and price. As such, a more holistic approach is taken. The methodology is described in detail by Pudi et al. [74] and in section S5 of the supplementary information. A brief description is provided here. Eight potential alternatives to toluene are pre-selected based on their water immiscibility from the large pool of solvent candidates assessed in the CHEM21 selection guide [75]: 1-butanol, 2-methyl tetrahydrofuran (2-MeTHF), anisole, benzyl alcohol, butyl acetate, ethyl acetate, isopropyl acetate, and methyl isobutyl ketone (MIBK). The environmental impact factors (EIFs) of these eight candidates and toluene are calculated using the ProPred toolbox in the Integrated Computer-Aided System [76]. These EIFs are then normalized within each category, weighted, and summed to obtain the environmental impact of each solvent as a single score (SS). In this work, all the impact categories are weighted equally.

Electronic structure calculations based on density functional theory (DFT) and the implicit solvent Conductor-like Screening Model (COSMO) [77] were carried out for the nine solvents and all the other species in the system using the quantum chemistry program package TURBOMOLE, version 7.4 [78,79].
Transition state structures for the two reactions, one each for the formation of the two products, were obtained from performing reaction path optimization. All the optimized DFT-COSMO molecular structures were transferred to COSMOtherm Release 19 [80], where COSMO for real solvents (COSMO-RS) [81,82] was used to obtain the partition coefficients and the free energies of solvation of various species in the biphasic solvent systems. Free energy of each reactant, product, and transition state in solution is then obtained as the sum of its ground state electronic energy in gas phase, thermal corrections to the ground state energy, and free energy of solvation. Free energies of activation and reaction are subsequently calculated using the same procedure as reported by Hellweg and Eckert [83]. From here on, R1 and R2 refer to the reactions that result in the formation of benzyl mercaptan (BnMer) and dibenzyl sulfide (DBS), respectively (see section S2 in the supplementary text).

![Graph](attachment:image.png)

**Figure 6** Comparison of toluene against the selected alternatives in terms of their environmental impact single scores (broken down by category), prices, and boiling points.
With this information gathered, the solvents were evaluated based on several important criteria: environmental impact, solvent price, ease of product separation, partition behavior, reaction energy barriers, and reaction free energies. As initially noted, the main objective here is to find greener alternatives to toluene. As such, the environmental impact of each of the eight candidates relative to toluene is the most important criterion. Figure 6 compares the environmental impact scores (SS), prices, and boiling points of all the solvents. Butyl acetate is the only solvent to have a worse SS than toluene, making it the only candidate that cannot be considered a greener substitute to toluene in this process. On the other hand, ethyl acetate, isopropyl acetate, MIBK, and benzyl alcohol exhibit much better/lower SS than the other alternatives. It could be argued that ATP is the most crucial EIF in an offshore platform since any solvent purged from the process is likely added to the produced water that’s discharged to the sea. Therefore, it is imperative to choose a solvent with a low normalized ATP score, especially in comparison to toluene. The four solvents with low SS also meet this criterion, further proving their ability to be green substitutes for toluene. In addition, they come at prices that are not too far from that of toluene, thus making a minimal impact on the previously discussed NPV analysis.

Ease of product recovery affects both the economics and the total environmental footprint of the process. Organic solvents with lower volatilities and higher boiling points correspond to lower emissions of volatile organic components. However, such solvents require a more expensive product recovery since the most common method for product recovery is distillation. In terms of the specific chemical system under study, BnMer is the lower-boiling product with a boiling point is 195 °C. Therefore, an easier product recovery requires the boiling point of the solvent to be lower than 195 °C to a satisfactory extent. However, it can’t be too low since the reaction temperature is about 60 °C and since the boiling point of an immiscible liquid mixture is lower than the boiling point of each of the constituents. Figure 6 shows that anisole, benzyl alcohol, and butyl acetate make bad candidates because their high boiling points are close to that of BnMer, while 2-MeTHF and ethyl acetate may pose a slight risk because their
boiling points are only about 20 °C higher than the reaction temperature. However, VLE data computed using COSMOtherm (see section S5.5) shows that the minimum boiling point of the binary mixtures of water with ethyl acetate and 2-MeTHF is more than 73 °C and 78 °C, respectively. Therefore, we conclude that there is no risk of any of the solvents boiling at the reaction conditions, especially in a closed system with proper temperature control.

Figure 7 Partition coefficients (organic/aqueous) of the three catalyst species in all solvents

In PTC, the partition behavior of the reactants, products, and the catalyst species is an important consideration. Partition coefficients for all the species were calculated in COSMOtherm. A value greater than 1 implies a preference for the organic phase, and a value less than 1 implies a preference for the aqueous phase. A good organic solvent for this application would result in high partition coefficients for BnCl and BnMer. Since all the considered solvents exhibit this behavior, this becomes irrelevant for screening. Nevertheless, proper partition behavior of the active and inactive catalyst forms is crucial for an efficient and effective PTC. The active catalyst form, tetrabutylammonium hydrogen sulfide (QSH), must transfer to the organic phase to form BnMer, while the resulting inactive catalyst forms, QBr and
QCl, must simultaneously transfer to the aqueous phase to regenerate QSH from the dissolved bisulfide ions. Ideally, the partition coefficient of QSH should be greater than 1, and the partition coefficients of QBr and QCl should be less than 1. Figure 7 shows (in log-scale) how the solvent candidates affect the partition coefficients of the three catalyst forms. All three species have high partition coefficients in 1-butanol, 2-MeTHF, and benzyl alcohol, making them less favorable in terms of process performance. On the other hand, ethyl acetate, butyl acetate, and anisole cause the desired partition behavior. Overall, alcohols seem to perform worse than the other classes, while alkyl acetates (or more generally, alkyl esters) seem to be the best solvents. Interestingly, an ether (anisole) provides a much better partitioning behavior in comparison to a cyclic ether (2-MeTHF). These observations could prove useful in a more rigorous computer-aided solvent design.

![Figure 8](image_url)

**Figure 8** Free energies of (a) activation and (b) reaction in all the considered solvents

Faster kinetics lead to a smaller reactor volume, while better thermodynamic favorability results in higher yield. The free energies of activation and reaction are a measure of kinetics and thermodynamic favorability, respectively. Figure 8a shows that, except for 1-butanol and benzyl alcohol, all the candidates result in an improved R1 kinetics in comparison to toluene with 3.4–7.9% reduction in the reaction energy barrier. In the case of R2, all the alternatives perform on par or better than toluene with 1-butanol and butyl acetate providing the best margin. However, neither of them can be used due to
their ineffectiveness in other criteria. When it comes to thermodynamics (see Figure 8b), all the solvents perform similarly with highly negative reaction free energies in the case of R1. However, in the case of R2, the reaction free energies are much lower, making the choice of solvent more impactful. Here, all the alternatives perform better than toluene. However, except for anisole, the rest provide significantly better thermodynamics by a factor of 2–4.

Table 3 presents a short and easy summary of the screening results. Considering all the screening criteria used in this work, ethyl acetate comes out as the best solvent with isopropyl acetate and MIBK also doing quite well, warranting a further look into their capability to replace toluene. As seen, alkyl esters generally performed much better than the other classes of solvents, making them an attractive option for more comprehensive optimization-based solvent design.

**Table 3** Color-Coded Summary of the Green Solvent Screening Results. Green indicates that a candidate is the best in relation to others and toluene. Orange indicates that a candidate is okay. Red indicates that a candidate is not suited.
Conclusions

The future of manufacturing is smart and flexible, and it is on the horizon. 20th century solutions are insufficient to satisfy the requirements of today and tomorrow. Alongside digitalization and additive manufacturing (which are not discussed in this article), modular and intensified processes will play a key role in shaping and designing the next-gen flexible manufacturing that goes beyond the traditional realm of chemical engineering. Stakeholders who take the steps today towards such a change are likely to reap the benefits of a more developed technology toolkit. Because financial feasibility is the main barrier to moving towards modular plants, a combination of modularity and intensification is proposed as the way forward to create more flexible manufacturing plants while eliminating the increased costs associated with decreasing the scale of equipment. In this work, one such MCPI-derived solution is applied to a current industrial need of natural gas sweetening on offshore platforms. This solution, based on phase transfer catalysis, is shown to provide significant advantages in terms of cost and sustainability when compared to the more established alternatives. Compared to the triazine-based process, the proposed PTC-based strategy offers a net profit of nearly US$100M over a 5-year operational period. The sale of the product(s) creates significant revenue for the PTC strategy, particularly at higher inlet H$_2$S concentrations. In addition, several green solvents are evaluated for the PTC process, and ethyl acetate is chosen as the best candidate with respect to various technical, economic, and environmental criteria.
The successful capture and utilization of H$_2$S by converting it to value-added products realizes the three objectives of sustainability: waste minimization, resource utilization, and environmental pollution reduction. Future work should focus on experimental validation followed by more rigorous modeling and simulation, especially in the space of green solvent exploration. Furthermore, this strategy can be extended to CO$_2$ capture and utilization to produce valuable chemicals, such as cyclic carbonates and methanol.

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CRediT Author Statement

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Declaration of interests

☐ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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