

Article

Concept for Temperature-Cascade Hydrogen Release from Organic Liquid Carriers Coupled with SOFC Power Generation

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SUMMARY

For a sustainable hydrogen economy, large-scale transportation and storage of hydrogen becomes increasingly important. Typically, hydrogen is compressed or liquified, but both processes are energy intensive. Liquid organic hydrogen carriers (LOHCs) present a potential solution for mitigating these challenges while making use of the existing fossil fuel transportation infrastructure. Here, we present a process intensification strategy for improved LOHC dehydrogenation and an example of clean power generation using solid oxide fuel cells. Four LOHC candidates—ammonia, biphenyl-diphenylmethane eutectic mixture, *N*-phenylcarbazole, and *N*-ethylcarbazole—have been compared as stand-alone and integrated systems using comprehensive process simulation. “Temperature cascade” dehydrogenation was shown to increase the energy generated per unit mass (kWh/kg LOHC) by 1.3–2 times in an integrated system compared to stand-alone LOHC systems, thus providing a possibility for a positive impact on a LOHC-based hydrogen supply chain.

INTRODUCTION

Utilization of hydrogen as an energy vector in a renewable-energy economy presents significant advantages as it is abundant, has a high gravimetric energy density¹ (120 MJ/kg), and the production can (and has been) integrated with surplus renewable electricity. The price of H₂ produced from renewably generated electricity can be expected to decrease as the total global supply of renewable electricity sources increases. Coupled with emerging high-efficiency conversion technologies such as reversible protonic ceramic electrochemical cells, with a reported 97% electric-to-hydrogen efficiency,² the price of renewably generated hydrogen should become more competitive as the industrial supply of these conversion technologies is increased within the next decade, according to Glenk and Reichelstein.³ One of the challenges of the utilization of hydrogen as an energy vector lies in storage and transportation technologies. Elemental hydrogen under 25°C and 1 atm has a low density of 0.08375 kg/m³ (8.38e⁻⁵ kg/L),⁴ and thus the lowest volumetric energy density of ~0.01 MJ/L compared to most of the commonly used fuels. Consequently, to store (for later utilization or transport) any quantity of hydrogen, most common, large-scale hydrogen storage techniques such as compressed hydrogen (200–700 bar),⁵ and liquefied hydrogen (–253°C)⁶ require somewhat rigorous conditions of pressure and temperature, which translate into significant energy expenditures and safety considerations. As this inhibits the progress of the hydrogen economy, alternative hydrogen storage techniques in the form of a liquid organic carrier have been extensively researched and developed in the past decade.^{7–10}

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Numerous advantages of liquid organic hydrogen carriers (LOHCs) as attractive storage compounds, owing to their cyclical nature, relative safety, well developed production technology, and comparative lack of carbon emissions, have been widely reported elsewhere, particularly by Reuß et al.,^{11,12} Preuster et al.,¹³ Runge et al.,¹⁴ and many others, and are not discussed here in detail. Compared to conventional storage technologies, the use of LOHCs essentially allows for a wide selection of globally produced and well-characterized chemicals (in terms of their handling and chemistry), providing a suitable selection for integration in the present liquid-fuel-based transportation infrastructure. A suitable LOHC candidate can ideally be used to bridge the distance between locations of high renewable energy supply and high clean energy demand, which are, on a global average, non-urban and urban, respectively. On the energy-demand side, hydrogen must be released from its LOHC in an endothermic chemical reaction at high temperatures. The source of the energy for that release (fossil fuel or renewable) and the efficiency of its usage will directly affect the overall efficiency of the LOHC-derived renewable electricity. Dehydrogenation processes for various LOHC candidates have been designed and theoretically evaluated.^{15–17} Eypasch et al.¹⁸ have modeled and performed an economic evaluation of an industrial-scale LOHC system based on dibenzyltoluene (DBT) (an LOHC used partly in this work) and have concluded that an energy-self-sufficient system (at this time) has a 47% higher cost per unit of electricity produced than an average grid price.

This study presents a previously unexplored process-intensification strategy for LOHC dehydrogenation, with an ultimate goal of increasing the energy efficiency of the process and consequently improving the viability of LOHCs in a hydrogen-based energy economy. A dehydrogenation process has been designed and simulated in which four LOHC chemicals are combined in a cascading fashion using the same energy source. This process is then compared with single LOHC systems, which use equal energy input and are compared based on the hydrogen yield. Finally, all of the processes are integrated with a solid oxide fuel cell (SOFC) model and compared based on the total kilowatt hours generated per kilogram of the LOHC chemical. The temperature-cascade (TC) LOHC dehydrogenation process was demonstrated to yield substantial performance improvements compared to systems that use single LOHC chemicals and can be used (e.g., in SOFCs) for a carbon-free stationary power generation system.

RESULTS AND DISCUSSION

Heat Source Flowrate and Temperature Comparison

Single LOHC systems and the TC LOHC system were first compared based on the same fixed heat source (flue gas), with a flowrate and temperature of 240 kg/h and 580°C, respectively (Figure 1). As can be observed, when compared to LOHC systems 2, 3, and 4, the TC LOHC system performs second best in terms of H₂ generated (4.36 kg/h) and the best in terms of total electrical energy produced per kilogram of LOHC (1.06 kWh/kg). This value was chosen as a goal to be maximized as it potentially has a beneficial impact on the LOHC supply chain, since more energy can be cleanly produced for the same mass of LOHC.

An obvious outlier here is the LOHC 1 system (ammonia), which produces 2.53 kWh/kg (LOHC). Although comparatively higher, this value must be observed differently from other systems because ammonia converts into hydrogen and nitrogen and therefore cannot be hydrogenated again, but rather synthesized. Therefore, in terms of the LOHC supply chain, the energy difference of ammonia synthesis and LOHC

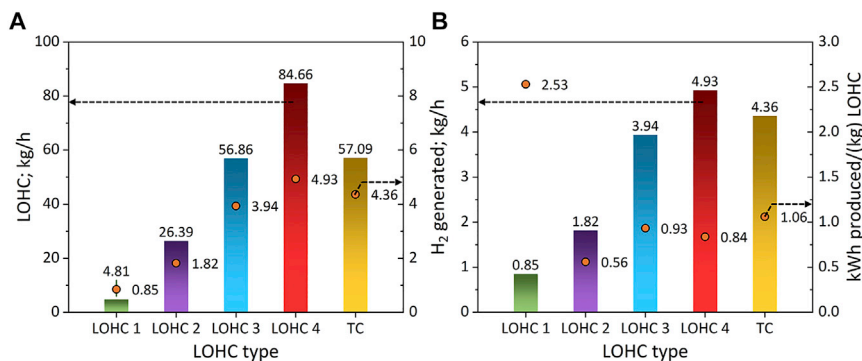


Figure 1. Hydrogen and Electricity Generation Comparison

(A) Comparison of single LOHC and TC systems based on optimal LOHC flowrate and resulting H₂ generated.

(B) Electricity produced in kilowatt hours per kilogram of LOHC.

hydrogenation (charging) must be taken into account, which is beyond the scope of the present study.

Heat Source Variable Temperature Comparison and Relative System Sizing

As the temperature of the flue gas can be adjusted by varying the air:fuel ratio, a sensitivity analysis was performed to account for the effect of different temperatures (and, consequently, flows) of the heat source (Figures 2A and 2B). It can be observed that the change in total power generated is the greatest in LOHC systems that have higher reaction temperatures (LOHC 1 and 2) as the temperature of the heat source increases. For the temperature variation in [580°C–730°C], the TC system outperforms LOHCs 2–4 in terms of power generated per kilogram of LOHC by an average factor of 1.3 compared to LOHC 3, and an average factor of 2 compared to LOHC 2, while LOHC 4 is between LOHCs 2 and 3. Analogous to Figure 1, LOHC 1 is an outlier in terms of the sharp increase of kilowatt hours per kilogram LOHC with respect to increasing temperature and must be viewed differently because of the reasons outlined earlier.

To provide a relative comparison of the reactor system complexity between the single and TC LOHC systems' reactors, they were sized assuming a tube bundle continuous reactor with set dimensions of a single tube of 1.5 m length and 0.15 m diameter (Figure 2C). The total reactor volume (as a sum of single tube volume integers) for complete dehydrogenation was assessed assuming the dehydrogenation kinetics for DBT-pentahydrate (DBT-18) published by Peters et al.¹⁹ It was not presumed that these kinetics accurately model the actual kinetics of LOHCs in this study. Rather, they were used simply to compare the relative reactor size differences since the same LOHCs are compared in both single and TC systems.

Under this assumption, the total number of reactor tubes was determined for each case (Figure 2D). The total number is an average for each LOHC system considering the heat source temperature variation. From Figure 2D, it can be observed that the average total number of reactor tubes for the TC system is comparable to LOHC 3, which is between LOHCs 4 and 2. Net power produced per reactor tube, however, provides a clear advantage for the TC LOHC system as it can generate more power with the same number of reactor tubes compared to the single LOHC systems. This metric is even higher than the outlier LOHC 1, which further demonstrates the increase in efficiency (in terms of energy generated per same amount of equipment)

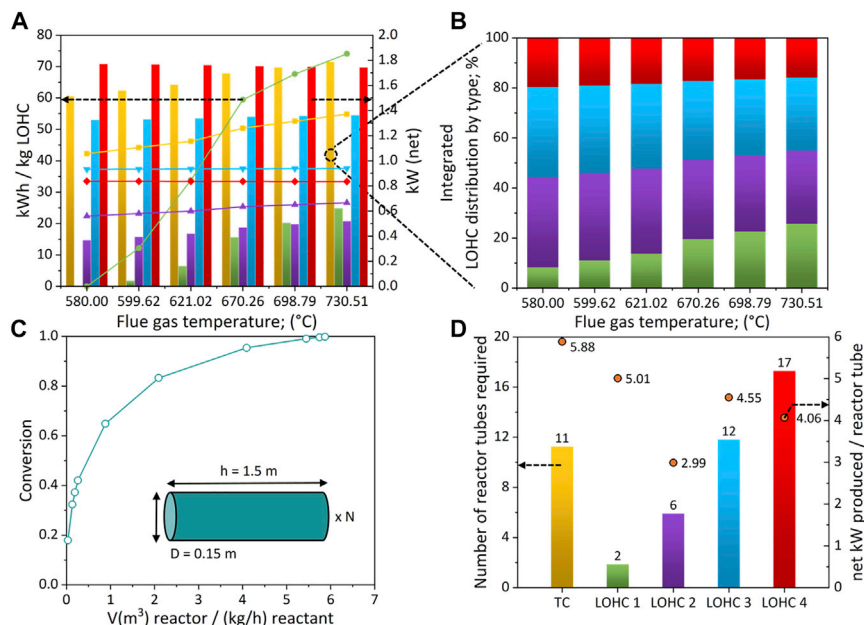


Figure 2. Heat Source Temperature Variation and Relative Reactor Sizing

(A) Heat source temperature variation effect on the total power produced and power per kilogram of LOHC.

(B) TC system LOHC optimal distribution in relation to flue gas temperature.

(C) Required reactor volume in relation to LOHC conversion.

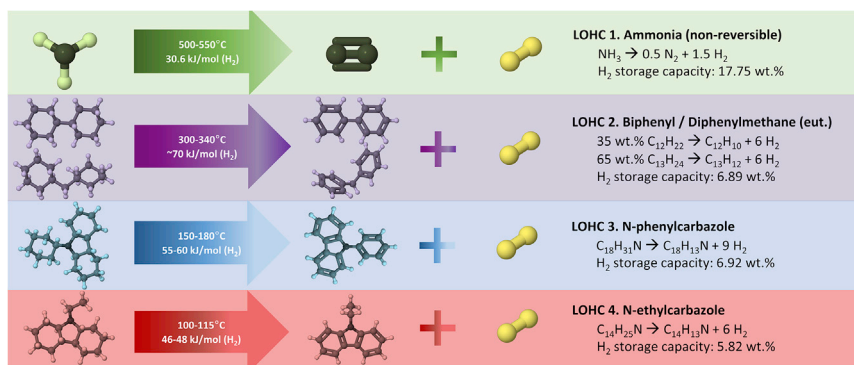
(D) Required number of reactor tubes for LOHC system and electricity generated per reactor tube. LOHC 1, 2, 3, 4, and TC: green, purple, blue, red, and yellow, respectively.

compared to LOHCs with the highest H₂ storage potential. Compared to single LOHC systems, the net power generated per reactor tube for TC LOHC increased by a factor range of 1.17–1.97.

To summarize, in this study, a process-intensification strategy for the increased efficiency of LOHC dehydrogenation was comprehensively evaluated. Four LOHC candidates were combined into a single system that uses the same heat source for dehydrogenation through LOHC reaction temperature differentials in a TC fashion. Such a system was integrated with an SOFC stack for zero-carbon electricity generation and compared with dehydrogenation systems which use only a single LOHC.

TC LOHC outperforms its single LOHC counterparts in virtually every metric besides the already-discussed outlier of ammonia. Including the temperature variability of the heat source, TC outperformed other systems in electrical energy generated per kilogram of LOHC by a factor of 1.3–2. Finally, it was shown how this integrated system can considerably increase the amount of clean energy generated (1.17–1.97 times) with a reactor system of the same complexity compared to single LOHC systems.

This leads us to draw the conclusion that the utilization of energy conservative and integrated process designs such as TC dehydrogenation can notably increase the hydrogen yield at zero-carbon emissions, thus providing a more energy-efficient process for clean hydrogen release and utilization. A detailed techno-economic assessment is required to fully define and quantify whether the benefits (increased efficiency) outweigh the possible drawbacks (more expensive LOHC transport) of



Scheme 1. LOHC Structures and Relevant Properties

Selected and color-coded LOHC candidates with their respective dehydrogenation reaction stoichiometries, structures, and H₂ storage capacities, ordered from top to bottom by descending reaction temperature.

this design. If that were the case, then this strategy may be required if the economic viability of renewable energy vectors and their faster implementation are to be achieved.

EXPERIMENTAL PROCEDURES

LOHC Selection

Considering that the purpose of LOHCs is the secure transport and storage of hydrogen, the likely candidates should possess some combination, or ideally all, of the following features: (1) high H₂ storage density (amount of H₂ it can carry per unit mass or volume); (2) preferably liquid with low volatility (for safe and convenient transport and storage); (3) preferably low reaction enthalpy and temperature (relative to other candidates); (4) high reversibility (low degradation during H₂ charging and discharging); and (5) low environmental hazard and cost.

As the central point of this work is improving the energy efficiency of the hydrogen release process using a shared energy source, the temperature gradient between the LOHC candidates' reaction temperatures was the additional feature considered in their selection. Four selected candidates (Scheme 1) were described in experimental and/or thermodynamic-computational dehydrogenation studies and were deemed viable as LOHCs.

Ammonia (Scheme 1, denoted in green) is one of the prime LOHC candidates in the literature, due to its outstanding hydrogen storage capacity (17.75 wild-type [WT]%) and a well-established synthesis process (Haber-Bosch), which is a *de facto* industry standard.²⁰ Experimental study of energy-self-sufficient ammonia dehydrogenation and subsequent electricity generation through SOFC by Cha et al.²¹ formed the basis of the present study. Although more recent research has demonstrated the possibility of using ammonia directly in a fuel cell, Zhao et al.²² note that, while promising, it still must develop to the point of the efficiency of the present fuel cell systems.

A eutectic mixture of biphenyl and diphenylmethane (Scheme 1, denoted in purple) is a novel LOHC candidate exhibiting properties similar to the somewhat popular DBT-H18.²³ Its dehydrogenation reaction conditions were determined in the Jang et al.²⁴ study and subsequently discussed applications for fuel cell integration.

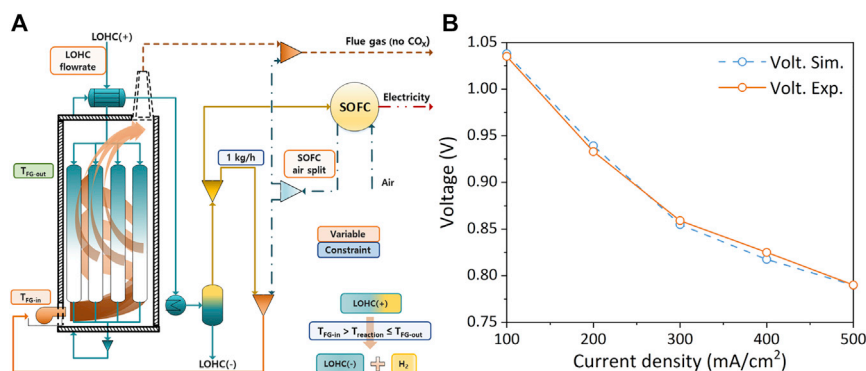


Figure 3. Optimization Logic and SOFC Validation

(A) LOHC dehydrogenation simulation setup.

(B) SOFC model validation; experimental versus simulated current-voltage (I-V) curve.

Carbazole-based LOHCs *N*-phenylcarbazole and *N*-ethylcarbazole (Scheme 1 denoted in blue and red, respectively) were assessed in a thermodynamic computational study by Shin et al.,²⁵ which was used as a basis for consideration in the present study due to their high hydrogen storage capacities and reaction temperatures, which are suitable for the TC process.

LOHC-SOFC Simulation, Validation, and Optimization Setup

The LOHC dehydrogenation and SOFC unit were simulated using Aspen Plus version 11. For dehydrogenation, a thermodynamic property method based on the Soave-Redlich-Kwong (SRK) cubic equation of state (EOS) was used as it is suitable for nonpolar mixtures such as hydrocarbons and light gases. SOFC simulation used the thermodynamic property method based on the Peng-Robinson cubic EOS due to its applicability domain, which extends to high-temperature and high-pressure regions.

SOFC was chosen as a representative hydrogen-to-electricity conversion route since it is widely accepted as a future standard for stationary clean power generation.²⁶ The experimental SOFC was a planar-type, co-flow fuel cell stack, with an active area of 18 cm², and inlet temperature and pressure conditions of 700°C and 1 atm, respectively. The air:hydrogen standard volume flow ratio was set to 4.5. Subsequently, the SOFC model was designed and validated against the experimental voltage-density measurements (Figure 3B). When scaled up linearly (achievable by increasing the number of stacks), 1 kg/h of H₂ produces ~18.04 kW of power, which translates into 54.1% SOFC efficiency and was used as a basis for the assessment of the total power produced from an LOHC system.

Single LOHC dehydrogenation (Figure 3A) was conceptualized as a plug-flow tube bundle reactor, which is heated to reaction temperature by a countercurrent flow of hot flue gas (analogous to the natural gas reformer furnace). The system was scaled to a constant 1 kg/h H₂, which was used as a fuel for the dehydrogenation. Essentially, the flowrate of fresh LOHC (+) (and consequently the produced H₂) was optimized to maximize the energy extraction from a constant fuel flowrate. Constraints ensured that the system operates within a thermodynamic reality. For the reaction to even occur, heat is required to bring the LOHC (+) to reaction temperature and to ensure enough energy to satisfy the endothermic heat of dehydrogenation.

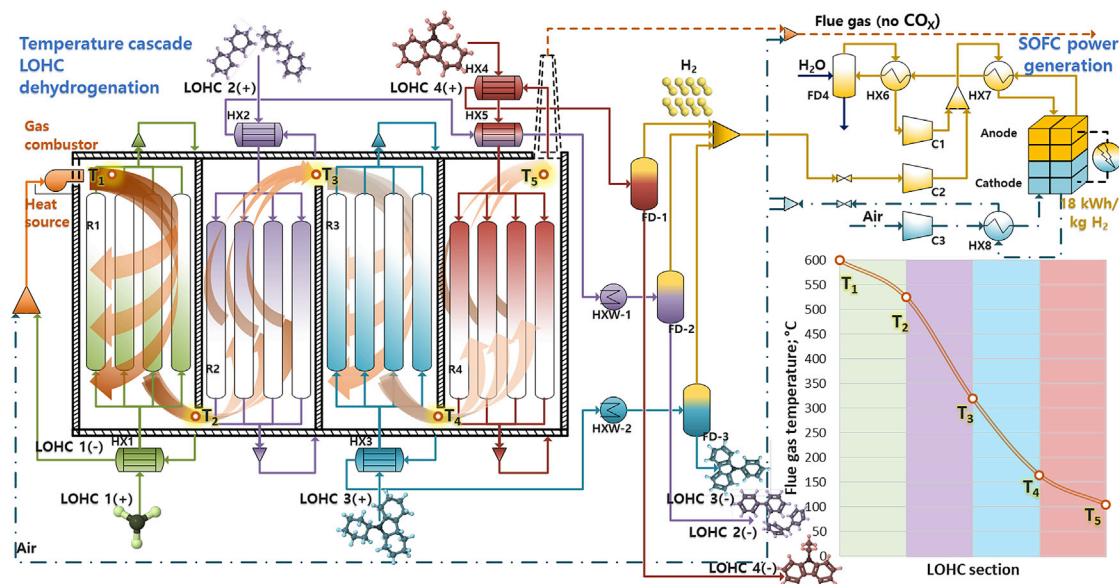


Figure 4. Main Process Design

TC LOHC dehydrogenation and SOFC electricity generation conceptual process design. The chart depicts flue gas temperature as it travels through the system. (+), H₂ charged; (-), H₂ discharged; C, compressor; FD, flash drum; HX, heat exchanger; HXW, water cooler; R, reactor.

Therefore, the flue gas temperature at the inlet must be higher than the reaction temperature of the particular LOHC system (ΔT of 50°C was set as the minimum). The flue gas temperature (and the flowrate) was controlled by varying the air:fuel ratio. The second constraint ensured that the flue gas exited the system at a temperature that was no greater than the reaction temperature of the LOHC system. The bulk of the produced hydrogen was used in an SOFC stack for direct electricity generation, and the part of the H₂ was used as a fuel. The flue gas contained no CO₂, yielding virtually zero-carbon electricity generation. It must be noted that trace amounts of dehydrogenated LOHC vapors may remain in the hydrogen stream, which would produce some CO_x when combusted. This, however, is a negligible amount when compared to fossil fuel combustion and can be further minimized with appropriate separation operations (adsorption and H₂ selective membranes, to name a few).

TC Process Design and Description

The main premise for the TC process design was the fact that the flue gas will exit the LOHC system at the temperature close to but not lower than the reaction temperature of that system. It is conceivable that one could use a combination of LOHC chemicals with high enough reaction temperature gradients so that the flue gas exiting one system would still have a high enough temperature for the dehydrogenation of the next system. This concept was designed and tested using four LOHCs (Scheme 1), which were dehydrogenated in a series (TC) using the same heat source from combusting 1 kg/h of H₂ (Figure 4). As detailed earlier, the flowrates of all LOHCs were maximized while being kept within the thermodynamic constraints for each system. Here, the air:fuel ratio was set (with reference to maximum flue gas temperature) so that the flowrate of LOHC 1 (ammonia) would produce the required 1 kg/h H₂ in a mixture with nitrogen. This was chosen since other LOHCs are comparatively much simpler to separate from the produced hydrogen (cooling + flash drum) and are recovered for subsequent charging. Ammonia, however, is non-reversible from the perspective of LOHCs and is therefore used entirely as a non-carbon heat source for a stand-alone LOHC

dehydrogenation system. LOHCs (+) (charged) 2, 3, and 4 are pre-heated in a heat exchanger by a hot mixture by hot reactor products and dehydrogenated in a temperature range tuned to that particular system (Figure 4). The LOHCs were cooled and separated from the produced hydrogen in flash drums, after which they were pumped to storage. Hydrogen from each LOHC system was combined and sent to the SOFC stack for electricity generation.

To determine the benefits of this design concept, the TC system was compared to each stand-alone LOHC system. All of the comparisons were made based on the same fuel input (1 kg/h H₂). Conductive and convective heat losses from the flue gas to the reactor and the LOHCs were not assessed for this comparison as they would equally affect the stand-alone systems and each LOHC system in the TC concept. The primary objectives of this comparison were to quantify the amount of electricity generated from each system, the amount of electricity generated per kilogram of LOHC used, and finally the amount of electricity generated per total number of reactor vessels (as a potential indicator of reduction of complexity of the reactor system).

DATA AND CODE AVAILABILITY

All of the data associated with the study are included in the paper or are available from the authors upon reasonable request.

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AUTHOR CONTRIBUTIONS

Conceptualization & Methodology, Writing – Original Draft, Writing – Review & Editing, B.B.; Investigation & Formal Analysis, B.B., B.L., and R.D.; Validation, Software, S.K. and J.J.L.; Project Administration, B.L.; Supervision, Funding Acquisition, H.L.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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