

Lab on a Chip

Lab on a chip for a low-carbon future

Journal:	Lab on a Chip
Manuscript ID	LC-PER-01-2022-000020.R1
Article Type:	Perspective
Date Submitted by the Author:	23-Dec-2022
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Abstract.

Transitioning our society to a sustainable future, with low or net-zero carbon emissions to the atmosphere, will require a wide-spread transformation of energy and environmental technologies. In this perspective article, we describe how Lab-on-a-Chip (LoC) systems can help address this challenge by providing insight into the fundamental physical and geochemical processes underlying new technologies critical to this transition, and developing the new processes and materials required. We focus on six areas: (I) subsurface carbon sequestration, (II) subsurface hydrogen storage, (III) geothermal energy extraction, (IV) bioenergy, (V) recovering critical materials, and (VI) water filtration and remediation. We hope to engage the LoC community in the many opportunities within the transition ahead, and highlight the potential of LoC approaches to the broader community of researchers, industry experts, and policy makers working toward a low-carbon future.

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Introduction.

The 6th report of the Intergovernmental Panel on Climate Change (IPCC)¹ emphasizes that "the only path out [of climate change] demands intensive, immediate efforts to bring net-carbon emissions to zero, combined with unprecedented efforts to extract carbon from the atmosphere." The International Energy Agency (IEA) Net Zero by 2050 report² provides some clarity on how this transition may be accomplished by mid-century. As noted in the IEA report, it will require "a wide-spread transformation of the energy systems that underpin our economies… [because] the energy sector is the source of around three-quarters of greenhouse gas emissions today and holds the key to averting the worst effects of climate change, perhaps the greatest challenge humankind has faced."

Lab-on-a-Chip (LoC) systems can help address this challenge by levering the attributes of high spatiotemporal resolution and excellent environmental control, and do so in two distinct ways. First, LoCs can provide insight into the fundamental physical and geochemical processes underlying many of the solutions outlined in the IEA roadmap, ranging from carbon sequestration to alternative approaches to energy storage and extraction. Second, they can be used for exploration, development, and testing of new processes and materials required to develop such solutions. LoC techniques can provide critically important insights for the design and optimization of environmental/engineered approaches and – when combined with physical upscaling approaches to translate the results of idealized microscale studies to the field – aid in our transition to a low-carbon future.

In this perspective article, which brings together speakers from a recent symposium on *Microfluidics & Energy*, we outline some of the most important opportunities for LoC systems in this future. Our goal is not to present a comprehensive overview of all the literature in this field, but rather, to highlight some areas of research we consider to be particularly promising, and outline the challenges and opportunities ahead. Some of the application areas are relatively new to the LoC community, whereas in others there is a track record of LoC contributions. In the more established areas of application, we highlight more comprehensive review articles that describe important developments using LoC systems, and instead focus on assessing the benefits provided by LoC and point to the challenges and opportunities ahead.

We first describe the use of LoC systems applied to subsurface carbon sequestration (§I) – considered a key pillar of decarbonization² – subsurface hydrogen storage (§II), as well as the related process of geothermal energy extraction (§III). We then describe applications of LoC systems for bioenergy (§IV), another key pillar in decarbonization plans. Transition to a low-carbon future will also require electrification of vehicular transport and industrial processes with a concomitant expansion of renewable electricity generation. The scaling of solar, wind, and battery production, will demand incredible material resources and present another opportunity for LoC systems: to inform the development of sustainable approaches for extracting these critical material resources (§V). Finally, we note that progress toward a low-carbon future requires advances not just in the management of greenhouse gas emissions and in the adoption of alternative energy sources, but also requires water security. Thus, we end by describing ways in which LoC systems can address challenges associated with water filtration and remediation (§VI). Microscale transport phenomena, more generally, are integrated within a wide array of relevant energy systems including fuel cells³ and electrolyzers for CO₂-reduction⁴ and hydrogen-generation⁵;

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however, we limit the scope here to chip-based approaches native to the LoC community. Likewise, we note that there are areas of importance for which the microfluidic toolkit appears to be poorly suited: for example, renewable power generation by solar, wind, and nuclear energy – all areas relevant to our low carbon future, but largely out of reach for LoC methods. Our motivation for this article is to engage the LoC community in considering the many opportunities within the transition ahead. Likewise, we highlight the potential of LoC approaches to the broader community of researchers, industry experts, and policy makers supporting the energy transition.

§I. Sequestering Carbon.

The IEA net-zero report² notes that CO₂ capture, utilization, and sequestration (CCUS) "is the only group of technologies that contributes both to reducing emissions in key sectors directly and to removing CO₂ to balance emissions that cannot be avoided", and is essential in the pursuit of net-zero. CCUS can take several forms beyond capturing and storing emissions from natural gas and from coal-fired power plants to include: the production of low-carbon hydrogen via steam methane reforming combined with CCUS (termed "blue" hydrogen), and carbon dioxide removal from the atmosphere via bioenergy with CCS (BECCS) or direct air capture (DAC) and subsequent storage. The most established means of CO₂ storage is geological, injecting CO₂ into the subsurface (Figure 1A)⁶⁻⁹. Here we outline ways in which LoC systems have provided, and continue to provide, scientific insights to inform effective approaches to geological carbon storage.

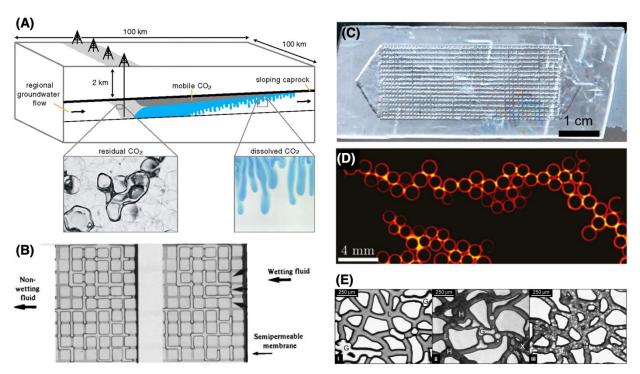


Figure 1. Lab-on-a-chip visualization of multiphase flow processes relevant to carbon sequestration. (A) Schematic of subsurface CO₂ storage describing the two key trapping mechanisms that contribute to storage capacity, residual trapping (left inset) and solubility trapping (right)¹⁰. (B) Snap-off of wetting films leading to capillary trapping of gas during imbibition¹¹. (C) Geometrically and chemically representative real-rock micromodel for reservoir engineering studies¹². (D) Snapshot from strong imbibition illustrating invasion by corner flow: the invading fluid advances by coating the perimeters of the posts rather than by filling the pore bodies; pendular rings link the coated posts, forming chains¹³. (E) Snapshots of methane hydrate

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formation in a micromodel¹⁴. i: bubbles of methane (G) and liquid (L) prior to hydrate formation. ii: newly formed hydrates (H), including encapsulated gas bubbles (X), prior to complete conversion to clathrate. iii: Hydrate redistribution after 2 days.

Multiphase flow physics underlying subsurface CO₂ storage. Geological CO₂ storage in the subsurface is controlled by the dynamics of multiphase flow through porous media^{15–17}. In particular, several of the trapping mechanisms like capillary trapping and solubility trapping depend strongly on the interplay between capillary forces, viscous forces and wettability characteristics of the porous rock. Given that most porous media (including all natural rocks) are disordered, rough and opaque, microfluidics has played a key role in advancing our understanding of multiphase flow at the pore level by enabling precise control of geometry and surface properties, and allowing direct visualization. This is perhaps best exemplified by early work employing glass-etched networks^{11,18,19} to uncover the pore-scale mechanisms responsible for fluid-fluid displacement and trapping (Fig. 1B), which allowed synthesizing the behavior of drainage (the displacement of a wetting fluid by a nonwetting fluid) in the form of a now classic phase diagram²⁰.

In addition to fluid transport through networks, CO₂ is highly reactive with minerals found naturally into the subsurface. CO2 injection into a brine-filled carbonate formation typically results in the formation of acid that dissolves carbonate minerals, forming wormholes and large channels that can affect the integrity of the rock. LoC technology has evolved and gained sufficient sophistication to provide progressively more realistic analogues of geologic media, including so-called 2.5D chips²¹, transparent 3D packings of particles^{22–24}, and micromodels that replicate the pore space and chemistry in actual rocks²⁵ and the pressure-temperature conditions in the subsurface^{12,26,26,27} (Fig. 1C). The reproducibility of such micromodels provides a unique opportunity to disentangle the effect of fluid mechanics and thermodynamics from rock/soil pore geometry and mineralogy, particularly in the context of unstable multiphase flows.^{28,29}

Given the importance of the wettability characteristics of the rock in the process of CO₂ migration and trapping^{30–32}, there has been an increased interest in extending our understanding of multiphase flow in porous media from drainage to imbibition (the displacement of a nonwetting fluid by a wetting fluid)³³. Here, once again, LoCs have been a key enabling technology allowing for precise control over the wettability characteristics, including fabrication techniques based on polymers whose surface properties can be engineering through exposure to UV light^{34,35}. These and other microfluidic fabrication protocols have then been used to elucidate wetting transitions in imbibition^{13,36,37} (Fig. 1D), help benchmark pore-scale simulation models of multiphase flow³⁸, and guide the extension of Lenormand's diagram to all wettability characteristics³⁹.

Microfluidics have provided new insights into CO_2 behavior for subsurface storage, including invasion patterns of CO_2 displacing the fluids in model porous media^{40–43}, characterizing the physical properties of CO_2 in the supercritical state^{44–46}, and screening CO_2 solubility in various solvents and brines for delivery^{47,48}. Although CO_2 mineralization occurs on geological timescales, microfluidic devices have provided new insights into how $CaCO_3$ precipitates along the transverse mixing zone, thus substantially reducing porosity and permeability, which can limit the formation of more stable carbonate phase, such as calcite^{49–52}.

Reproducing the chemical properties of subsurface media, in addition to their structure, can be necessary to assess and predict system reactivity during carbon injection and storage. This need has led to the development of functionalized microfluidics devices whose surfaces are directly coated by geomaterials⁵³. More recent advancements have enabled researchers to directly embed real rock samples in microfluidic chips, enabling observations of chemical reaction progression and resulting alterations in pore space topology in real time⁵⁴, thereby helping to elucidate feedbacks between multiphase transport and chemical reactions^{55–57}.

While microfluidics has already enabled a wealth of observations on the fluid-flow physics of carbon storage in the past two decades^{58–60}, several knowledge gaps remain. We expect that LoC systems will continue to offer a fantastic testbed to address those gaps, including in: (i) Elucidating the influence of mixed wettability, both at the sub-pore level^{61,62} and at the level of heterogenous patches involving many pores⁶³, on the flow behavior; (ii) Upscaling dynamic macroscopic multiphase flow descriptors such as relative permeability and capillary pressure^{64,65}; (iii) Examining the influence of partial dissolution⁶⁶ and compositional effects on multiphase flows in multicomponent systems, which we briefly describe next; and (iv) Studying chemical reactions (mineral dissolution and carbonate precipitation) coupled with permeability evolution in in situ CO₂ mineralization⁵¹. In each of these examples, LoC systems are particularly useful, given that they enable researchers to create repeatable experimental samples and change flow and thermodynamic conditions systematically—providing insights that could help practitioners select optimal operating conditions. Another emerging application of LoC systems is as sensors of CO₂ leakage into water, providing a potentially fast, cheap, and reliable way to monitor reservoir integrity⁶⁷.

 CO_2 Foams. CO_2 foams are being applied for to enhanced oil recovery and there is significant interest in using foams to assist with improved subsurface sequestration^{68,69}. The use of CO_2 as foam in the subsurface can control the mobility of the gas and limit the extent of fingering and gravity override²⁹. Foams in microfluidic porous media have shown to increases the volume of gas trapped in the swept zone and reduces the gas entering thief zones^{70,71}. Foams are typically generated by mixing gas with aqueous chemical surfactants and its stability is highly dependent on gas-type and capillary pressure⁷², which Is influenced by rock geometry^{73,74}. Although strong foams can be generated, they typically collapse in the presence of organics, thus limiting their transport deep into the reservoir. Microfluidic studies of foams in porous media have shown in situ foam generation mechanisms that are able to regenerate foam in porous media. In recent years, studies of foam dynamics in microchannels and porous-media chips are providing fundamental new insights into CO_2 foam formation mechanism, oil and water displacements, and the effect of permeability on CO_2 foam migration³⁵⁻³⁸. Deep learning algorithms have also been employed to process images taken during the tests, allowing the determination of quantitative information from visual monitoring³⁹.

An avenue to obtain a stable foam is the use of nanoparticles with or without conventional surfactant, as nanoparticles adsorb irreversibly at the gas-liquid interface⁷⁵. This extraordinary property of nanoparticles enables the formation of strong foams even for gas fractions as large as 97 vol.% of CO_2 —a feature that has sparked the exploration of CO_2 foams in applications like geologic CO_2 storage, geothermal energy production, and enhanced oil recovery^{76–79}.

Traditionally, identification of the optimal conditions for the foam-nanoparticle system requires costly and time-consuming experiments in conventional column-flood setups. The possibility of conducting chemical screening experiments using microfluidic systems is therefore an attractive alternative.

In gas-injection operations where the gas phase is delivered as a foam and it is supposed to react with the liquid and the rock, the knowledge of mass transfer kinetics from gas-bubble to the liquid is critical. This is the case of carbon sequestration in deep aquifers or fractured rocks where CO_2 is injected as a foam, dissolves into the liquid phase, and in some cases it can react with the rock minerals. To study mass transfer kinetics at reservoir conditions⁸⁰, high-pressure microfluidic systems have been built. In the application by Martin Ho *et al.* (2021) the analysis of the evolution of single CO_2 bubbles³² allowed to conclude that CO_2 mass transfer kinetics at

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reservoir conditions is controlled by the Reynolds number and independent from the pressure, contrarily to CO₂ mass transfer kinetics at atmospheric conditions⁸¹.

Given this tremendous potential, we expect that moving forward, LoC systems will provide useful insights with which to address the remaining challenges: (i) Elucidating nanoparticle-foam dynamics, including quantitatively describing coalescence and trapping of the bubbles and nanoparticle interaction with the gas-liquid interface; (ii) Exploring foam dynamics in porous media of varying and mixed wettabilities akin to natural systems; (iii) Monitoring the injection process to ensure safety and validate models; and (iv) screening surface-active materials, such as surfactants, nanoparticles, or combinations of them, to select for optimal compositions and concentrations to guide larger-scale testing at reservoir conditions. Future research should focus on these next challenges and provide a deep understanding of nanoparticle-foam evolution and transport in porous media.

CO₂ and Methane Hydrates. A gas hydrate is an ice-like solid that forms from a gas-water mixture under sufficiently high pressure and low temperature. For CO₂ and methane (CH₄), the conditions for hydrate stability are reached in the ocean floor at depths of about 500 meters and deeper, or in permafrost regions. Methane, in particular, is a potent greenhouse gas, yet methane fluxes in both terrestrial and marine settings are poorly understood, raising fundamental questions about the role of methane hydrates in past and future climatic change⁸². Experimental observations using microfluidics have provided important insights into the mode of hydrate growth in multiphase gas-water systems^{14,83,84} (Fig. 1E), including the role of wettability⁸⁵. These laboratory observations, along with many field observations, suggest that most hydrate systems are out of equilibrium⁸⁶, a feature that explains the co-existence of gas and hydrate in nature⁸⁷, and points to challenges in scaling up the tantalizing proposal of simultaneous CO₂ storage and CH₄ production from natural gas hydrate⁸⁸. LoC systems can help address these challenges moving forward—for example, in improving quantitative descriptions of the kinetics of hydrate formation and dissociation in porous media, including near wellbores, where the permeability reduction due to hydrate growth may impede fluid transport. Indeed, given the stochasticity inherent in hydrate formation, LoC systems may provide a useful way to obtain statistically significant data that may otherwise be challenging to obtain^{89,90}. However, it is important to note that in field conditions, ice and hydrates—which may be challenging to visually differentiate from each other—can coexist; thus, it may be necessary to couple LoC systems with non-invasive analytical techniques such as micro-Raman spectroscopy^{91–93}.

§II. Storing Hydrogen.

The transition from fossil fuels to renewable energy sources is one of the key measures to mitigate climate change and build a sustainable, reliable, and secure energy supply system¹. However, widespread application of renewable energy will require a means of storing electrical energy on an unprecedented scale, which will likely require chemical forms of energy storage. Hydrogen (H₂) is an emission-free energy carrier that can be produced from surplus renewable electricity ("green" hydrogen), with specific energy (MJ/kg) almost three times higher than natural gas. Hence, H₂ can have a major role in the energy transition; indeed, as noted in the IEA net-zero roadmap², "The initial focus for hydrogen use in the NZE (Net Zero Emissions) plan is the conversion of existing uses of fossil energy to low-carbon hydrogen in ways that do not immediately require new transmission and distribution infrastructure. This includes hydrogen use in industry and in refineries and power plants, and the blending of hydrogen into natural gas for distribution to end-users." Available storage capacity today is, however, insufficient to balance supply and local demand for future renewable energy source implementation, and H₂

storage capacity must improve to accommodate widespread implementation. Similar to CO₂ sequestration, subsurface, porous, geological formations like saline aquifers and depleted natural gas reservoirs represent widely available large-scale H₂ storage capacity⁹.

Subsurface porous formations like saline aquifers and depleted gas reservoirs offer large working gas capacities, worldwide distribution, and were recently identified as the most cost-effective H_2 subsurface storage option⁹⁴. However, storage in porous geological formations will cause a variety of effects that impact storage capacity including H_2 migration, leakage, trapping and activation of indigenous microbial metabolic groups (Fig. 2A). Therefore, similar to the case of geological sequestration of CO_2 , understanding fluid migration patterns is crucial. Additionally, intermittent H_2 withdrawal requires new fundamental understanding of coupled subsurface H_2 flow/microbial processes. We describe here ways in which LoC systems provide scientific insights to inform effective approaches to geological H_2 storage.

Multiphase flow physics underlying subsurface H_2 storage. With scarce literature, few research projects worldwide, limited industrial testing at scale, and lack of dedicated laboratory data there is an obvious need for focused studies to assess coupled processes for subsurface H_2 storage in porous media. To our knowledge, only few laboratory studies assessing porous media H_2 flow is available at the time of writing 95–97. Relevant publications mostly use numerical simulation 98, with conceptual approaches on e.g., feasible strategies and impact of geological heterogeneity. Overall, most simulations use scaled or extrapolated flow data not specifically measured for H_2 ; this approach represents a significant uncertainty. A broader characterization of coupled parameters that determine working gas capacity, deliverability and injection rate is needed, including flow functions, biochemical reactions and biotic gas consumption and conversion. An even more basic uncertainty that also needs to be addressed is whether H_2 permeates through caprocks that are known to be effectively sealing to CO_2 and CH_4 : are they sealing to CO_2 and if not, what is the leakage rate? Despite the extensive knowledge developed on CO_2 storage in the past decades, the unique physical, chemical, and molecular properties of CO_2 render its dynamics and interactions with geologic seals, the host rock, the brine and resident fluids largely unknown.

Flow functions and physically unrecoverable gas largely determines working gas capacity, deliverability and injection rates. Residual gas trapping, relevant for saline aquifer H₂ storage and frequently discussed for CO₂ sequestration 16,99, is not described in the context of H₂ subsurface porous media storage. Residual trapping occurs when the injected gas breaks into discontinuous bubbles that becomes trapped, reducing working gas capacity as residually trapped H₂ is not available for withdrawal. Disconnected bubbles may reconnect during subsequent H₂ injection, described as hysteresis. This is particularly important when establishing a H₂ storage site. Hysteresis is rarely discussed for H₂ subsurface porous media, although described for nano-porous carbon tubes 100. Contact angle measurements improve understanding of wettability and capillary pressure and relative permeability hysteresis. Hydrogen contact angles are often still not appropriately investigated, with lack of consistent and systematic approaches. Hydrogen contact angles were derived for basaltic 101 and measured for quartz 102 and sandstone¹⁰³ rocks. Overall, pore-scale displacement and trapping mechanisms are widely investigated for CO₂ storage¹⁰⁴⁻¹⁰⁶, but remains largely unaddressed for hydrogen. To the best of our knowledge, the first systematic experimental study on a H₂/water/sandstone rock system at the core scale (mm/cm scale) under shallow and deep aquifer pressure and temperature conditions was only performed recently%, providing useful X-ray CT visualization of the flow behavior of hydrogen in initially water saturated rock for both drainage and imbibition. The authors also determined the wettability of the system and identified complex displacement patterns including Lab on a Chip Page 8 of 29

gravity segregation and enhancement of spreading. These forms of direct pore scale observations of hydrogen displacement and trapping mechanisms at the sub-mm scale are crucial for understanding of hydrogen flow physics during underground storage. Storage in depleted gas reservoirs must account for H₂ mixing and interaction with CH₄, and dedicated H₂ flow functions with increasing CH₄ share are needed. Additionally, a fundamental understanding of the impact of injection-withdrawal cycles on trapping, mixing and reactions is necessary. As demonstrated in the context of the formation of chemical hotspots due to intermittent oxygen delivery in fractured rocks¹⁰⁷, temporal cycling may lead to subsurface biogeochemistry to be spatially and temporally dynamic. New modular designs of microfluidics allowing the creation of temporally varying inlet concentration signals¹⁰⁸ could provide a way to elucidate the impact of H₂ cycling on many important biogeophysicochemical processes.

Microbial effects. Microbial activity determines H₂ loss in the subsurface ¹⁰⁹, and increased understanding of microbial processes (activity, growth rates and gas conversion) is crucially needed. Depleted gas reservoirs and saline aquifers are not sterile environments (microbial life occurs up to 120°C without thresholds for pressure or brine salinity), and contain a variety of indigenous or anthropogenically introduced anaerobic microorganisms (both bacteria and archaea) that use H₂ as electron donor. Methanogenic, acetogenic, and sulphate-reducing bacteria are the most important groups¹¹⁰ as they can convert H₂ to CH₄, generate hydrogen sulfide (H₂S) that renders produced gases toxic and unusable, influence water chemistry, and reduce injection rates. Many microbial issues for hydrocarbon production, CO₂ sequestration and geothermal storage are known^{111–113}, whereas biogeochemical reactions relevant for subsurface H₂ storage are poorly understood and often only inferred from shallow environments like lakes or soil. Hence, little is known about the extent of H₂ loss in subsurface porous media for specific microbial groups, and how they are influenced by high H₂ partial pressures. It is known that higher H₂ partial pressure in shallow environments can strongly inhibit H₂-related enzymes and growth¹¹⁴; it is not known if microbial activity is suppressed during subsurface H₂ storage. Hence, LoC approaches to studying microbial activity in porous media (such as that shown in Fig. 2B-C ¹¹⁵, as well as those further detailed in §IV and other recent publications^{116–118}) will be critical to addressing these uncertainties.

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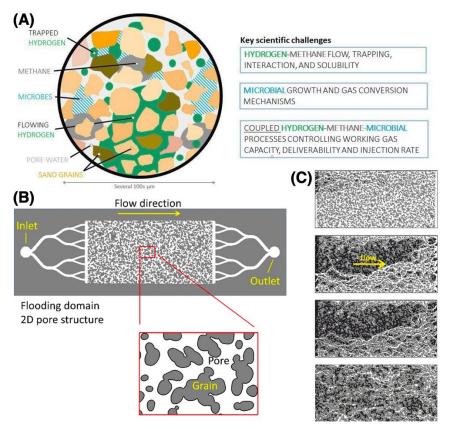


Figure 2. Lab-on-a-chip visualization of multiphase flow processes relevant to hydrogen storage. (A) Schematic of subsurface H₂ storage describing the underlying pore-scale multi-phase interactions. (B) Example of a 2D microfluidic chip for studying the relationship between microbial growth and hydraulic properties at the pore scale¹¹⁵. (C) Images from the chip in (B) showing the phase distribution in time steps during a nutrient flooding (NF) experiment in the total flooding domain; top to bottom after 16, 23, 29, and 40 h of NF, respectively. The images are segmented in three phases: white is the open pore space, light gray denotes the grains, and black is the biomass.

§III. Enabling geothermal energy.

Geothermal energy—the accessible thermal energy resource in the subsurface—holds the potential to provide 150 GWe of electrical energy by 2050, or ~ 5% global electricity generation¹¹⁹. It is renewable and flexible; geothermal resources can be found globally and run continuously, with the potential to provide for baseload power needs, or respond to demand. Current geothermal electricity generation installed capacity is roughly 10% of this 2050 goal (with 16 GWe). Geothermal has a particularly important niche among renewables: it is dispatchable, and can thus be responsive to swings in demand or other supplies. In contrast, solar and wind are intermittent. Other forms of low-carbon energy generation such as nuclear energy can provide for steady, baseload, but they cannot respond to demand spikes nor can they compensate for the intermittency of solar and wind. Geothermal energy is unique in this regard.

As noted by the US Department of Energy in the description of a recent grant opportunity¹²⁰, "Only a fraction of this potential has been realized due to technical and non-technical barriers that constrain industry growth". Some of the key challenges to massively scaling geothermal energy by 2050 are developing means to extract more energy from conventional geothermal hot-spots, and the means to tap energy from the deep

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subsurface-a breakthrough that would enable both scaling and much broader application of renewable geothermal power production.

LoC systems are contributing to the expansion of geothermal energy recovery in two ways: micromodel-based study of transport in the subsurface; and thermal fluid testing and development. Here we describe these two approaches and the opportunities they present.

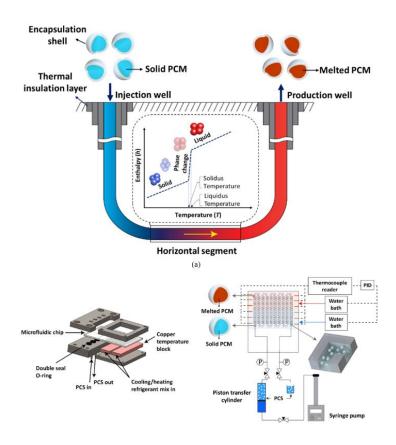


Figure 3. Lab-on-a-chip analysis of processes relevant to geothermal energy. A closed-loop geothermal system (a) is a closed system, and can benefit from specialized working fluids. The work highlighted here employs LoC methods (b) to assess the potential for a microencapsulated phase change slurry as a working fluid, with testing at temperatures, pressures and shear-rates relevant to large scale operations¹²¹.

Micromodels of geothermal reservoirs. Here the field can leverage the extensive toolkit of microfluidic methods developed for oil and gas technologies¹²², and particularly those applied to higher temperatures and pressures, such as steam-on-chip¹²³. Many enhanced geothermal sources suffer from poor fluid distribution through the subsurface, limiting the heat transfer potential. Thus, a pressing current challenge is to develop ways to control flow of fluids in subsurface geothermal reservoirs to promote heat transfer. Micromodel-based studies of fluid and heat transport, particularly in structurally heterogeneous¹²⁴ and fractured media, have yielded important insights in oil/gas recovery—and will therefore be particularly useful in addressing these conceptually similar problems in geothermal settings. For example, micromodels provide a unique opportunity to screen the utility of methods traditionally used for conformance control in oil/gas recovery (e.g., use of gels¹²⁵, polymers^{126,127},

foams¹²²) in promoting fluid mixing, homogeneous transport, and heat transfer from hot rock to address this challenge in geothermal energy.

Thermal fluid testing and development. Efforts to develop advanced thermal working fluids can benefit from the characteristic control and speed offered by LoC testing, but with a new emphasis—thermal properties. To date, LoC approaches have focused on chemical, biological and to a lesser extent, physical properties of fluids. To enable the next generation of thermal working fluids, for geothermal and other applications, LoC approaches that can provide thermal properties with high accuracy and throughput are needed^{121,128}. This is a multifaceted fluid optimization challenge, with the full combination of thermal properties (such as heat capacity) and physical properties (such as density and viscosity) and other factors such as stability and cost influencing the ultimate applicability^{121,128}. LoC systems can provide critically important data with which to navigate this broad landscape of physical, chemical and thermal properties.

Looking ahead, there are additional opportunities to expand the pressure and temperature range of LoCs—both for microfluidic testing methods, but also for in-field operations. One of the challenges in predicting and optimizing the power output of a geothermal plant is to estimate the flow rate and temperature based on the injection rate, fluid composition, and the structure of the medium. Seismic data for each site provide some of the basis needed for computational modeling and assessment of the viability of an operation. However, this area could be greatly accelerated with machine learning, suitably combined with the high-throughput approaches afforded by LoC systems, as in other application areas^{129–131}. In particular, the application of machine learning here will require large training data sets under controlled experimental conditions, well beyond that available currently. The development and testing of machine learning algorithms are identified as major research challenges by the US Department of Energy^{132,133}. Here, there is a clear path for LoCs to contribute as a provider of large quantities of accurate measurements to serve as training data sets. Here the community can make use of fabrication techniques, and approaches to control wetting conditions and withstand high pressures, as developed previously for the oil and gas industry¹³⁴. There are additional opportunities for LoCs in assessing upscaling (physics-based) models¹³⁵. The LoC community has many relevant technologies, as well as new opportunities, to aid in the expansion of renewable geothermal energy harvesting.

§IV. Enabling bioenergy.

Decarbonization of transportation is a critical step in reducing global greenhouse gas emissions. Electrification of light-duty vehicles is already playing a role, but not all types of transportation can be readily electrified, e.g. shipping, aviation. Fuels derived from organic matter (biofuels) are expected to contribute to the equivalent of $40-50 \text{ Mboe/day} \, ^{136,137}$, $^{\sim}8\%$ of the world's primary energy demand by 2030. Assuming an average oil production carbon intensity of $10.3 \, \text{gCO}_2/\text{MJ} \, ^{138}$, the annual volume of biofuels would represent an addition $1 \, \text{GT} \, \text{CO}_2$ equivalent of emissions were these barrels produced from fossil fuels. Biofuels represent a sustainable fuel option that could help close the carbon cycle: plants utilize CO_2 to produce organic compounds that can be converted into hydrocarbons suitable for ethanol, biodiesel, and other fuels. If applied with carbon capture and sequestration, energy production from biofuels (bioenergy) has potential for net-negative emissions—but a great deal of technological innovation is needed to realize that potential (Fig. 4).

One outstanding issue in first generation biofuels is that, in practice, crops use valuable land and water resources at large scale; ways to address this demand are therefore of critical importance, particularly because

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most of the crops used are food sources as well. Indeed, while widespread application of chemical fertilizers enabled crop yields to increase fast enough to meet growing demand over the past century, such yield increases are slowing and leveling off. One way of addressing this issue is through scientific research to develop new ways to increase crop yield. As noted in the IEA Net Zero 2050 report²: "Most liquid biofuels produced today come from dedicated bioenergy crops such as sugarcane, corn or oil crops, often known as conventional biofuels. The expanded use of feedstocks and arable land to produce these biofuels can conflict with food production." Thus, in the roadmap proposed by the IEA, there needs to be "a shift towards the use of sustainable, certified agricultural products and wood... also on advanced bioenergy feedstocks, including waste streams from other processes, short-rotation woody crops and feedstocks that do not require the use of arable land." However, this promising approach of using second generation biofuels is not exempt from technological challenges that still need to be resolved through scientific research; as described below, LoC systems can be of great help due to the myriad of variables to be tested.

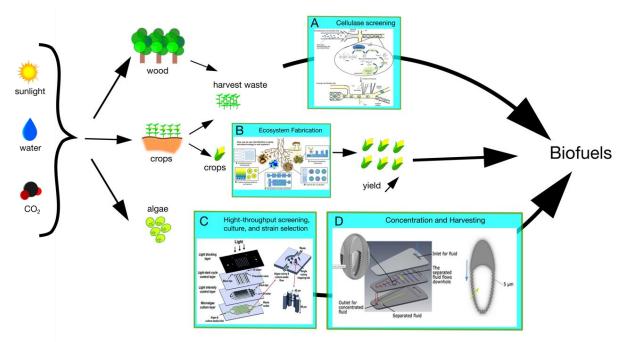


Figure 4. Lab-on-a-chip studies of processes relevant to bioenergy. A variety of feedstocks, each one having its own pathway, will contribute to the production increase to meet demand in biofuels. Lab-on-a-chip approaches can substantially help in optimizing industrial processes, for example by: (A) increasing high-throughput screening of cellulase activity¹³⁹, (B) understanding soil function and optimizing interactions¹⁴⁰, (C) easily screening multiple relevant conditions to increase microalgae yield¹⁴¹, and (D) finding the physico-chemical conditions allowing to pre-concentrate and harvest microalgae¹⁴².

Biofuels from vegetables (crops and waste). Growing understanding of the soil microbiome—the collection of microorganisms that inhabit and play key roles in the functioning of soil—has started to yield new insights into how to sustain healthy and productive soils. However, soil microbiome experiments have traditionally been laborious, time-consuming, and low-throughput. Recent developments in microfluidics have started to overcome these limitations 140,143. For example, Ecosystem Fabrication (EcoFAB) is an approach recently developed using traditional microfluidic technology to create controlled laboratory habitats for growing bioenergy plants on a

chip and thereby enable mechanistic studies of plant-microbe interactions within specific environmental conditions¹⁴⁴. By enabling new investigation of how soil microbes interact with and help sustain plant roots, and potentially how the soil microbiome can be manipulated to improve plant growth, such approaches have potential to address current limitations in bioenergy. Moving forward, integrating such microfluidic approaches with developments in e.g. experimental tools to study processes in 3D "artificial soils" will potentially enable such lab-based studies to be even more representative of processes occurring in more complex natural media.

In second generation biofuels, a key step in converting biomass to useful fuels is the degradation of lignocellulosic plant cell walls, and the subsequent hydrolysis of cellulosic macromolecules into fermentable sugars. Selection of the right enzyme cocktail using a traditional approach is labor intensive and costly. The development and use of high-throughput screening microfluidic platforms have paved the way to overcome this hurdle 139,148, with 250-fold increases in throughput compared to automated systems reported in some cases. Further work is needed to screen for the right enzyme cocktails for different uses, particularly in a directed manner. Gaining insight on the hydrolysis mechanism will allow to direct enzyme screening studies. High resolution imaging techniques, such as synchrotron radiation, combined with microfluidics have already shown the potential to improve our understanding on this path 149. The existing data are limited, and more experiments combining microfluidics and analytics are needed in this area.

Biofuels from microalgae. Algae is one alternative to conventional biofuel crops that does not require potable water or arable land; however, current production costs make algae biofuels uncompetitive in a market dominated by relatively cheap fossil fuels. Thus, further research—much of which can be done with LoC systems—is needed to develop ways to improve biofuel production from algae. Unlike crops, algae can produce large quantities of the lipids required for biofuel production without needing arable land and can be grown with non-potable water. Since algae are ~20-50% lipid by weight, they have potential to produce ~100 times more lipids per acre than traditional crop-based biofuel feedstocks. However, this potential still has not been realized due to the high energy cost of photobioreactors used to cultivate algae. Such reactors are designed to concentrate and pump CO₂, manage oxygen accumulation (which is detrimental to algae growth), and maintain set temperatures so algae can optimally perform photosynthesis and maximize biomass production. However, identifying optimal photobioreactor conditions is still a challenge; therefore, microfluidic technologies have been used to do so in a high-throughput manner, as well as to screen different microalgae for optimal production 141,141,150-154.

Most of the work performed thus far has focused on selecting strains and determining light and nutrient conditions to optimize growth rate and lipid production. The enormous number of conditions to be tested make this topic ideally suited for LoC systems to address, and certainly there is still work to be done along these lines. Nevertheless, algae production is only the first step of the overall process. The main bottleneck to make biofuel from microalgae economically viable is harvesting, which accounts for 60% of the costs¹⁴¹. Microfluidic chip designs have already been tested to preconcentrate microalgae streams, but with limited success¹⁴². A LoC approach allowing for parallelized high-throughput testing would be ideal to tackle this problem; it would allow screening of conditions leading to flocculation of the algae either by controlling physical-chemical conditions of by co-culture of different species that could induce flocculation via polymer-mediated depletion interactions, for example. Co-culture of different species could also have other benefits as rendering cultures more resilient, less prone to harmful contamination and widen the effective range of working conditions for the process.

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§V. Recovering materials for renewable energy.

Transitioning toward a low-carbon future requires the large-scale adoption of renewable energy technologies. However, this adoption requires massive volumes of critical materials, such as arsenic, gallium, germanium, indium, tellurium, aluminum, rare earth elements, cobalt, lithium, graphite, and manganese¹⁵⁵. Between 2018 and 2019, global consumption of Li increased ~ 18%, and by 2040, global demand for rare earth elements is expected to grow three to seven-fold its current (i.e., 2021) consumption^{156,157}. Current methods for production of these material resources rely on the extraction (e.g., surface mining, in situ mining) of ore and subsequent physicochemical processes (e.g., grinding, flotation, acid leaching, smelting, solvent extraction, electrolytic refining) to separate the elements of interest. Due to the mineral heterogeneity of the ore, minerals extraction and separation are energy intensive and involve massive volumes of environmental wastes. For example, the primary energy consumption required to produce titanium from ore is ~ 263 MJ/kg due to its high melting point and bond strength¹⁵⁸. Greenhouse gas emissions from mineral and metals productions, as a result, totaled ~ 3.6 × 10¹² kg CO₂e in 2018, or about 10% of total energy-related emissions that year¹⁵⁹.

Thus, a set of opportunities arise for the LoC community to help produce the material resources required for low-carbon systems. First, significant effort is required to optimize the current extraction and separation processes used to reduce energy consumed by the mining sector (~1200 TBtu/year). Extracting metals from ore involves the reactive transport of reagents and metals of interest at sub-millimeter length scales that match those of microfluidics. Importantly, LoCs are well-suited both to understand physico-chemical processes and their coupling, such as reactivity and mixing ^{12,26}, and to help develop and screen new reagent chemistries and reaction conditions to optimize the extraction process ^{134,160}. For example, Yang et al. 2020 developed a microfluidic screening method to predict and remediate acid mine drainage, an environmental waste product of the mining industry ¹⁶¹. Even more recently, Le et al. (2022) ¹⁶² applied microfluidics to investigate the performance of various solvent extraction methods for the recovery of critical minerals—minimizing the use of reactants and the time required for experiments. However, it is important to note that while LoC systems provide a fast and cost-effective way to screen multiple separation processes suitable for mineral extraction, larger-scale tests are essential for scaling up, given the limitations of LoC systems to mix fluids at small scales.

Second, new methods are needed to process and recycle existing end-of-life resources¹⁶³. Industrial wastes such as electronic devices and coal ash contain concentrated quantities of the metals that are mined. Currently, only ~17% of total e-wastes are recycled, whereas a total of ~54 Mt of e-wastes are generated annually. Circularity in the mineral economy will need improved methods for recovering metals out of electronic devices, many that are designed to be miniature in size. Here, microfluidics offers an opportunity to understand the reactive transport and reagent chemistries required to recover these metals in an environmentally benign manner. For example, geochemical microfluidics^{12,26,112,164–166} enable the visual probing of reactive transport interactions between reagents and metals in porous end-of-life materials to understand and design methods of recycling metals from industrial wastes.

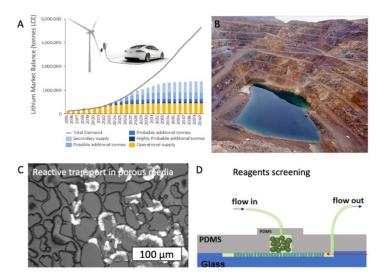


Figure 5. Opportunities for microfluidics in recovering critical mineral resources. (A) A significant demand gap is projected for mineral resources such as lithium over the next 20 years. (B) Geologic resources hosting the critical materials required. (C) Porous microfluidics (i.e., micromodels) present an opportunity to understand the reactive transport phenomena controlling minerals recovery from geologic and waste materials. (D) Microfluidic platforms provide an approach to screen reagents rapidly and economically to improve mineral recovery. Figures adapted from Benchmark Minerals Intelligence, New York Times, and refs ^{112,161}.

§VI. Ensuring water security.

Water and energy are scarce and valuable resources that are closely linked¹⁶⁷: cool and clean water is used to produce energy (e.g., cooling or hydropower production for electric power, hydraulic fracturing for oil and gas), which generates hot, contaminated water that requires additional energy-intensive purification and treatment processes (e.g., wastewater treatment, desalination). Thus, moving toward a low-carbon future also requires advances in approaches to ensuring water security. Indeed, access to clean water is already an issue faced by billions of people globally today, and unfortunately, while technologies already exist to make use of subsurface and surface water sources (aquifers, lakes, rivers, the ocean), they often have high energy demands. Thus, development of ways to remediate dirty water sources is inherently part of reaching a low-carbon future. Toward this goal, the circular economy approach in Figure 6 outlines how reuse (of waste) and repair (of contaminated resources) will become increasingly critical—necessitating improved design and optimization of the underlying processes. LoC approaches provide a way to do so, as we describe below by highlighting two key examples: remediation of contaminated sites using nanoparticles or beneficial microbes (Fig. 6E-F), and water desalination (Fig. 6B,G,I), in addition to traditional uses of LoC systems for contaminant detection, which continue to be critically important logical logicalogical logical logical logical logical logical logical logical lo

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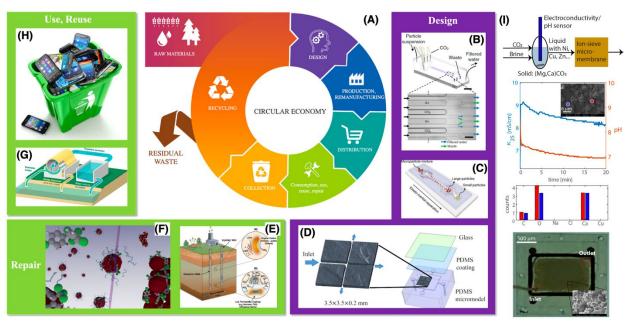


Figure 6. Opportunities for LoC approaches in ensuring water security. (A) A description of material use, reuse, repair, and design in a circular economy¹⁷⁰, along with the role of LoC. (B-D) Examples of microfluidic devices used to assess the efficacy of (B) membraneless water filtration using dissolved CO₂ ¹⁷¹; (C) membraneless water filtration using the elasto-inertial effect in a non-Newtonian fluid¹⁷²; (D) multiscale dissolution dynamics in natural rocks⁵⁴. (E) Schematic of bioremediation of contaminated water sources¹⁷³. (F) Schematic of nanoparticles used to remediate contaminated water¹⁷⁴. (G) Schematic of a seawater desalination process¹⁷⁵. (H) Photograph (from Bet_Noire/Getty Images/iStockphoto) showing materials to be recycled. (I) A microfluidic approach to studying brine desalination combined with CO₂ mineralization; top to bottom show the reactor scheme, CaCO₃ precipitation in the presence of copper (inset SEM image shows produced CaCO₃), EDS analysis of the produced CaCO₃, and 3D printed HMO-coated micro-membrane, respectively.

Groundwater remediation. More than half of the world's drinking and agriculture water supply comes from porous groundwater aquifers. However, these sources are frequently susceptible to contamination from industrial processes. In particular, non-aqueous phase liquid (NAPL) and poorly-soluble contaminants, such as perchloroethylene and trichloroethylene, persist in groundwater aquifers years after their initial introduction—in many cases taking several decades to completely dissolve—leading to negative environmental, ecological, and health consequences. For example, the environmental and ecological consequences of contamination by organic compounds like these includes enhanced morbidity of fish and local plants, ecosystem dysfunction, and reduced biodiversity. Moreover, human use of contaminated water is directly linked with diseases such as hepatitis, dysentery, and cancer. Remediating contaminated groundwater aquifers is, therefore, critical to protecting water security.

While "pump-and-treat" approaches have been used for decades, these are energy- and time-intensive, requiring continual removal of contaminated water from the source and costly annual upkeep. As a result, many groundwater sources remain contaminated for extended periods of time. Time and cost efficient in situ methods of water remediation are increasingly being sought to replace outdated pump-and-treat remedies, which are energy and time intensive, requiring complete removal of contaminated water from an aquifer and costly annual upkeep. LoC approaches provide a useful way to assess alternative approaches to groundwater remediation.

One such approach is using subsurface injection of chemically active nanoparticles, which have a large reactive surface area and are theorized to be able to travel long distances and pervade the small pores that are frequently occupied by contaminants within an aquifer. However, two challenges currently limit widespread adoption of catalytic nanomaterials for groundwater remediation. First, in many cases, aqueous suspensions of these particles aggregate quickly, reducing their overall chemical reactivity and reducing their ability to travel long enough distances to reach contaminants in an aquifer. Second, these particles do not specifically localize at trapped NAPL interfaces, reducing their remediation efficacy and requiring the addition of large amounts of nanoparticles and repeated injection at multiple sites, since contaminants can be difficult to locate. While core flood-type experiments have been instrumental in helping to shed light on these limitations, and provide a useful way to assess ways to overcome them¹⁷⁶, such approaches are often limited in their ability to probe pore-scale transport in opaque 3D porous media. Microfluidic technologies can play a useful role in shedding light. For example, micromodels can be used to develop a deeper understanding of the processes controlling how injected nanomaterials spread through a porous medium¹⁷⁷ and how they interact with a trapped non-aqueous phase¹⁷⁸. Microfluidics with functionalized surfaces particularly can help to improve the accuracy of the testing by mimicking the mineralogy of natural soil¹⁷⁹. Future studies using such approaches to screen different classes of nanomaterials for in situ groundwater remediation will be useful.

Another way to improving the delivery of reactive nanoparticles to contaminated subsurface sites is using foam as a carrier. Wet foam (or simply foam) is a complex fluid consisting of gas bubbles dispersed in a liquidphase. It is often referred to waterless fluid because the volume fraction of the gas-phase can be up to 97 vol.%. Foam is injected into the subsurface to improve the gas mobility control as it increases the effective gas viscosity and allows to divert the gas towards low permeability zones¹⁸⁰. It has been successfully employed in gas-injection operations such as enhanced oil recovery (EOR)^{181–184} and suggested as an alternative option to continuous gas injection in geological CO₂ sequestration^{78,185–190} (see §I), hydraulic fracturing for geothermal energy¹⁹¹, and shalegas extraction 192-194. Furthermore, foam has been proposed for the remediation of contaminated sites as an effective way to: deliver amendments^{195–199}, mobilize non-aqueous phase liquid contaminants²⁰⁰, and confine a source zone of contaminants in groundwater²⁰¹. In addition, since foam stability is significantly enhanced when surfactant and nanoparticles are added and nanoparticles adsorb irreversibly at gas-liquid interface, foam has been also considered as a carrier of nanoparticles towards selected targets in the subsurface²⁰²⁻²⁰⁴. However, for the description and the design of a remediation technique based on foam and nanoparticle flooding a model is required. Current models consider only foam stabilized with surfactant focusing on the description of foam texture and water/gas saturation and yet missing the description of nanoparticle migration²⁰⁵⁻²¹². To address this issue, it is necessary to gain an insight into physical and chemical processes such as surfactant and nanoparticle adsorption/desorption at the gas-liquid interface, nanoparticle interactions with a reactive porous medium in the presence of bubbles, and diversion of foam into low permeability zone in the presence of nanoparticles. To gain these insights, experiments where foam and nanoparticle transport dynamics can be observed at the pore scale should be performed. Microfluidic systems would provide valuable laboratory tools for direct observation of physical phenomena as well as more efficient and cost- effective execution than traditional column-flood tests. However, deep learning algorithms for image processing would be required to convert visual observations into quantitative information²¹³.

Yet another way of remediating contaminated groundwater aquifers by manipulating chemical reactions in situ is by using bacteria that move through the pore space towards trapped contaminant and metabolize it. While such bioremediation approaches have shown promise in groundwater remediation, their efficacy and

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widespread use has been limited by a poor understanding of how to effectively target trapped contaminants. Addressing this challenge requires fundamental studies of bacterial transport in disordered porous media. It is well known that diverse strains of bacteria can direct their swimming motion towards and metabolize trapped partly-soluble contaminants in groundwater aquifers. This process of bioremediation has been explored for several decades due to its immense promise for rapid aquifer remediation²¹⁴. However, widespread application of bioremediation has been hindered by several gaps in knowledge. Specifically, it is unknown how the time taken for bacteria, either naturally present in an aquifer or exogenously introduced, to move toward and degrade trapped contaminant depends on (i) the geometry of the pore space, (ii) the amount and spatial distribution of contaminant, (iii) the amount and spatial distribution of bacteria, (iv) the ability of the bacteria to sense and metabolize the contaminant, and (v) the ambient fluid flow. As a result, bioremediation strategies often proceed on a trial-and-error basis and, in many cases, the kinetics of aquifer remediation are too slow to yield appreciable improvement in water quality. Just as with the other examples noted in previous subsections of how LoC approaches can help shed light on microbial processes (e.g., in influencing H₂ storage, for bioenergy), micromodel studies of bacteria sensing and remediation of contaminants in situl^{147,215–217} will be important to overcome this issue.

Desalination and its products. Due to the scarcity of potable water resources, water remediation and desalination are becoming more and more critical for the supply of clean water to the world population²¹⁸. In continental semiarid and arid regions, subsurface brine is an attractive alternative to surface water and groundwater not only for drinking water use as well as for applications to agricultural fields²¹⁹. However, subsurface brine requires complex treatments that often imply significant energy-intensive chemical processes²²⁰. Therefore, microfluidic approaches play a critical role in assessing new low-carbon ways of desalinating water (e.g., Fig. 6I)^{171,221,222}.

Desalination processes conventionally employ membrane reactors because of their high selectively and separation performance without requiring chemicals and large reactor volume, unlike large-scale settling tanks and filtration columns. Thus, LoC systems are increasingly being used to study membrane processes²²³; in this context, they are of growing interest because they provides a platform for testing new materials to reduce fouling on membrane surfaces and increase permeate flux across membranes²²⁴.

LoC approaches also yield a powerful way to examine how desalination products may be useful in sustainable processes. For example, the combination of water desalination with carbon storage through ex situ CO₂ mineralization¹⁵ has been proposed to mitigate the impact of the technology and increase its sustainability^{225–227}. Indeed, the waste brine generated through desalination is a potential source for CO₂ mineralization, because in brine the concentration of divalent cations (i.e., Ca²⁺ and Mg²⁺) necessary for binding CO₂ into stable minerals, is almost twice that of ordinary seawater. For instance, the process proposed by Oh et al. (2018)²²⁵ can achieve up to 230 tonne CO₂ reduction/yr in addition to an economical benefit of water desalination. However, the precipitation kinetics of carbonate minerals changes with brine composition and an intense experimental activity is required to determine the rates. This type of investigation involves the use of comprehensive batch and continuous flow-through reactors and extensive time. Therefore, it would be envisaged to use microfluidic systems which require smaller amounts of chemicals and allow faster execution of tests. Monitoring tools including microscope and high-speed camera coupled with X-ray Diffraction (XRD), Fourier transform infrared (FTIR) and Raman spectroscopy, which are generally employed in CO₂ mineralization experiments off-line characterization, could be implemented for online monitoring.

Microfluidic systems have been used in few recent works to study carbonate precipitation^{228,229} and they have shown a significant advantage over conventional experimental approaches such as the highly reproducible control over crystal polymorph, size, and shape. Therefore, future research should focus on developing frameworks for experimental design and data processing to determine the kinetics of CO₂ mineralization using desalination brine of various chemical compositions; one such approach is shown in Fig. 6I. Indeed, conventional experimental approaches typically involve the use of large reactors and solution volumes, as well as costly monitoring tools for online and offline characterization. LoC could be uniquely helpful in miniaturizing such experiments, thereby reducing the amount of infrastructure and resources required and shorting amount of time needed to perform tests. Due to their versatility and ease of use, LoC systems could also be useful in investigating multiple systems (e.g., with different chemistries), especially when integrated with other spectroscopy, microscopy, and XRD tools.

Conclusion.

Sustainability is at the forefront of decision making in industry and governments worldwide. Although the time and length scales of LoCs are typically much smaller than conventional processes, the application and adaptation of LoCs to enable a low carbon future presents a tremendous opportunity. In many of the examples described above, LoCs provide new understanding and insights in the fundamental transport, thermodynamic, and kinetics governing processes involving sustainable energy. Combined with controlled surface chemistry and the ability to conduct high-throughput screening, microfluidics offers a path to quickly evaluate new methods and processes.

While LoC systems provide tremendous opportunities, they also have limitations. Many energy systems involving geological media are intrinsically spatially heterogeneous and characterized by multiple physicochemical processes across a broad range of length scales - LoC systems cannot capture this full complexity. How can we improve our LoC systems to be more representative of the larger-scale systems they are intended to represent? How do we build quantitative correlations/maps between what we observe and understand at the submm scale using LoC systems and what we predict at larger scales? What scale of heterogeneity should/could a microfluidic chip represent? Can we be more deliberate in designing experiments such that results can be more readily translated across scales—for example, using hierarchies of LoC systems at different scales 166? Addressing these questions is an important ongoing challenge for this field.

The many areas outlined above present a balance of precedence for LoC technologies, and new challenges. The transition period ahead, leading to 2050, corresponds to the professional lifetime of current graduate students and postdoctoral fellows. These decades present the opportunity to leverage the world's smallest fluid systems to address the world's largest fluids challenge and achieve a low-carbon future.

Acknowledgements.

SSD acknowledges funding in part from NSF grants CBET-1941716 and DMR-2011750, the Eric and Wendy Schmidt Transformative Technology Fund at Princeton, the Donors of the American Chemical Society Petroleum Research Fund for partial support of this research through grant PRF 59026-DNI9, and the New Jersey Water Resources Research Institute (NJWRRI) junior faculty grant program. MF was funded in part by the Norwegian Research Council under project 325457 – HyPE. RJ acknowledges funding from the U.S. Department of Energy (Grant No. DE-SC0018357). SP acknowledges funding from the Donors of the American Chemical Society

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Petroleum Research Fund for partial support of this research through grant PRF 62566-DNI9. VP would like to thank the American Chemical Society Petroleum Research Fund (ACS-PRF) under the grant number PRF# 57739-DNI9. WS acknowledges funding in part from DOE grants EE0009440 and FE0031791, from the Donors of the American Chemical Society Petroleum Research Fund for partial support of this research through grant PRF 61218-DNI9, and the University of Texas at Austin Energy Institute. DS acknowledges support of the Natural Sciences and Engineering Research Council of Canada (NSERC), and the Canada Research Chairs (CRC) Program.

References.

- 1 Climate Change 2021: The Physical Science Basis; IPCC AR6 WGI, 2021.
- 2 Net Zero by 2050 Analysis IEA, https://www.iea.org/reports/net-zero-by-2050, (accessed January 5, 2022).
- 3 E. Kjeang, N. Djilali and D. Sinton, *Journal of Power Sources*, 2009, **186**, 353–369.
- 4 C.-T. Dinh, T. Burdyny, M. G. Kibria, A. Seifitokaldani, C. M. Gabardo, F. P. G. de Arquer, A. Kiani, J. P. Edwards, P. D. Luna, O. S. Bushuyev, C. Zou, R. Quintero-Bermudez, Y. Pang, D. Sinton and E. H. Sargent, *Science*, 2018, **360**, 783–787.
- 5 S. M. H. Hashemi, M. A. Modestino and D. Psaltis, *Energy & Environmental Science*, 2015, **8**, 2003–2009.
- 6 A. Majumdar and J. Deutch, *Joule*, 2018, **2**, 805–809.
- 7 L. Joppa, A. Luers, E. Willmott, S. J. Friedmann, S. P. Hamburg and R. Broze, *Nature 2021 597*:7878, 2021, **597**, 629–632.
- 8 F. M. Orr Jr., *SPE Journal*, 2018, **23**, 2444–2455.
- 9 Krevor, S., de Coninck, H., Gasda, S., Ghaleigh, N. S., de Gooyert, V., Hajibeygi, H., Juanes, R., Neufeld, J., Roberts, J. J., and Swennenhuis, F., *Nature Reviews Energy & Environment*, 2023, Accepted, In press.
- 10 M. L. Szulczewski, C. W. MacMinn, H. J. Herzog and R. Juanes, *Proceedings of the National Academy of Sciences of the United States of America*, 2012, **109**, 5185–5189.
- 11 R. Lenormand, C. Zarcone and A. Sarr, *Journal of Fluid Mechanics*, 1983, 135, 337–353.
- 12 W. Song, T. W. De Haas, H. Fadaei and D. Sinton, *Lab on a Chip*, 2014, **14**, 4382–4390.
- 13 B. Zhao, C. W. MacMinn and R. Juanes, *Proceedings of the National Academy of Sciences of the United States of America*, 2016, **113**, 10251–10256.
- 14 B. Tohidi, R. Anderson, R. W. Burgass and A. B. Biderkab, *Geology*, 2001, 867.
- 15 Carbon Dioxide Capture and Storage IPCC, https://www.ipcc.ch/report/carbon-dioxide-capture-and-storage/, (accessed January 6, 2022).
- 16 R. Juanes, E. J. Spiteri, F. M. Orr and M. J. Blunt, *Water Resources Research*, 2006, **42**, 12418.
- 17 H. E. Huppert and J. A. Neufeld, *Annual Review of Fluid Mechanics*, 2014, 46, 255–272.
- 18 C. D. Tsakiroglou and A. C. Payatakes, *Advances in Colloid and Interface Science*, 1998, **75**, 215–253.
- 19 M. I. J. van Dijke, K. S. Sorbie, M. Sohrabi, D. Tehrani and A. Danesh, *All Days*, , DOI:10.2118/75192-MS.
- 20 R. Lenormand, E. Touboul and C. Zarcone, *Journal of Fluid Mechanics*, 1988, **189**, 165–187.
- 21 K. Xu, T. Liang, P. Zhu, P. Qi, J. Lu, C. Huh and M. Balhoff, *Lab on a Chip*, 2017, **17**, 640–646.

- 22 A. Anbari, H.-T. Chien, S. S. Datta, W. Deng, D. A. Weitz, J. Fan, A. Anbari, H. Chien, J. Fan, S. S. Datta, W. Deng and D. A. Weitz, *Small*, 2018, **14**, 1703575.
- 23 M. Stöhr, K. Roth and B. Jähne, *Exp Fluids*, 2003, **35**, 159–166.
- 24 P. Sharma, P. Aswathi, A. Sane, S. Ghosh and S. Bhattacharya, *Rev Sci Instrum*, 2011, **82**, 113704.
- 25 R. F. Neumann, M. Barsi-Andreeta, E. Lucas-Oliveira, H. Barbalho, W. A. Trevizan, T. J. Bonagamba and M. B. Steiner, *Sci Rep*, 2021, **11**, 11370.
- 26 M. L. Porter, J. Jiménez-Martínez, R. Martinez, Q. McCulloch, J. W. Carey and H. S. Viswanathan, *Lab on a Chip*, 2015, **15**, 4044–4053.
- 27 Y. Q. Zhang, A. Sanati-Nezhad and S. H. Hejazi, *Lab Chip*, 2018, **18**, 285–295.
- 28 A. Ferrari, J. Jimenez-Martinez, T. L. Borgne, Y. Méheust and I. Lunati, *Water Resources Research*, 2015, **51**, 1381–1400.
- 29 B. Ling, J. Bao, M. Oostrom, I. Battiato and A. M. Tartakovsky, *Advances in Water Resources*, 2017, **105**, 29–38.
- 30 D. N. Espinoza and J. C. Santamarina, Water Resources Research, 2010, 46, 7537.
- 31 S. Iglauer, Accounts of Chemical Research, 2017, 50, 1134–1142.
- 32 T.-H. M. Ho, J. Yang and P. A. Tsai, *Lab Chip*, 2021, **21**, 3942–3951.
- 33 M. Rücker, W.-B. Bartels, K. Singh, N. Brussee, A. Coorn, H. A. van der Linde, A. Bonnin, H. Ott, S. M. Hassanizadeh, M. J. Blunt, H. Mahani, A. Georgiadis and S. Berg, *Geophysical Research Letters*, 2019, **46**, 3225–3234.
- 34 D. Bartolo, G. Degré, P. Nghe and V. Studer, *Lab on a Chip*, 2008, **8**, 274–279.
- 35 B. Levaché, A. Azioune, M. Bourrel, V. Studer and D. Bartolo, *Lab on a Chip*, 2012, **12**, 3028–3031.
- 36 C. Odier, B. Levaché, E. Santanach-Carreras and D. Bartolo, *Physical Review Letters*, 2017, **119**, 208005.
- 37 T. Lan, R. Hu, Z. Yang, D.-S. Wu and Y.-F. Chen, *Geophysical Research Letters*, 2020, 47, e2020GL089682.
- 38 B. Zhao, C. W. MacMinn, B. K. Primkulov, Y. Chen, A. J. Valocchi, J. Zhao, Q. Kang, K. Bruning, J. E. McClure, C. T. Miller, A. Fakhari, D. Bolster, T. Hiller, M. Brinkmann, L. Cueto-Felgueroso, D. A. Cogswell, R. Verma, M. Prodanović, J. Maes, S. Geiger, M. Vassvik, A. Hansen, E. Segre, R. Holtzman, Z. Yang, C. Yuan, B. Chareyre and R. Juanes, *Proceedings of the National Academy of Sciences of the United States of America*, 2019, 116, 13799–13806.
- 39 B. K. Primkulov, A. A. Pahlavan, X. Fu, B. Zhao, C. W. MacMinn and R. Juanes, *Journal of Fluid Mechanics*, 2021, **923**, 34.
- 40 C. Zhang, M. Oostrom, J. W. Grate, T. W. Wietsma and M. G. Warner, *Environ. Sci. Technol.*, 2011, **45**, 7581–7588.
- 41 S. Morais, N. Liu, A. Diouf, D. Bernard, C. Lecoutre, Y. Garrabos and S. Marre, *Lab Chip*, 2016, **16**, 3493–3502.
- 42 F. Kazemifar, G. Blois, D. C. Kyritsis and K. T. Christensen, *Advances in Water Resources*, 2016, **95**, 352–368.
- 43 L. Zuo, C. Zhang, R. W. Falta and S. M. Benson, *Advances in Water Resources*, 2013, **53**, 188–197.
- 44 C. Chang, Q. Zhou, T. J. Kneafsey, M. Oostrom, T. W. Wietsma and Q. Yu, *Advances in Water Resources*, 2016, **92**, 142–158.

Lab on a Chip Page 22 of 29

- 45 B. Pinho, S. Girardon, F. Bazer-Bachi, G. Bergeot, S. Marre and C. Aymonier, *Lab Chip*, 2014, 14, 3843–3849.
- 46 H. Yoon, K. N. Chojnicki and M. J. Martinez, *Environ. Sci. Technol.*, 2019, **53**, 14233–14242.
- 47 P. N. Perera, H. Deng, P. J. Schuck and B. Gilbert, J. Phys. Chem. B, 2018, 122, 4566–4572.
- 48 J. Xu and M. T. Balhoff, Advances in Water Resources, 2022, 164, 104200.
- 49 C. Zhang, K. Dehoff, N. Hess, M. Oostrom, T. W. Wietsma, A. J. Valocchi, B. W. Fouke and C. J. Werth, *Environ. Sci. Technol.*, 2010, 44, 7833–7838.
- 50 H. Yoon, A. J. Valocchi, C. J. Werth and T. Dewers, *Water Resources Research*, DOI:10.1029/2011WR011192.
- 51 M. Nooraiepour, H. Fazeli, R. Miri and H. Hellevang, *Environ. Sci. Technol.*, 2018, **52**, 6050–6060.
- 52 H. Fazeli, M. Nooraiepour and H. Hellevang, Ind. Eng. Chem. Res., 2020, 59, 450–457.
- 53 Y. Zhang, G. Yesiloz, H. J. Sharahi, H. Khorshidian, S. Kim, A. Sanati-Nezhad and S. H. Hejazi, *Advanced Materials Interfaces*, 2019, **6**, 1900995.
- 54 B. Ling, M. Sodwatana, A. Kohli, C. M. Ross, A. Jew, A. R. Kovscek and I. Battiato, *Proceedings of the National Academy of Sciences*, 2022, **119**, e2122520119.
- 55 J. Jiménez-Martínez, J. D. Hyman, Y. Chen, J. W. Carey, M. L. Porter, Q. Kang, G. Guthrie Jr. and H. S. Viswanathan, *Geophysical Research Letters*, 2020, 47, e2020GL087163.
- 56 C. Soulaine, S. Roman, A. Kovscek and H. A. Tchelepi, *Journal of Fluid Mechanics*, 2017, **827**, 457–483.
- 57 C. Soulaine, S. Roman, A. Kovscek and H. A. Tchelepi, *Journal of Fluid Mechanics*, 2018, **855**, 616–645.
- 58 N. K. Karadimitriou and S. M. Hassanizadeh, *Vadose Zone Journal*, 2012, 11, vzj2011.0072.
- 59 N. S. K. Gunda, B. Bera, N. K. Karadimitriou, S. K. Mitra and S. M. Hassanizadeh, *Lab Chip*, 2011, **11**, 3785–3792.
- 60 A. Gerami, Y. Alzahid, P. Mostaghimi, N. Kashaninejad, F. Kazemifar, T. Amirian, N. Mosavat, M. Ebrahimi Warkiani and R. T. Armstrong, *Transp Porous Med*, 2019, **130**, 277–304.
- 61 A. R. Kovscek, H. Wong and C. J. Radke, AIChE Journal, 1993, 39, 1072–1085.
- 62 T. Hiller, J. Ardevol-Murison, A. Muggeridge, M. Schröter and M. Brinkmann, *SPE Journal*, 2018, **24**, 200–214.
- 63 Irannezhad, A., Primkulov, B. K., Juanes, R., and Zhao, B., *Physical Review Fluids*, 2023, Accepted, in Press.
- 64 J. W. Barker and S. Thibeau, SPE Reservoir Engineering, 1997, 12, 138–143.
- 65 S. E. Gasda and M. A. Celia, in *Developments in Water Science*, eds. C. T. Miller, M. W. Farthing, W. G. Gray and G. F. Pinder, Elsevier, 2004, vol. 55, pp. 793–804.
- 66 N. Liu, C. Aymonier, C. Lecoutre, Y. Garrabos and S. Marre, *Chemical Physics Letters*, 2012, **551**, 139–143.
- 67 A. Schaap, D. Koopmans, M. Holtappels, M. Dewar, M. Arundell, S. Papadimitriou, R. Hanz, S. Monk, M. Mowlem and S. Loucaides, *International Journal of Greenhouse Gas Control*, 2021, **110**, 103427.
- 68 G. Jian, C. A. Fernandez, M. Puerto, R. Sarathi, A. Bonneville and S. L. Biswal, *Journal of Petroleum Science and Engineering*, 2021, **202**, 108447.
- 69 W. R. Rossen, R. Farajzadeh, G. J. Hirasaki and M. Amirmoshiri, OnePetro, 2022.

- 70 K. Ma, R. Liontas, C. A. Conn, G. J. Hirasaki and S. L. Biswal, *Soft Matter*, 2012, **8**, 10669–10675.
- 71 S. Xiao, Y. Zeng, E. D. Vavra, P. He, M. Puerto, G. J. Hirasaki and S. L. Biswal, *Langmuir*, 2018, **34**, 739–749.
- 72 Y. Zeng, R. Farajzadeh, A. A. Eftekhari, S. Vincent-Bonnieu, A. Muthuswamy, W. R. Rossen, G. J. Hirasaki and S. L. Biswal, *Langmuir*, 2016, **32**, 6239–6245.
- 73 R. Liontas, K. Ma, G. J. Hirasaki and S. L. Biswal, *Soft Matter*, 2013, **9**, 10971–10984.
- 74 E. Vavra, M. Puerto, C. Bai, K. Ma, K. Mateen, L. Biswal and G. Hirasaki, *Journal of Colloid and Interface Science*, 2022, **621**, 321–330.
- 75 N. Yekeen, M. A. Manan, A. K. Idris, E. Padmanabhan, R. Junin, A. M. Samin, A. O. Gbadamosi and I. Oguamah, *Journal of Petroleum Science and Engineering*, 2018, **164**, 43–74.
- 76 V. Prigiobbe, A. J. Worthen, K. P. Johnston, C. Huh and S. L. Bryant, *Transport in Porous Media*, 2016, **1**, 265–285.
- 77 A. J. Worthen, H. G. Bagaria, Y. Chen, S. L. Bryant, C. Huh and K. P. Johnston, *Journal of colloid and interface science*, 2013, **391**, 142–151.
- 78 A. U. Rognmo, S. Heldal and M. A. Fernø, *Fuel*, 2018, **216**, 621–626.
- 79 H. Hosseini, J. S. Tsau, K. Shafer-Peltier, C. Marshall, Q. Ye and R. B. Ghahfarokhi, *Industrial and Engineering Chemistry Research*, 2019, **58**, 9431–9449.
- 80 T. Gothsch, C. Schilcher, C. Richter, S. Beinert, A. Dietzel, S. Büttgenbach and A. Kwade, *Microfluid Nanofluid*, 2015, **18**, 121–130.
- 81 M. Abolhasani, M. Singh, E. Kumacheva and A. Günther, Lab Chip, 2012, 12, 1611–1618.
- 82 C. D. Ruppel and J. D. Kessler, *Reviews of Geophysics*, 2017, **55**, 126–168.
- 83 S. Almenningen, J. Flatlandsmo, A. R. Kovscek, G. Ersland and M. A. Fernø, *Lab on a Chip*, 2017, **17**, 4070–4076.
- 84 L. P. Hauge, J. Gauteplass, M. D. Høyland, G. Ersland, A. Kovscek and M. A. Fernø, *International Journal of Greenhouse Gas Control*, 2016, **53**, 178–186.
- 85 A. Touil, D. Broseta and A. Desmedt, *Langmuir*, 2019, **35**, 12569–12581.
- 86 X. Fu, L. Cueto-Felgueroso and R. Juanes, *Physical Review Letters*, 2018, **120**, 144501.
- 87 X. Fu, J. Jimenez-Martinez, T. P. Nguyen, J. William Carey, H. Viswanathan, L. Cueto-Felgueroso and R. Juanes, *Proceedings of the National Academy of Sciences of the United States of America*, 2020, **117**, 31660–31664.
- 88 B. Kvamme, Canadian Journal of Chemistry, 2015, 93, 897–905.
- 89 M. L. Martinez de Baños, O. Carrier, P. Bouriat and D. Broseta, *Chemical Engineering Science*, 2015, **123**, 564–572.
- 90 D. Atig, A. Touil, M. Ildefonso, L. Marlin, P. Bouriat and D. Broseta, *Chemical Engineering Science*, 2018, **192**, 1189–1197.
- 91 W. Chen, B. Pinho and R. L. Hartman, *Lab Chip*, 2017, **17**, 3051–3060.
- 92 W. Chen and R. L. Hartman, *Energy Fuels*, 2018, **32**, 11761–11771.
- 93 J. D. Wells, W. Chen, R. L. Hartman and C. A. Koh, J. Chem. Phys., 2021, 154, 114710.
- 94 R. Tarkowski, Renewable and Sustainable Energy Reviews, 2019, 105, 86–94.
- 95 A. E. Yekta, J. C. Manceau, S. Gaboreau, M. Pichavant and P. Audigane, *Transport in Porous Media*, 2018, **122**, 333–356.
- 96 M. Boon and H. Hajibeygi, *Sci Rep*, 2022, **12**, 14604.
- 97 M. Lysyy, G. Ersland and M. Fernø, Advances in Water Resources, 2022, 163, 104167.

Lab on a Chip Page 24 of 29

- 98 P. Gabrielli, A. Poluzzi, G. J. Kramer, C. Spiers, M. Mazzotti and M. Gazzani, *Renewable and Sustainable Energy Reviews*, 2020, **121**, 109629.
- 99 M. A. Hesse, F. M. Orr and H. A. Tchelepi, Journal of Fluid Mechanics, 2008, 611, 35-60.
- 100 S. H. Barghi, T. T. Tsotsis and M. Sahimi, *International Journal of Hydrogen Energy*, 2014, **39**, 1390–1397.
- 101 A. Al-Yaseri and N. K. Jha, *Journal of Petroleum Science and Engineering*, 2021, **200**, 108387.
- 102 S. Iglauer, M. Ali and A. Keshavarz, *Geophysical Research Letters*, 2021, **48**, e2020GL090814.
- 103 L. Hashemi, W. Glerum, R. Farajzadeh and H. Hajibeygi, *Advances in Water Resources*, 2021, **154**, 103964.
- 104M. Buchgraber, A. R. Kovscek, L. M. Castanier, M. Buchgraber, · A R Kovscek and L. M. Castanier, *Transport in Porous Media 2012 95:3*, 2012, **95**, 647–668.
- 105 S. C. Cao, S. Dai and J. Jung, *International Journal of Greenhouse Gas Control*, 2016, **44**, 104–114.
- 106C. Chang, Q. Zhou, M. Oostrom, T. J. Kneafsey and H. Mehta, *Advances in Water Resources*, 2017, **100**, 14–25.
- 107 O. Bochet, L. Bethencourt, A. Dufresne, J. Farasin, M. Pédrot, T. Labasque, E. Chatton, N. Lavenant, C. Petton, B. W. Abbott, L. Aquilina and T. Le Borgne, *Nat. Geosci.*, 2020, **13**, 149–155.
- 108B. Ling and I. Battiato, Micromachines, 2022, 13, 774.
- 109 V. Reitenbach, L. Ganzer, D. Albrecht and B. Hagemann, *Environmental Earth Sciences*, 2015, **73**, 6927–6937.
- 110S. P. Gregory, M. J. Barnett, L. P. Field and A. E. Milodowski, *Microorganisms 2019, Vol. 7, Page 53*, 2019, **7**, 53.
- 111 L. Pellizzari, T. Lienen, M. Kasina and H. Würdemann, *Environmental Earth Sciences*, 2017, **76**, 1–19.
- 112 W. Song, F. Ogunbanwo, M. Steinsbø, M. A. Fernø and A. R. Kovscek, *Lab on a Chip*, 2018, **18**, 3881–3891.
- 113 R. L. Tyne, P. H. Barry, M. Lawson, D. J. Byrne, O. Warr, H. Xie, D. J. Hillegonds, M. Formolo, Z. M. Summers, B. Skinner, J. M. Eiler and C. J. Ballentine, *Nature 2021* 600:7890, 2021, **600**, 670–674.
- 114E. W. J. Van Niel, P. A. M. Claassen and A. J. M. Stams, *Biotechnology and Bioengineering*, 2003, **81**, 255–262.
- 115 N. Hassannayebi, B. Jammernegg, J. Schritter, P. Arnold, F. Enzmann, M. Kersten, A. P. Loibner, M. Fernø and H. Ott, *Transport in Porous Media*, 2021, **139**, 579–593.
- 116Combining Fluidic Devices With Microscopy And Flow Cytometry To Study Microbial Transport In Porous Media Across Spatial Scales Video, https://www.jove.com/t/60701/combining-fluidic-devices-with-microscopy-flow-cytometry-to-study, (accessed December 22, 2022).
- 117P. de Anna, A. A. Pahlavan, Y. Yawata, R. Stocker and R. Juanes, *Nat. Phys.*, 2021, **17**, 68–73
- 118G. Ceriotti, S. M. Borisov, J. S. Berg and P. de Anna, *Environ. Sci. Technol.*, 2022, **56**, 17471–17480.

- 119 E. Jolie, S. Scott, J. Faulds, I. Chambefort, G. Axelsson, L. C. Gutiérrez-Negrín, S. Regenspurg, M. Ziegler, B. Ayling, A. Richter and M. T. Zemedkun, *Nature Reviews Earth & Environment 2021 2:5*, 2021, **2**, 324–339.
- 120 Department of Energy (DOE) Office of Energy Efficiency and Renewable Energy (EERE) Innovative Methods to Control Hydraulic Properties of Enhanced Geothermal Systems Funding Opportunity Announcement (FOA) Number: DE-FOA-0002498, https://eere-exchange.energy.gov/FileContent.aspx?FileID=3062faca-bd37-4795-a1d0-7a287d10406c, (accessed January 6, 2022).
- 121 V. Soni, S. Saber, H. McPhee, J. Riordon, M. Zargartalebi, M. Holmes, M. Toews and D. Sinton, *Energy Fuels*, 2021, **35**, 10293–10302.
- 122 C. A. Conn, K. Ma, G. J. Hirasaki and S. L. Biswal, *Lab on a chip*, 2014, 14, 3968–3977.
- 123 T. W. De Haas, H. Fadaei, U. Guerrero and D. Sinton, Lab on a Chip, 2013, 13, 3832-3839.
- 124 Y. Zhang, C. Zhou, C. Qu, M. Wei, X. He and B. Bai, *Lab on a Chip*, 2019, **19**, 4071–4082.
- 125 B. Bai, Y. Liu, J.-P. Coste and L. Li, *All Days*, DOI:10.2118/89468-MS.
- 126C. A. Browne and S. S. Datta, Science Advances, 2021, 7, 2619.
- 127 C. A. Browne, A. Shih and S. S. Datta, Small, 2020, 16, 1903944.
- 128 J. Zhong, V. Soni, S. Saber, J. Riordon, B. Schwarz, M. Toews and D. Sinton, *Energy Fuels*, 2020, **34**, 11219–11226.
- 129 K. Lee, S. E. Kim, J. Doh, K. Kim and W. K. Chung, Lab on a Chip, 2021, 21, 1798–1810.
- 130 H. Cai, Z. Ao, Z. Wu, S. Song, K. Mackie and F. Guo, Lab on a Chip, 2021, 21, 2194–2205.
- 131 K. M. Graczyk and M. Matyka, *Scientific Reports* 2020 10:1, 2020, **10**, 1–11.
- 132 Using Machine Learning to Predict Future Temperature Outputs in Geothermal Systems (Conference) | OSTI.GOV, https://www.osti.gov/biblio/1710149, (accessed January 6, 2022).
- 133 Energy Department Awards \$5.5 Million to Apply Machine Learning to Geothermal Exploration | Department of Energy, https://www.energy.gov/eere/articles/energy-department-awards-55-million-apply-machine-learning-geothermal-exploration, (accessed January 6, 2022).
- 134 V. A. Lifton, *Lab on a Chip*, 2016, **16**, 1777–1796.
- 135 B. Ling, M. Oostrom, A. M. Tartakovsky and I. Battiato, *Physics of Fluids*, 2018, **30**, 076601.
- 136 Total Energy Outlook 2020.
- 137 Bp, Energy Outlook 2020 edition.
- 138 M. S. Masnadi, H. M. El-Houjeiri, D. Schunack, Y. Li, J. G. Englander, A. Badahdah, J. C. Monfort, J. E. Anderson, T. J. Wallington, J. A. Bergerson, D. Gordon, J. Koomey, S. Przesmitzki, I. L. Azevedo, X. T. Bi, J. E. Duffy, G. A. Heath, G. A. Keoleian, C. McGlade, D. Nathan Meehan, S. Yeh, F. You, M. Wang and A. R. Brandt, *Science*, 2018, 361, 851–853.
- 139 R. Ostafe, R. Prodanovic, W. L. Ung, D. A. Weitz and R. Fischer, *Biomicrofluidics*, 2014, **8**, 041102.
- 140 K. Aleklett, E. T. Kiers, P. Ohlsson, T. S. Shimizu, V. E. Caldas and E. C. Hammer, *The ISME Journal 2018 12:2*, 2017, **12**, 312–319.
- 141 H. S. Kim, T. L. Weiss, H. R. Thapa, T. P. Devarenne and A. Han, *Lab on a Chip*, 2014, **14**, 1415–1425.
- 142 B. K. Hønsvall, D. Altin and L. J. Robertson, *Bioresource Technology*, 2016, **200**, 360–365.
- 143 C. E. Stanley and M. G. A. van der Heijden, *Trends in Microbiology*, 2017, 25, 610–613.

Lab on a Chip Page 26 of 29

- 144 J. Gao, J. Sasse, K. M. Lewald, K. Zhalnina, L. T. Cornmesser, T. A. Duncombe, Y. Yoshikuni, J. P. Vogel, M. K. Firestone and T. R. Northen, *Journal of Visualized Experiments: JoVE*, 2018, **2018**, 57170.
- 145 L. Ma, Y. Shi, O. Siemianowski, B. Yuan, T. K. Egner, S. V. Mirnezami, K. R. Lind, B. Ganapathysubramanian, V. Venditti and L. Cademartiri, *Proceedings of the National Academy of Sciences of the United States of America*, 2019, **166**, 11063–11068.
- 146T. Bhattacharjee and S. S. Datta, *Nature Communications 2019 10:1*, 2019, **10**, 1–9.
- 147 T. Bhattacharjee, D. B. Amchin, J. A. Ott, F. Kratz and S. S. Datta, *Biophysical Journal*, 2021, **120**, 3483–3497.
- 148 M. Najah, R. Calbrix, I. P. Mahendra-Wijaya, T. Beneyton, A. D. Griffiths and A. Drevelle, *Chemistry & Biology*, 2014, **21**, 1722–1732.
- 149 M. F. Devaux, F. Jamme, W. André, B. Bouchet, C. Alvarado, S. Durand, P. Robert, L. Saulnier, E. Bonnin and F. Guillon, *Frontiers in Plant Science*, 2018, **9**, 200.
- 150 P. J. Graham, J. Riordon and D. Sinton, Lab Chip, 2015, 15, 3116-3124.
- 151 H. S. Kim, S. C. Hsu, S. I. Han, H. R. Thapa, A. R. Guzman, D. R. Browne, M. Tatli, T. P. Devarenne, D. B. Stern and A. Han, *Plant Direct*, 2017, 1, e00011.
- 152B. Ozdalgic, M. Ustun, S. R. Dabbagh, B. Z. Haznedaroglu, A. Kiraz and S. Tasoglu, *Biotechnology and Bioengineering*, 2021, **118**, 1716–1734.
- 153 S. Mishra, Y. J. Liu, C. S. Chen and D. J. Yao, *Energies 2021, Vol. 14, Page 1817*, 2021, **14**, 1817.
- 154E. E. Jung, M. Kalontarov, D. F. R. Doud, M. D. Ooms, L. T. Angenent, D. Sinton and D. Erickson, *Lab on a Chip*, 2012, **12**, 3740–3745.
- 155 E. Alonso, A. M. Sherman, T. J. Wallington, M. P. Everson, F. R. Field, R. Roth and R. E. Kirchain, *Environmental Science and Technology*, 2012, **46**, 3406–3414.
- 156Mineral Commodity Summaries 2020 | U.S. Government Bookstore, https://bookstore.gpo.gov/products/mineral-commodity-summaries-2020, (accessed January 6, 2022).
- 157 The Role of Critical Minerals in Clean Energy Transitions Analysis IEA, https://www.iea.org/reports/the-role-of-critical-minerals-in-clean-energy-transitions, (accessed January 6, 2022).
- 158U. Department of Energy, .
- 159 M. Azadi, S. A. Northey, S. H. Ali and M. Edraki, *Nature Geoscience 2020 13:2*, 2020, **13**, 100–104.
- 160 T. W. de Haas, B. Bao, H. Acosta Ramirez, A. Abedini and D. Sinton, *Energy Fuels*, 2021, **35**, 7866–7873.
- 161 D. Yang, R. Fan, C. Greet and C. Priest, *Environmental Science and Technology*, 2020, **54**, 14000–14006.
- 162 T. N. Q. Le, Q. D. Tran, N. N. Tran, C. Priest, W. Skinner, M. Goodsite, C. Spandler, N. J. Cook and V. Hessel, *Green Chem.*, 2022, **24**, 8879–8898.
- 163 V. Forti, C. P. Balde, R. Kuehr and G. Bel, *United Nations University (UNU)/United Nations Institute for Training and Research (UNITAR) co-hosted SCYCLE Programme, International Telecommunication Union (ITU) & International Solid Waste Association (ISWA), Bonn/Geneva/Rotterdam*, 2020, 1–119.
- 164 A. Gerami, P. Mostaghimi, R. T. Armstrong, A. Zamani and M. E. Warkiani, *International Journal of Coal Geology*, 2016, **159**, 183–193.

- 165 A. L. Harrison, G. M. Dipple, W. Song, I. M. Power, K. U. Mayer, A. Beinlich and D. Sinton, *Chemical Geology*, 2017, **463**, 1–11.
- 166B. Ling, H. J. Khan, J. L. Druhan and I. Battiato, *Energies 2021, Vol. 14, Page 21*, 2020, **14**, 21.
- 167E. Metzger, P. Reig, W. H. Wen, R. S. Young and B. Owens, .
- 168 P. K. Rai, M. Islam and A. Gupta, Sensors and Actuators A: Physical, 2022, 347, 113926.
- 169 J. Saez, R. Catalan-Carrio, R. M. Owens, L. Basabe-Desmonts and F. Benito-Lopez, *Analytica Chimica Acta*, 2021, **1186**, 338392.
- 170 Circular economy: the importance of re-using products and materials | News | European Parliament,
 - https://www.europarl.europa.eu/news/en/headlines/economy/20150701STO72956/circular-economy-the-importance-of-re-using-products-and-materials, (accessed January 6, 2022).
- 171 S. Shin, O. Shardt, P. B. Warren and H. A. Stone, *Nature Communications 2017 8:1*, 2017, **8**, 1–6.
- 172 J. Nam, H. Lim, D. Kim, H. Jung and S. Shin, Lab on a Chip, 2012, 12, 1347–1354.
- 173 K. H. Williams, J. R. Bargar, J. R. Lloyd and D. R. Lovley, *Current opinion in biotechnology*, 2013, **24**, 489–497.
- 174New study shows how nanoparticles can clean up environmental pollutants | MIT News | Massachusetts Institute of Technology, https://news.mit.edu/2015/nanoparticles-clean-environmental-pollutants-0721, (accessed January 6, 2022).
- 175 E. C. La Plante, D. A. Simonetti, J. Wang, A. Al-Turki, X. Chen, D. Jassby and G. N. Sant, *ACS Sustainable Chemistry & Engineering*, 2021, **9**, 1073–1089.
- 176T. Pak, L. F. De Lima Luz, T. Tosco, G. S. R. Costa, P. R. R. Rosa and N. L. Archilha, *Proceedings of the National Academy of Sciences of the United States of America*, 2020, **117**, 13366–13373.
- 177 N. Bizmark, J. Schneider, R. D. Priestley and S. S. Datta, *Science Advances*, 2020, **6**, 2530–2543.
- 178 J. Schneider, R. D. Priestley and S. S. Datta, *Physical Review Fluids*, 2021, 6, 014001.
- 179 S. M. Seyedpour, M. Janmaleki, C. Henning, A. Sanati-Nezhad and T. Ricken, *Science of The Total Environment*, 2019, **686**, 1272–1281.
- 180 R. F. Li, W. Yan, S. Liu, G. J. Hirasaki and C. A. Miller, SPE Journal, 2010, 15, 928–942.
- 181 W. R. Rossen, Foams, 2017, 413–464.
- 182 T. C. Ransohoff and C. J. Radke, SPE Reservoir Engineering, 1988, 3, 573–585.
- 183 G. J. Hirasaki, C. A. Miller, R. Szafranski, D. Tanzil, J. B. Lawson, H. Meinardus, M. Jin, J. T. Londergan, R. E. Jackson, G. A. Pope and W. H. Wade, DOI:10.2118/39292-MS.
- 184R. Farajzadeh, A. Andrianov, R. Krastev, G. J. Hirasaki and W. R. Rossen, *Advances in colloid and interface science*, 2012, **183–184**, 1–13.
- 185 A. J. Worthen, P. S. Parikh, Y. Chen, S. L. Bryant, C. Huh and K. P. Johnston, *Energy Procedia*, 2014, **63**, 7929–7938.
- 186 M. Lotfollahi, I. Kim, M. R. Beygi, · Andrew, J. Worthen, · Chun Huh, · Keith, P. Johnston, M. F. Wheeler and D. A. Dicarlo, *Transport in Porous Media*, 2017, **116**, 687–703.
- 187 F. Guo, S. A. Aryana, Y. Wang, J. F. McLaughlin and K. Coddington, *International Journal of Greenhouse Gas Control*, 2019, **87**, 134–141.
- 188 T. Føyen, B. Brattekås, M. A. Fernø, A. Barrabino and T. Holt, *International Journal of Greenhouse Gas Control*, 2020, **96**, 103016.

Lab on a Chip Page 28 of 29

- 189Ø. Eide, M. Fernø, S. Bryant, A. Kovscek and J. Gauteplass, *Journal of Natural Gas Science and Engineering*, 2020, **80**, 103378.
- 190 K. R. Chaturvedi and T. Sharma, Chemical Engineering Science, 2021, 235, 116484.
- 191 W. A. Nugroho, S. Hermawan, B. H. Lazuardi and R. Mirza, *Society of Petroleum Engineers SPE/IATMI Asia Pacific Oil and Gas Conference and Exhibition 2017*, , DOI:10.2118/186922-MS.
- 192 Q. Lv, Z. Li, B. Li, S. Li and Q. Sun, *Industrial and Engineering Chemistry Research*, 2015, **54**, 9456–9477.
- 193 W. A. M. Wanniarachchi, P. G. Ranjith, M. S. A. Perera, A. Lashin, N. Al Arifi and J. C. Li, *Geomechanics and Geophysics for Geo-Energy and Geo-Resources*, 2015, 1, 121–134.
- 194C. Xiao, S. N. Balasubramanian and L. W. Clapp, *Industrial and Engineering Chemistry Research*, 2017, **56**, 8340–8348.
- 195 C. N. Mulligan and S. Wang, Engineering Geology, 2006, 85, 75–81.
- 196L. Zhong, N. P. Qafoku, J. E. Szecsody, P. E. Dresel and Z. F. Zhang, *Vadose Zone Journal*, 2009, **8**, 976–985.
- 197L. Zhong, J. E. Szecsody, F. Zhang and S. V. Mattigod, *Vadose Zone Journal*, 2010, **9**, 757–767.
- 198 J. Maire, A. Coyer and N. Fatin-Rouge, *Journal of hazardous materials*, 2015, **299**, 630–638.
- 199 J. Maire, E. Brunol and N. Fatin-Rouge, *Chemosphere*, 2018, **197**, 661–669.
- 200 S. Liao, Z. Saleeba, J. D. Bryant, L. M. Abriola and K. D. Pennell, *Water research*, DOI:10.1016/J.WATRES.2021.116975.
- 201 C. Portois, C. S. Boeije, H. J. Bertin and O. Atteia, *Transport in Porous Media*, 2018, **124**, 787–801.
- 202 J. Quinn, C. Geiger, C. Clausen, K. Brooks, C. Coon, S. O'Hara, T. Krug, D. Major, W. S. Yoon, A. Gavaskar and T. Holdsworth, *Environmental science & technology*, 2005, **39**, 1309–1318.
- 203 X. Shen, L. Zhao, Y. Ding, B. Liu, H. Zeng, L. Zhong and X. Li, *Journal of Hazardous Materials*, 2011, **186**, 1773–1780.
- 204 Y. Ding, B. Liu, X. Shen, L. Zhong and X. Li, *Journal of Environmental Engineering*, 2013, **139**, 1206–1212.
- 205 O. Li and V. Prigiobbe, Transport in Porous Media, 2020, 131, 269–288.
- 206 W. R. Rossen, Journal of Colloid and Interface Science, 1990, 136, 1–16.
- 207 S. S. Mohammadi, D. A. Coombe and V. M. Stevenson, *Journal of Canadian Petroleum Technology*, 1993, **32**, 49–54.
- 208 A. R. Kovscek, T. W. Patzek and C. J. Radke, *Chemical Engineering Science*, 1995, **50**, 3783–3799.
- 209 S. I. Kam and W. R. Rossen, *Proceedings SPE Annual Technical Conference and Exhibition*, 2002, 3137–3150.
- 210 M. Kun, G. Ren, K. Mateen, D. Morel and P. Cordelier, *Proceedings SPE Symposium on Improved Oil Recovery*, 2014, **2**, 1041–1065.
- 211 M. Lotfollahi, R. Farajzadeh, M. Delshad, A. Varavei and W. R. Rossen, *Journal of Natural Gas Science and Engineering*, 2016, **31**, 184–197.
- 212Z. H. Zhou and W. R. Rossen, SPE Advanced Technology Series, 1995, 3, 154–162.
- 213 Q. Li and V. Prigiobbe, Chemical Engineering Science, 2020, 215, 115427.

- 214R. B. Marx and M. D. Aitken, *Environmental Science and Technology*, 2000, **34**, 3379–3383.
- 215 X. Wang, T. Long and R. M. Ford, *Biotechnology and Bioengineering*, 2012, **109**, 1622–1628.
- 216X. Wang, J. Atencia and R. M. Ford, *Biotechnology and Bioengineering*, 2015, **112**, 896–904
- 217 X. Wang, L. M. Lanning and R. M. Ford, *Environmental Science and Technology*, 2016, **50**, 165–172.
- 218 P. Greve, T. Kahil, J. Mochizuki, T. Schinko, Y. Satoh, P. Burek, G. Fischer, S. Tramberend, R. Burtscher, S. Langan and Y. Wada, *Nature Sustainability 2018 1:9*, 2018, **1**, 486–494.
- 219 E. Jones, M. Qadir, M. T. H. van Vliet, V. Smakhtin and S. mu Kang, *Science of The Total Environment*, 2019, **657**, 1343–1356.
- 220 D. Xevgenos, K. Moustakas, D. Malamis and M. Loizidou, *New pub: Balaban*, 2014, **57**, 2304–2314.
- 221 B. D. MacDonald, M. M. Gong, P. Zhang and D. Sinton, Lab on a Chip, 2014, 14, 681–685.
- 222 S. H. Roelofs, A. Van Den Berg and M. Odijk, *Lab on a Chip*, 2015, **15**, 3428–3438.
- 223 J. de Jong, R. G. H. Lammertink and M. Wessling, *Lab Chip*, 2006, **6**, 1125–1139.
- 224 H. S. Santana, J. L. Silva, B. Aghel and J. Ortega-Casanova, SN Appl. Sci., 2020, 2, 395.
- 225 J. Oh, D. Jung, S. H. Oh, K. Roh, J. Chung, J. I. Han and J. H. Lee, *IFAC-PapersOnLine*, 2018, **51**, 85–90.
- 226 J. H. Bang, Y. Yoo, S. W. Lee, K. Song and S. Chae, *Minerals 2017, Vol. 7, Page 207*, 2017, 7, 207.
- 227 J. H. Bang, S. C. Chae, S. W. Lee, J. W. Kim, K. Song, J. Kim and W. Kim, *Journal of CO2 Utilization*, 2019, **33**, 427–433.
- 228 A. Yashina, F. Meldrum and A. DeMello, *Biomicrofluidics*, 2012, 6, 022001.
- 229 K. Y. Ko and I. H. Kim, *Biotechnology and Bioprocess Engineering 2016 21:3*, 2016, **21**, 453–462.