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Soft alchemy: a comprehensive guide to chemical reactions for pneumatic soft actuation

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Abstract

Soft robotics has emerged as a transformative field, leveraging bio-inspired novel actuation mechanisms to enable more adaptable, compliant, and sophisticated robotic systems. However, the portability of soft pneumatic actuators is typically constrained by the tethering to bulky power sources. This review offers a thorough analysis of autonomous power alternatives facilitated by chemical reactions for gas generation and absorption, a concept analogous to biological energy conversion processes. These bio-inspired strategies propel soft pneumatic actuators towards new horizons of autonomy and portability, essential for real-world applications. This comprehensive review explores the critical intersection of gas evolution reactions (GERs) and gas consumption reactions (GCRs) as a power source for pneumatic actuation in soft robotics. We here emphasize the importance and impact of bio-inspired design, control, efficiency, safety, and sustainability within soft robotics to not only mimic biological motions but to enhance them. This review explores the fundamentals of both pneumatic and chemically powered actuation, highlighting the need for careful consideration of reaction kinetics. Additionally, this work highlights key aspects of smart materials that draw from biological structures and response mechanisms, along with state-of-the-art techniques for precise pressure modulation. Finally, we chart prospective development pathways and provide a future outlook for bio-inspired soft robotics, emphasizing the transformative impact of integrating chemical actuation methods. This exploration underlines the quest for further autonomy in soft robotic systems and points towards the future opportunities in this exciting and fast-developing field.

Keywords: Soft robotics, chemistry, pneumatic actuation, design, chemical power source, oscillating systems, chemical reactions, chemically driven pneumatic actuation



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INTRODUCTION

The evolution of robotics over the past decades has transformed the field and yielded more interactive and collaborative systems^[1-3]. From agriculture and commodities to science, medicine, and manufacturing industries, robotic advancements have become embedded as critical components in diverse sectors of human development^[4,5]. Historical events such as global pandemics and wars have underscored the significance of contingency robotic systems. Contingency systems capable of operating in hazardous environments or performing repetitive and collaborative tasks will allow humans to shift their focus to capital and knowledge-intensive industries^[6,7].

The field of soft robotics draws inspiration from nature to create systems that replicate the adaptability, fluidity, and function of biological entities, with performance approaching that of the mimicked natural systems^[8]. Beyond mimicking biological structures, soft robotics also aims to develop biocompatible systems to interact synergically with biological structures. As soft robotics and robotics evolve, we witness more autonomous, adaptable, modular, safer, and controllable systems that positively influence and benefit society^[9]. It is paramount for industry and researchers to take safety and sustainability into consideration when designing or conceptualizing new systems^[10]. By developing systems that can mimic, interact, and preserve biological systems, a complete soft robotics “triad” can be achieved^[11] [Figure 1A].

Developing systems capable of interacting with soft biological tissues (e.g., in nature to collect biological samples, or interacting with humans, as outlined below) is a major goal in soft robotics. Pneumatic soft actuators are a popular choice for developing bio-inspired systems by arranging multiple positive and negative pressure-actuated systems to mimic limbs. Hu *et al.* introduced positive and negative pressure-driven pneumatic actuators arranged in the shape of a glove system for hand rehabilitation [Figure 1B]^[12]. Recently, non-invasive pneumatic and hydraulic systems for sample collection and exploration in shallow waters have been proposed and tested^[13-15]. Sinatra *et al.* developed a soft robotic system with ultra-gentle manipulation for interacting with softer species such as jellyfish [Figure 1C]^[16]. Other systems with the ability for gentle manipulation can also interact with other natural specimens such as fruits and vegetables using pneumatic grippers. Wang *et al.* proposed a novel soft robotic gripper comprised of positive pressure-driven robotic fingers and a negative pressure-driven suction cup for apple harvesting [Figure 1D]^[17].

However, the realm of soft robotics is not limited to non-human interactions. A key research focus lies in developing implantable soft robots designed to address and prevent muscular failure-related issues. These medical conditions, including urinary incontinence (UI)^[18], muscular dystrophy (MD)^[19], sarcopenia, and other muscle-related complications^[20], arise due to disease, aging, or trauma. There is widespread interest in preventing and restoring muscular malfunctioning of damaged muscle tissue and other medical conditions related to the mobility and control of smooth or rough muscles^[21-24]. Rehabilitation gloves^[25], apparel that improves mobility^[26], and other important topics related to well-being and opportunities to increase life quality are of significant interest to soft roboticists^[27,28]. By exploring innovative soft robotic solutions, researchers aim to improve treatment and rehabilitation options for patients suffering from these debilitating conditions.

While several mechanisms drive soft robots, pneumatic soft actuators are among the most popular and researched soft actuators; their simple mechanism and compliance make them great candidates for achieving high mechanical forces (up to ~140 N)^[29] and specific control. Their reliance on changes in pressures - particularly gases - to induce deformation underpins their popularity^[30]. Among the different pressure sources commonly used to power soft actuators (see Section “Power sources”), gas evolution reactions (GERs) and gas consumption reactions (GCRs) have a promising role as innovative power sources

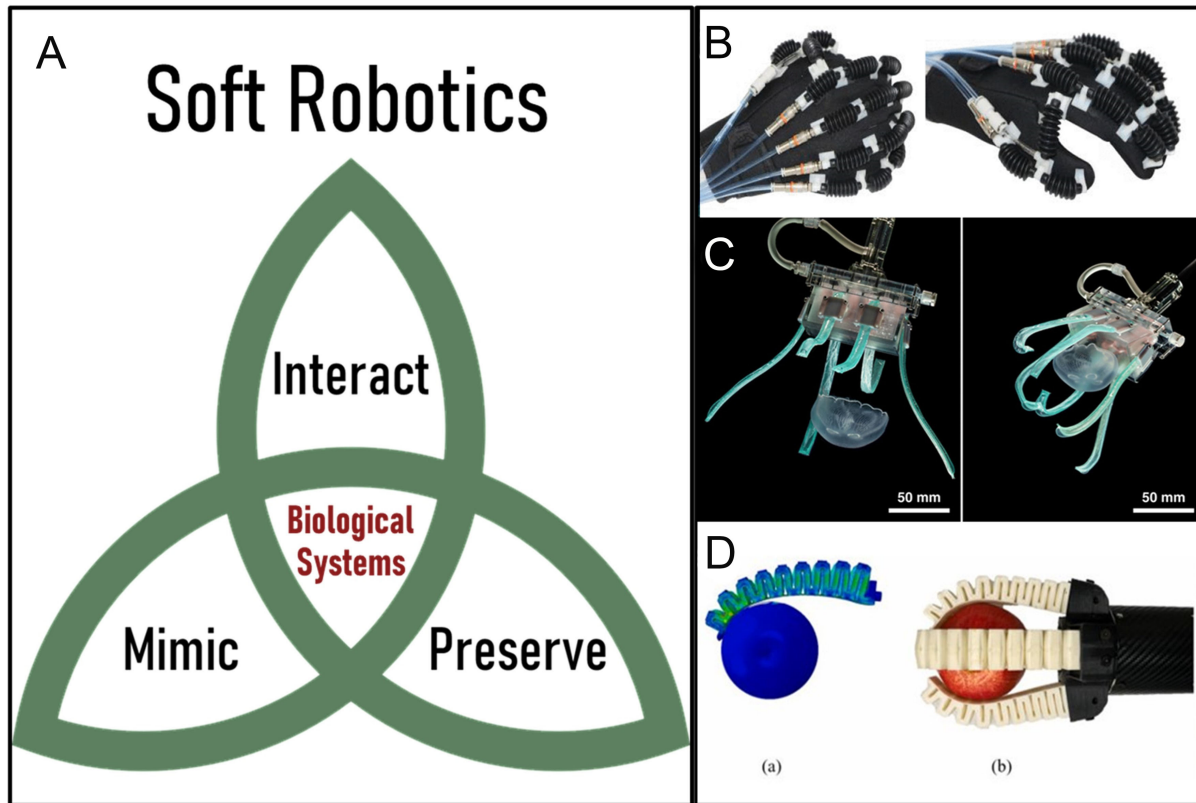


Figure 1. Soft robotics triad: factors and examples regarding (A) the safe interaction, effective mimicking, and environmental preservation for soft robotics; (B) Pneumatic gloves for movement restoration and muscular rehabilitation. Reproduced with permission^[12]. Copyright 2020, IEEE; and (C) Soft grippers for gentle biological sampling under water. Reproduced with permission^[16]. Copyright 2019, AAAS; and (D) Soft system for apple harvesting. Reproduced with permission^[17]. Copyright 2023, Elsevier. AAAS: American Association for the Advancement of Science.

for oscillating and cyclic pneumatic systems^[31]. As we believe this chemical-based approach has the potential to revolutionize the mechanism of soft pneumatic actuation, this review explores and describes the potential of implementing GERs and GCRs for chemo-driven pneumatic actuation. To aid in designing future chemo-driven pneumatic actuation systems, we also carefully consider and address some of the safety and power requirements of these novel power sources.

PNEUMATIC ACTUATION

Soft pneumatic actuators predominantly exhibit actuation strain and stress comparable to natural muscles^[32]. Moreover, a considerable number of materials employed in their fabrication possess Young's modulus values within the same range (10^5 - 10^7 Pa) as skeletal muscles^[33-36]. Owing to the gas pressure driving the actuation, soft pneumatic actuators can accomplish distinct types of movements through designed tunability [Figure 2]. By integrating and configuring elastic and inelastic regions within the design, pneumatic actuation can achieve linear^[37], bending^[38], and twisting^[39] motions when a pressure difference (ΔP) is created between the internal pressure of the soft system and the atmospheric pressure (see Table 1 for examples).

Pneumatic soft actuation has been studied for a range of applications, from human assistance to hostile terrain exploration^[47-49]. Hashem *et al.* reported an arrangement of soft pneumatic actuators as a concept for mimicking the smooth muscle contractions found, for example, in the digestive system^[50]. The arrangement

Table 1. Summary of common muscular actions and the average force and power generated in contrast with pump-driven and chemically driven soft pneumatic actuators

Action	Typical average force (N)		Ref.
	Male	Female	
	Natural muscular actions		
Force to stand (one leg)	800 ^a		
Overhead seated pull	400*	222*	[40]
Overhead standing pull	400*	244*	
Straight push seated	227*	96*	
Straight push standing	251*	140*	
Contracting quadriceps (no age range)	460 ± 159		[41]
Hand grip left (20-29)	454 ± 94	254 ± 52	[42]
Hand grip right (20-29)	486 ± 87	285 ± 64	
Anal sphincter	7		[43]
	Power (W)		
Plantar flexors extension	630 ± 13		[44]
Knee extensors	606 ± 30		
Hip extensors	2,037 ± 181		
	Soft robotics		
	Force (N)		
High-force soft pneumatic actuator	70		[45]
Chemo-driven pneumatic actuator	15		[31]

^aBased on the average weight of a person in Europe as 80 kg^[46], *no error given as taken over a range of angles for the subjects and averaged.

of bellows-type pneumatic actuators surrounding the soft tissue emulates the peristaltic contraction movements through their linear motions when pressurized. Chen *et al.* described the development and arrangement of soft grippers^[51] that mimic the bending and twisting motions of a human wrist, providing precision and fine control for tasks such as delicate goods handling.

Geometry is a fundamental part not only of the motion achieved by soft pneumatic systems but also of their control and deployment. Origami, kirigami, and kerf patterns have served as inspiration for achieving controlled and tunable actuation^[52,53]. Jin *et al.* reported soft pneumatic actuators with programmable shapes using various kirigami patterns through computational optimization strategies^[54]. Furthermore, Melancon *et al.* developed large-scale multi-stable inflatable structures that lock in place after deployment by integrating bistable origami shapes into the associated pneumatic actuators^[55]. They also developed modular origami structures with different stabilities to achieve controlled and tunable motions^[56]. The significant advances in the design and control of soft pneumatic systems, as reported by Jin *et al.*^[54] and Melancon *et al.*^[56], undeniably showcase the vast potential of these systems [Figure 3]. The ability to achieve motion, control, and deployment in these systems, particularly those designed for portability, hinges critically on the power source. A major challenge for these and related systems is, therefore, the on-demand availability of portable pneumatic power sources. As these soft pneumatic systems become more compact and efficient, the need for compatible, lightweight, and efficient power sources becomes increasingly apparent to allow wide application of this approach.

Power sources

Despite the numerous advantages of soft pneumatic systems derived from their fluid-driven nature in terms of tunability, applications and design, these systems tend to rely on heavy, rigid, and non-portable power sources^[57-59]. Commonly used conventional power sources include air compressors and pumps. These

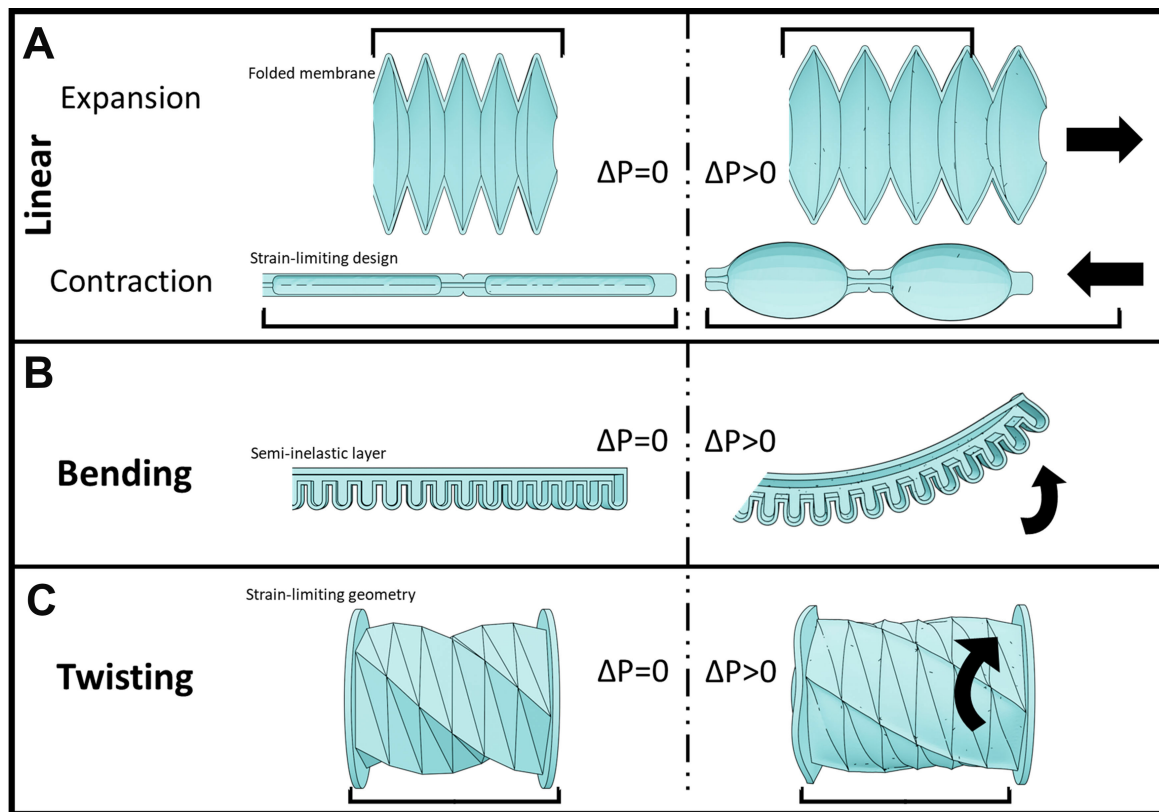


Figure 2. Schematic of the deformation motions obtained by soft pneumatic actuation. (A) Linear actuation using folded membrane and strain-limiting geometries^[37]; (B) bending actuation is achieved by the combination of a folded membrane and an inelastic region^[38]; (C) twisting actuation is obtained by the combination of strain-limiting geometries and origami folding^[39].

devices utilize energy from electrical power or fuel combustion to generate the initial mechanical force necessary for air compression. This compressed air is then used as the driving fluid for pneumatic systems or cylinders.

A further commonly used power source is pressurized cylinders. This source does not rely on external energy once it is fully pressurized, and the pressure and flow of the gas are driven by the inner pressure of the cylinder and controlled by a pressure regulator. The use of the previously described power sources limits the portability and autonomy of pneumatic soft systems, making them completely tethered systems. Electronic design advances and new manufacturing processes allow the manufacture of battery-powered onboard pumps as the first approach toward more compact and portable soft systems^[60,61]. The mechanical efficiency of soft pneumatic systems is dramatically reduced when the power sources are considered^[62]. The weights of soft pneumatic systems range from 0.01 to 1 kg, while gas cylinders and air compressors can weigh as high as 50 kg^[63-65]. When using air pumps, cylinders or micro-compressors, the final volume and weight of the set-up, considering the microcontrollers, batteries, tubing, and related components, significantly influences the produced power and energy densities of each system.

Breitman *et al.* have spearheaded efforts focused on the miniaturization and portability of pneumatic soft actuators^[66]. Through an in-depth analysis of the internal fluid mechanics of different actuator geometries, the relevance of viscosity and compressibility in miniaturized pneumatic channels was reported. Their work on actuator miniaturization contributes significantly to studying complex fluid mechanics, thereby

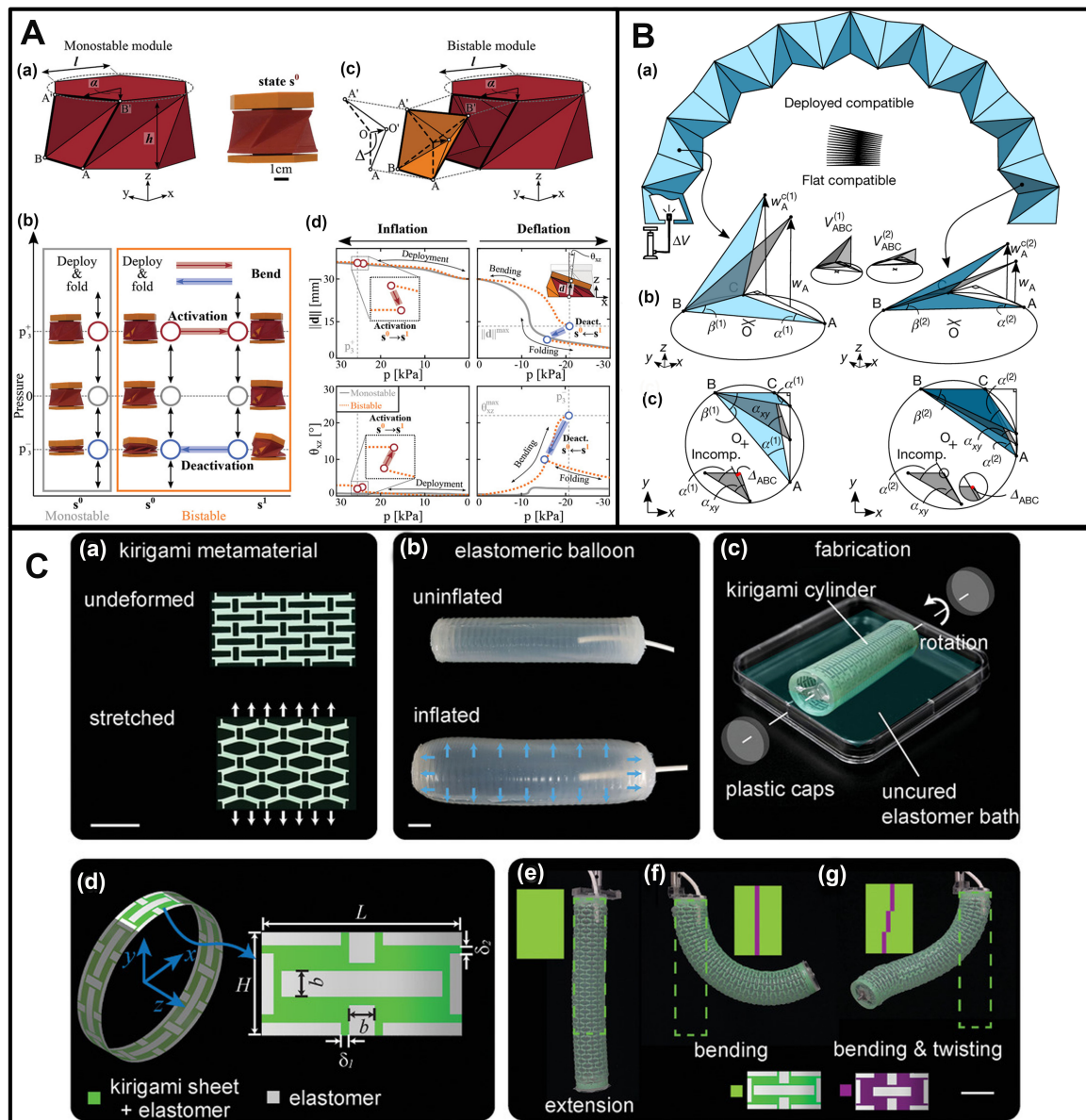


Figure 3. Geometry used as a resource for achieving tunable motions and controlled deployment. (A) and (B) portraying the use of origami patterns to move and deploy modular structures in a controlled way. Reproduced with permission^[56]. Copyright 2022, Wiley Materials. Reproduced with permission^[55]. Copyright 2021, Springer Nature; (C) shows pneumatic actuators with integrated kirigami patterns used to restrict specific regions of the actuator and achieve controlled actuation. Reproduced with permission^[54]. Copyright 2020, Wiley Materials.

influencing the optimized miniaturization of associated power sources. Microcompressors powered by batteries are a popular choice owing to various available options, each tailored to the energy and control requirements of the specific system^[60,67,68]. They work well for low-pressure systems; if larger pressures are required, a reconfiguration of multiple micro-compressors can be used, introducing more mass and volume to the total of the system. Another promising power source for pneumatic actuation is using soft electro-pneumatic pumps (EPPs) proposed by Diteesawat et al.^[69]. Using the dielectrophoretic liquid zipping (DLZ) principle, a small bead of dielectric liquid between the closest point(s) of two electrodes amplifies the

electrostatic forces. By arranging two distinct electrodes to 'zip' together when an electric charge is applied, high contraction is achieved, resulting in an EPP. The EPP, weighing only 5.3 grams, can deliver pressures up to 2.34 kPa and volumetric flow rates of 161 mL/min.

While these advances are significant, the high energy density of certain chemical reactions offers an untapped opportunity for onboard powering of pneumatic systems; this promising novel approach is explored in this review.

CHEMICALLY POWERED PNEUMATIC ACTUATION

Applying chemical reactions, specifically GERs and GCRs, has the potential to revolutionize powering pneumatic actuators by improving portability and energy density. Wehner *et al.* made a significant contribution to this approach by using the platinum nanoparticle (PtNP)-catalyzed chemical decomposition of hydrogen peroxide (H_2O_2) into O_2 and H_2O ^[70]. PtNPs were integrated using additive manufacturing techniques into the first entirely soft autonomous robot. A similar system exploring the decomposition of H_2O_2 into H_2O and O_2 as a power source was reported by Kim *et al.* using manganese dioxide (MnO_2) as the catalyst, further underscoring the potential of chemical reactions as viable power sources^[71].

While H_2O_2 -based reactions offer one set of possibilities, it is critical to also consider using negative pressures to drive soft pneumatic actuation. Fortunately, as shown in [Figure 4](#), chemical reactions are not confined to generating positive pressures; they can also be harnessed for negative pressure applications. Given this dual capability, a comprehensive understanding of the chemical energy sources and pressures generated is essential to advance this emerging field.

As recently demonstrated^[31], neutralization reactions are an untapped source to power pneumatic actuators. Owing to the small amount of starting material required, these GERs and GCRs stand out as perfect candidates for wearable and potentially even implantable, pneumatic soft actuators.

Prior to detailing the reactions that could be implemented in chemically driven pneumatic actuation, it is important to understand the factors influencing the efficiency of both GERs and GCRs such as the kinetics and thermodynamics behind the reactions, from both a safety and an actuation optimization point of view.

THEORETICAL CONSIDERATIONS

The molar volume of gas

The molar volume of a gas is dependent on the temperature and pressure of the system. In the case of an isothermal reaction, the system temperature will be constant; therefore, the initial conditions of the system can be set as room temperature and pressure (RTP) conditions. As an example, the utilized values can be defined as:

Temperature (T) = 293K;

Pressure (P) = 1 atm;

Molar volume of a gas (V_M) = 24 L·mol⁻¹

Ideal gas law

According to the ideal gas law, the pressure of the system will be given, among other parameters, by its volume and the number of moles of gas it contains.

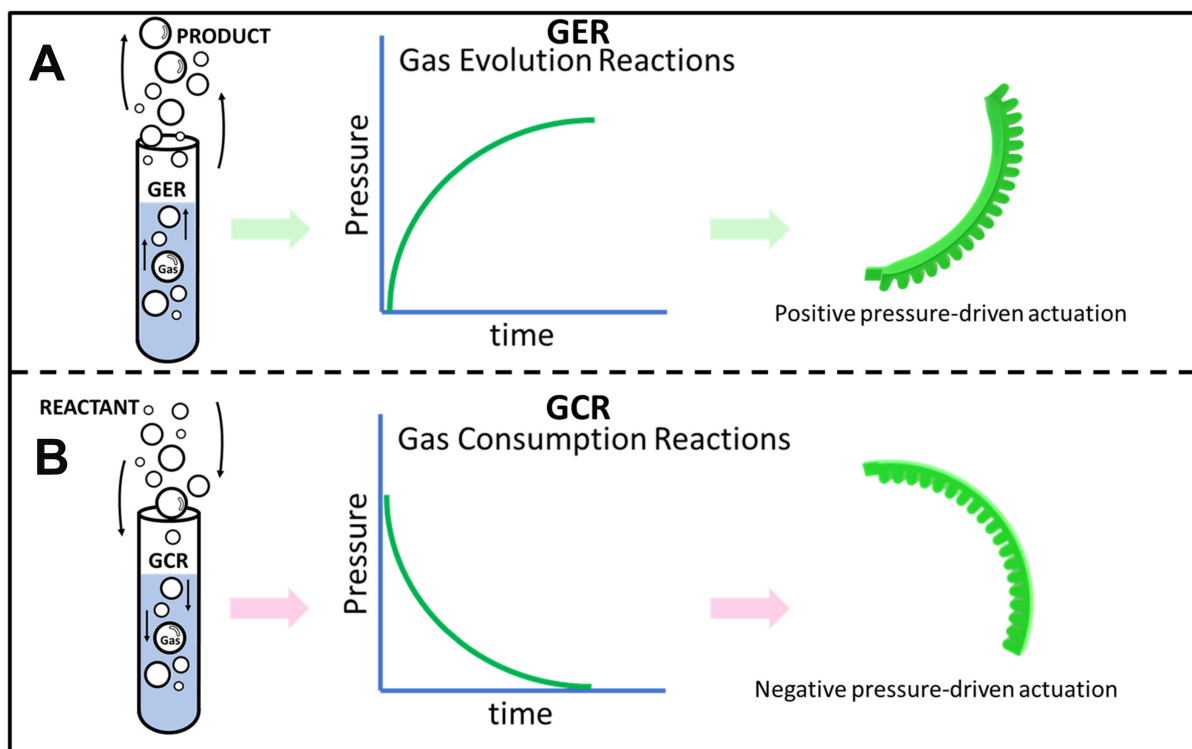


Figure 4. GERs and GCRs as power sources. (A) GERs: a gas is produced by the chemical reaction of one or more reactants, increasing pressure when the reaction takes place in a sealed system. When a soft pneumatic actuator is attached to the system, the increase in pressure results in the actuation; (B) GCRs: a gas is used as a reactant for reaction with other chemical species, resulting in the decrease of pressure when the reaction takes place in a sealed system. When a soft pneumatic actuator is attached to the system, the decrease in pressure results in actuation. GERs: Gas evolution reactions; GCRs: gas consumption reactions.

Boyle's, Gay-Lussac/Charles's, and Avogadro's laws collectively inform the behavior of gases under various conditions of pressure, volume, and temperature^[72-74]. The evolution and consumption of gases can be monitored by the change in pressure of a sealed system. By having a constant temperature and volume, the pressure change will be directly influenced by the change in the amount of moles (n) of gas resulting from the GER and GCR occurring inside the system.

The general gas equation in empirical form is expressed as:

$$PV_s = nRT$$

where P = pressure, V_s = volume of the system, n = number of moles of gas, R = universal gas constant, T = temperature.

By monitoring the system pressure during and after the GER and GCR total completion, the final number of moles of gas in the system can be calculated by:

$$n = \frac{PV_s}{RT}$$

Le Chatelier's principle

Le Chatelier's states that when a system at equilibrium is subjected to a change, it will shift its position to counteract that disturbance. In the context of GERs and GCRs, this principle is of paramount importance in predicting how changes in pressure, volume, temperature, or concentration will affect the equilibrium position of the system.

If the concentration, pressure or volume of a substance is increased, the equilibrium will shift towards the side that consumes that substance. Conversely, if the concentration, pressure or volume is decreased, the equilibrium will shift towards the side that produces that substance. Temperature changes can drastically affect the position of equilibrium, especially in endothermic and exothermic reactions. For endothermic reactions, an increase in temperature will shift the equilibrium towards the products, favoring the forward reaction. Conversely, decreasing the temperature will shift the equilibrium towards the reactants, favoring the reverse reaction.

For reactions involving the evolution or consumption of gases, changes in volume or pressure can lead to profound shifts in equilibrium:

GER: reactions that produce gas will increase the total number of gaseous moles. An increase in pressure or decrease in volume will shift the equilibrium to the side with fewer gas molecules, often favoring the reactants.

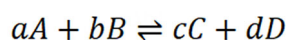
GCR: reactions that consume gases will decrease the total number of gaseous moles. An increase in pressure or decrease in volume can shift the equilibrium to the side consuming the gas, often favoring the products.

While Le Chatelier's principle provides insight into the thermodynamic aspects of a reaction with temperature changes, it is essential to remember that temperature also affects reaction rates (kinetics). A reaction may be thermodynamically favored at a particular temperature but kinetically inactive. For a simple yet elegant example, see the work on phase changes in alcohols to achieve actuation by Lee *et al.*^[75].

Reaction kinetics

Reaction kinetics is the study of the rates of chemical reactions and the factors that influence these rates. The rate of a reaction describes how quickly the concentration of reactants decreases or the concentration of products increases over time^[76].

For a general chemical reaction:



where A and B are reactants, C and D are products, and a , b , c , and d are their stoichiometric coefficients.

The rate r of the reaction can be expressed as:

$$r = -\frac{1}{a} \frac{\Delta[A]}{\Delta t} = -\frac{1}{b} \frac{\Delta[B]}{\Delta t} = \frac{1}{c} \frac{\Delta[C]}{\Delta t}$$

where t is time.

Experimentally, the rate law shows the relationship between the rate of a reaction and the concentrations of the reactants and takes the form:

$$r = k[A]^m[B]^n$$

where m and n are called reaction orders and are obtained experimentally and defined using graphical methods, distinct from the stoichiometric coefficients.

Integrated rate laws and their implications

For various reaction orders, rate laws, when integrated over time, correlate the reactant concentration to time:

$$\text{Zero order: } [A] = [A]_0 - kt$$

$$\text{First order: } \ln [A] = \ln [A]_0 - kt$$

$$\text{Second order: } \frac{1}{[A]} = \frac{1}{[A]_0} + kt$$

where k is the rate constant, t is time, and $[A]$ and $[A]_0$ are reactant concentration and concentration at $t = 0$.

Concept of half-life: half-life ($t_{1/2}$) denotes the time necessary for the concentration of the reactant to halve its initial value. For first-order reactions, it is expressed as:

$$t_{1/2} = \frac{0.693}{k}$$

It is noteworthy that the half-life remains unaffected by the starting concentration.

The Arrhenius equation and activation energy: Activation energy (E_a) signifies the minimum energy required to start a chemical reaction^[77]. The Arrhenius equation relates the rate constant, temperature, and the activation energy:

$$k = A \exp\left(\frac{E_a}{RT}\right)$$

where A is the pre-exponential factor representing a specific reaction-dependent constant.

In practice, temperature variations will influence the kinetics of the underlying chemical reactions and, consequently, the actuation behavior of these soft actuators. However, it is worth noting that the practical impact of temperature sensitivity can vary depending on factors such as the choice of reactants and the efficiency of the selected chemical reactions. For instance, as recently demonstrated by our group^[31], highly efficient reactions with low reactant consumption may exhibit minimal sensitivity to temperature changes ($\Delta T = \sim 2$ °C).

Energetic considerations

As described by Adami *et al.* for GERs and GCRs, the energy storage can be described in terms of fuel, even if not a conventional combustible fuel^[78]. To accurately describe a pressure generation method, it is essential to consider the quantity of gas that can be generated.

The net flow factor, denoted as $f_{n,v}$, is obtained based on the fuel's initial volume V_{fuel} prior to expansion and the resulting volume of gas V_{gas} after the expansion, as determined by:

$$f_{n,v} = \frac{V_{gas}}{V_{fuel}}$$

The gross flow factor $f_{g,v}$ is obtained by considering the total volume of the system V_{sys} (instead of the volume of the fuel):

$$f_{g,v} = \frac{V_{gas}}{V_{sys}}$$

To consider the varying densities of the reactants (fuel) and the fuel system, the flow capacity is also included. The net flow capacity $f_{n,m}$ is calculated (similar to the determination of the net flow factor), employing the mass of the fuel m_{fuel} . The gross flow capacity $f_{g,m}$ is calculated accordingly from the mass of the system m_{sys} :

$$f_{n,m} = \frac{V_{gas}}{m_{fuel}}, \quad f_{g,m} = \frac{V_{gas}}{m_{sys}}$$

The energy density of the reactants $\omega_{n,v}$ can then be defined as the energy E per volume of fuel, and the energy stored per fuel mass or net specific energy $\omega_{n,m}$ can be defined similarly.

$$\omega_{n,v} = \frac{E}{V_{fuel}}, \quad \omega_{n,m} = \frac{E}{m_{fuel}}$$

According to Adami *et al.*, the pressure-volume work $W_{1,2}$, of an isothermal process with the ideal gas considerations is calculated by^[78]:

$$W_{1,2} = n \cdot R \cdot T \cdot \ln\left(\frac{P_2}{P_1}\right) = P_1 \cdot V_1 \cdot \ln\left(\frac{P_2}{P_1}\right) = P_2 \cdot V_2 \cdot \ln\left(\frac{P_2}{P_1}\right)$$

As the energy leaves the GER or GCR system, the pressure-work value is considered negative. For GCRs, the pressure-volume work performed at an initial gas volume V_1 is considered to be the effective energy of reactants or fuel. The effective energy density is then calculated by:

$$\omega_{eff,v} = \frac{-W_{1,2}}{V_1} = \frac{P_1 \cdot V_1 \cdot \ln\left(\frac{P_2}{P_1}\right)}{V_1} = -P_1 \cdot \ln\left(\frac{P_2}{P_1}\right)$$

Similarly, effective specific energy for a system with solid reactants is calculated by:

$$\omega_{eff,m} = \frac{-W_{1,2}}{m_1}$$

Application of these equations will be shown in later sections of this review.

POSITIVE PRESSURE

Positive gauged pressure ($\Delta P > 0$) is the most common and accessible type of pressure when pneumatic actuation is required. In this context, a few GERs have been investigated as potential energy sources for pneumatic actuation in soft robotics. Combustion reactions involving methane and butane have been studied for their high theoretical energy densities of 1.11×10^{-2} and 1.08×10^{-2} MJ/kg, respectively^[78,79]. However, the highly explosive nature of combustion reactions requires using materials that can withstand the resulting localized elevated temperatures. These reactions generate elevated temperatures, which can range from 340 to 2,800 °C^[80,81]. Systems that employ combustion as a power source typically incorporate discrete mixing chambers and sparking sources to facilitate the reaction.

The decomposition of H_2O_2 is another popular studied chemical reaction for pneumatic actuation. Pure H_2O_2 has a specific energy of 2.9 MJ/kg, which reduces to 1.45 MJ/kg for a 50% w/v H_2O_2 solution^[78,82,83]. The O_2 evolution rate can be modulated by varying the addition rate of H_2O_2 to the catalyst.

Acid-carbonate reactions have been recently explored as an efficient power source for pneumatic soft actuators. Okui *et al.* evaluated the neutralization reaction of sodium bicarbonate ($NaHCO_3$) and citric acid as an inexpensive, nontoxic and ubiquitous alternative to power pneumatic soft actuators^[84]. The exothermic chemical reaction has a reported conversion efficiency of over 97%, meaning it can produce over 23.3 L of CO_2 per mol of starting material. Similarly, the GERs of CO_2 from acid-carbonate reactions were explored using potassium carbonate (K_2CO_3), sodium carbonate (Na_2CO_3) or calcium carbonate ($CaCO_3$), and adding hydrochloric acid (HCl)^[31]. The CO_2 evolution rates were controlled by adjusting the quantity of acid added to the carbonate-containing reservoir. Control over the reaction speed, and consequently pressure rise of GERs, provides an additional level of flexibility in the design and operation of chemically driven pneumatic actuation systems.

Other GERs, such as the hydrogen (H_2) displacement from HCl by Zn and O_2 evolution by water splitting, have also been briefly explored as means to power soft pneumatic actuators by harnessing direct gas release from the reactants^[31] or implementing mechano-electrochemical methods using electrodes^[85,86] [Table 2].

GERS

Gas, as one of the four states of matter, is characterized by the random and constant movement of molecules. The transition from solid to liquid and ultimately to gas occurs as temperature and pressure varies. The selection of appropriate reactants, which can efficiently transition to the gaseous phase and produce the desired volume of gas, is essential for the optimal performance of actuators.

Table 2. GERs previously used in soft robotics

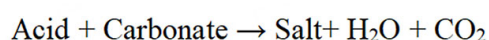
Type	Reaction	Gas evolved	Ref.
Water splitting	$2\text{H}_2\text{O} (\text{l}) \leftrightarrow 2\text{H}_2 (\text{g}) + \text{O}_2 (\text{g})$	O_2	[85]
Catalyzed decomposition of H_2O_2	$2\text{H}_2\text{O}_2 (\text{aq}) \rightarrow \text{O}_2 (\text{g}) + 2\text{H}_2\text{O} (\text{l})$	O_2	[31,70,71,82,83,87]
Methane combustion	$\text{CH}_4 (\text{l}) + 2\text{O}_2 (\text{g}) \rightarrow \text{CO}_2 (\text{g}) + 2\text{H}_2\text{O} (\text{l})$	CO_2	[88,89]
Butane combustion	$2\text{C}_4\text{H}_{10} (\text{l}) + 13\text{O}_2 (\text{g}) \rightarrow 8\text{CO}_2 (\text{g}) + 10\text{H}_2\text{O} (\text{l})$	CO_2	[79]
Acid-carbonate	$\text{C}_6\text{H}_8\text{O}_7 (\text{l}) + 3\text{NaHCO}_3 (\text{s}) \rightarrow \text{Na}_3\text{C}_6\text{H}_5\text{O}_7 (\text{aq}) + 3\text{H}_2\text{O} (\text{l}) + 3\text{CO}_2 (\text{g})$	CO_2	[84]
Acid-carbonate	$\text{K}_2\text{CO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow 2\text{KCl} (\text{aq}) + \text{CO}_2 (\text{g}) + \text{H}_2\text{O} (\text{l})$	CO_2	[31]
Acid-carbonate	$\text{Na}_2\text{CO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow 2\text{NaCl} (\text{aq}) + \text{CO}_2 (\text{g}) + \text{H}_2\text{O} (\text{l})$	CO_2	[31]
Acid-carbonate	$\text{CaCO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow \text{CaCl}_2 (\text{aq}) + \text{CO}_2 (\text{g}) + \text{H}_2\text{O} (\text{l})$	CO_2	[31]
Displacement	$\text{Zn} (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow \text{ZnCl}_2 (\text{aq}) + \text{H}_2 (\text{g})$	H_2	[31]
Reversible adsorption	$(2/x)\text{M} + \text{H}_2 \rightleftharpoons (2/x)\text{MHx}$	H_2	[86]

GERs: Gas evolution reactions.

One of the most important characteristics of soft robotics is their suitability for interacting with and mimicking biological systems. Therefore, chemical reactions that utilize or produce noxious gases such as carbon monoxide (CO), hydrogen sulfide (H_2S), ammonia (NH_3), or chlorine (Cl_2) will be briefly discussed but will not be considered as potential sources for soft pneumatic actuation. Fortunately, in contrast to the harmful gases previously discussed, various gases such as N_2 , CO_2 , or O_2 are not harmful to biological systems or even play important roles in biological processes such as respiration, photosynthesis, pH regulation and ion-mobility processes crucial for cell metabolism^[90]. Therefore, there is an extensive list of safe and appropriate chemical reactions with potential applications in soft robotics, specifically pneumatic soft actuators, as discussed below.

Acid-carbonate reactions

Acid-carbonate reactions are a type of acid-base reaction where the acid reacts with the carbonate to form a salt, H_2O , and CO_2 gas. These reactions occur through:



Most acid-carbonate reactions are exothermic reactions and will release heat, depending on the specific reactants and reaction conditions. Metal carbonates are of alkaline nature and some of the most abundant molecules on Earth, making them accessible to harvest and use within the soft robotics field^[91]. In addition, CaCO_3 has medical applications in treating and preventing osteoporosis and is also used as an antacid for indigestion^[92]. Therefore, acid-carbonate reactions are a suitable and safe type of GER to be used within bio-inspired soft systems [Table 3].

The acid-carbonate reactions would ideally run until the reactants have been consumed or limited by their stoichiometry. However, the effectiveness of these reactions depends on several factors that should be considered. Among these factors, the most practical and influencing are the acid concentration, the acid concentration at the surface of the carbonate, the reactant concentration at the center of the carbonate, and the effective diffusivity of the acid within the carbonate. These factors can be partially controlled by the spatial distribution of the reactants in the reaction chamber which should simultaneously be designed to favor the acid-carbonate chemical reaction. The chamber design should benefit the transport of the reaction products (CO_2 , H_2O , and salt) from the solid-liquid interface to the bulk fluid in an ideally controlled mass transfer step^[94,95].

Table 3. Acid-carbonate-based reactions for gas evolution^[93]

Acid-carbonate reactions
$\text{CaCO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow \text{CaCl}_2 (\text{aq}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$
$\text{MgCO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow \text{MgCl}_2 (\text{aq}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$
$\text{Na}_2\text{CO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow 2\text{NaCl} (\text{aq}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$
$\text{K}_2\text{CO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow 2\text{KCl} (\text{aq}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$
$(\text{NH}_4)_2\text{CO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow 2\text{NH}_4\text{Cl} (\text{aq}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$
$\text{BaCO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow \text{BaCl}_2 (\text{aq}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$
$\text{ZnCO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow \text{ZnCl}_2 (\text{aq}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$
$\text{CuCO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow \text{CuCl}_2 (\text{aq}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$
$\text{FeCO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow \text{FeCl}_2 (\text{aq}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$
$\text{PbCO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow \text{PbCl}_2 (\text{aq}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$
$\text{Ag}_2\text{CO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow 2\text{AgCl} (\text{s}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$
$\text{SrCO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow \text{SrCl}_2 (\text{aq}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$
$\text{Cr}_2(\text{CO}_3)_3 (\text{s}) + 6\text{HCl} (\text{aq}) \rightarrow 2\text{CrCl}_3 (\text{aq}) + 3\text{H}_2\text{O} (\text{l}) + 3\text{CO}_2 (\text{g})$
$\text{Al}_2(\text{CO}_3)_3 (\text{s}) + 6\text{HCl} (\text{aq}) \rightarrow 2\text{AlCl}_3 (\text{aq}) + 3\text{H}_2\text{O} (\text{l}) + 3\text{CO}_2 (\text{g})$
$\text{Li}_2\text{CO}_3 (\text{s}) + 2\text{HCl} (\text{aq}) \rightarrow 2\text{LiCl} (\text{aq}) + \text{H}_2\text{O} (\text{l}) + \text{CO}_2 (\text{g})$

Other GERs

Fermentation reactions are biological processes in which microorganisms break down organic compounds to produce gases, such as CO_2 and CH_4 , and other products such as alcohols, acids, and organic acids. These reactions have been widely studied and used to produce fuels, such as methane and hydrogen^[96], and in other industrial applications, such as manufacturing chemicals, biofuels, and food and beverage products. Fermentation reactions can be classified into several types, such as alcoholic fermentation, lactic acid fermentation, and acetic acid fermentation, each with distinct microorganisms and conditions to produce different by-products^[97,98]. Studies by Dagle *et al.* have shown that fermentation reactions can be a cost-effective and sustainable alternative to fossil fuels and other traditional energy sources^[99] [Table 4].

Displacement reactions involve replacing one element in a compound with another. These reactions are commonly used to generate gases such as H_2 and O_2 . One common application of displacement reactions is in producing H_2 gas, which serves as a fuel source in various industries, such as transportation and power generation. For example, the reaction between Zn and HCl generates H_2 gas and can be used in H_2 fuel cells to produce electricity^[100]. In addition to the reaction between Zn and HCl, another example of displacement reactions is in producing O_2 gas, which is applied in various industrial and medical settings. For example, the reaction between KMnO_4 and H_2SO_4 generates O_2 gas and is leveraged in O_2 generators in hospitals and submarines^[101]. Furthermore, displacement reactions are also implemented in generating Cl_2 gas, which is deployed in water treatment and disinfection. For example, the reaction between HCl and MnO_2 generates Cl_2 gas and is employed in water disinfection systems^[102] [Table 5].

Catalyzed decomposition of hydrogen peroxide

H_2O_2 is a strong oxidizing agent and is especially dangerous at high concentrations. It is crucial to handling it with care and use appropriate safety measures when working with it^[104]. Despite these risks, H_2O_2 has a wide range of applications due to its ability to decompose into H_2O and O_2 gas^[105,106]. The catalyzed decomposition of H_2O_2 is commonly used in the generation of O_2 gas and steam and has been widely employed in various fields such as rocket propulsion, water treatment, and medical sterilization.

Table 4. Fermentation GERs

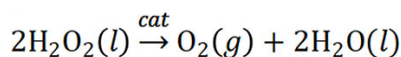
Fermentation reactions	Bacteria
$C_6H_{12}O_6(l) \rightarrow 3CH_4(g) + 3CO_2(g)$	<i>Methanogens</i>
$C_6H_{12}O_6(l) \rightarrow CH_3(CHOH)COOH(l) + CH_3(CH_2)OH(l) + CO_2(g)$	<i>Lactobacillus</i>
$C_6H_{12}O_6(l) \rightarrow 2CH_3(CH_2)OH(l) + 2H_2O(l) + 2CO_2(g)$	<i>Saccharomyces</i>
$C_{12}H_{22}O_{11}(l) + H_2O(aq) \rightarrow C_2H_5OH(l) + 4CO_2(g)$	<i>Escherichia coli</i>
$C_9H_{18}O_9(l) \rightarrow 2[CH_3(CH_2)COOH](l) + CH_3COOH(l) + H_2O(aq) + CO_2(g)$	<i>Clostridium</i>
$C_{12}H_{24}O_{12}(l) \rightarrow CH_3(CH_2)_2COOH(l) + 2(CH_3COOH)(l) + 4CO_2(g) + 6H_2(g)$	<i>Clostridium</i>

GERs: Gas evolution reactions.

Table 5. Displacement reactions for gas evolution^[103]

Displacement reactions
$Zn(s) + 2HCl(aq) \rightarrow ZnCl_2(aq) + H_2(g)$
$2HCl(aq) + MnO_2(s) \rightarrow MnCl_2(aq) + Cl_2(g) + H_2O(l)$
$Fe(s) + 2H_2SO_4(aq) \rightarrow FeSO_4(aq) + 2H_2O(l) + SO_2(g)$
$2Al(s) + 6H_2O(l) \rightarrow 2Al(OH)_3(s) + 3H_2(g)$
$Ni(s) + 2HCl(aq) \rightarrow NiCl_2(aq) + 2H_2(g)$
$Cu(s) + 4HNO_3(aq) \rightarrow Cu(NO_3)_2(aq) + 2NO_2(g) + 2H_2O(l)$
$2KMnO_4(s) + 3H_2SO_4(aq) + 5H_2O(l) \rightarrow K_2SO_4(aq) + 2MnSO_4(aq) + 8O_2(g) + 6H_2O(l)$

The decomposition of H_2O_2 in nature is typically catalyzed by enzymes such as catalase or peroxidase. These enzymes catalyze the breakdown of H_2O_2 into H_2O and O_2 gas through:



The reaction, when not catalyzed by enzymes, is highly exothermic and releases substantial amounts of energy, making it useful in generating heat and power. Among the most common catalysts used for this reaction are MnO_2 , silver (Ag), platinum (Pt), palladium (Pd), and iron (Fe).

MnO_2 is inexpensive, stable, and easy to obtain, making it a popular choice in various H_2O_2 -related fields. Ag is an effective catalyst for the H_2O_2 decomposition and is used in medical and industrial settings owing to its high catalytic activity. Pt is another highly active catalyst for the decomposition of H_2O_2 and is frequently employed in industrial contexts. Pd acts as a replacement for platinum in industrial usage. Fe, as a cheap and widely available catalyst, sees frequent use in industrial applications and water treatment.

The decomposition of H_2O_2 is also employed in the field of soft robotics for the actuation of pneumatic systems. This reaction serves as a means for inflating and deflating soft actuators, thus enabling motion in soft robotic devices. The reaction is safe, as it produces no toxic or harmful by-products. However, the storage of H_2O_2 , especially in concentrated form, poses risks to consider; lower concentrations are safer but result in decreased reactivity, potentially affecting the gas evolution performance.

Combustion

As mentioned in Section "POSITIVE PRESSURE", combustion reactions have already been explored in soft robotics as a power source for pressure-driven soft actuators. Their working principle, instead of relying on the quantity of gas produced by the reaction, is explained by the localized and momentary elevated

temperatures and, therefore, pressures reached during the combustion process. These reactions require O_2 to occur, and, as previously mentioned, the amount of gas (O_2) consumed is higher than the quantity of gas (CO_2) produced due to the formation of H_2O as a side product [Table 6]. Combustion reactions rely on heat rather than gas production to generate pressure, which is the reason that, regardless of their use and application in pneumatic soft actuators, these reactions are not considered GERs.

Thermal decomposition

Contrary to combustion reactions, thermal decomposition reactions deliver a gas surplus by the end of the reaction. Take, for example, the use of baking powder in cooking. When subjected to elevated temperatures, $NaHCO_3$ in the baking powder undergoes decomposition, releasing CO_2 , which contributes to the cake rising. Notably, the decomposition of $NaHCO_3$ commences at around $80\text{ }^\circ\text{C}$, a temperature already considered high for biological systems.

Other more industrially focused instances of thermal decomposition reactions, such as the generation of N_2 from sodium azide or the production of O_2 from mercury oxide, are known^[107]. However, these reactions occur at even higher temperatures of 275 and $500\text{ }^\circ\text{C}$, respectively.

One important aspect to highlight is the endothermic nature of thermal decomposition reactions. This necessitates considering supplemental energy sources to sustain the reaction. Furthermore, the requirement for a contained environment, especially at elevated temperatures, poses challenges in controlling these reactions. The need for precise temperature control and containment makes practically applying thermal decomposition reactions in certain settings particularly difficult [Table 7].

Vaporization

For lower temperature ranges, the evaporation of solvents such as methanol or ethanol has already been utilized in soft pneumatic actuators. Miriyev *et al.* proposed encapsulating these solvents in gas pockets using a polydimethylsiloxane (PDMS)-based silicon elastomer^[109]. The inner volume of each pocket or chamber increases due to the pressurization caused by the expansion of gases in the system when the solvent reaches its boiling point. Although these systems produce effective actuation, no chemical interaction occurs with the solvent and a secondary reactant. These vaporization processes are a physical phenomenon driven by external stimuli (temperature increases) resulting in a liquid-to-gas phase change, and thus not a chemical GER [Figure 5].

Lee *et al.* have recently reported the development of vaporization-based actuators using thermoelectric materials to achieve fast actuation responses via quick heating/cooling cycles^[75]. Even though a chemical reaction is not involved in vaporization, it can be categorized as a gas evolution process of interest for soft robotics. Chemical reactions can offer a higher volume:reactant ratio compared with simple evaporation. This means that a smaller quantity of reactants can generate a larger volume of gas, resulting in increased actuation capabilities. Additionally, chemical reactions can be controlled and triggered with precision, allowing for on-demand actuation and modulation of force without needing external power sources. The advantages of one method over the other can vary depending on factors such as the desired actuation force, response time, environmental considerations, and available resources.

NEGATIVE PRESSURE

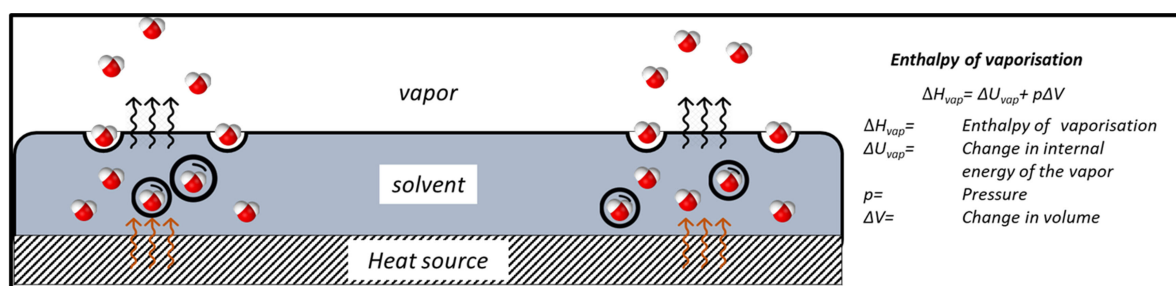
Within the pneumatic actuation field, only a few systems have exploited using negative pressures to achieve actuation compared to positive-pressure-driven actuators^[39,64,111-113]. Similarly to pneumatic actuators, many negative-pressure-driven soft actuators rely on mechanical vacuum pumps to operate successfully^[114].

Table 6. Combustion gas evolution^[103]

Combustion reactions
Combustion of methane
$\text{CH}_4 (\text{g}) + 2\text{O}_2 (\text{g}) \rightarrow \text{CO}_2 (\text{g}) + 2\text{H}_2\text{O}$
Combustion of ethane
$2\text{C}_2\text{H}_6 (\text{g}) + 7\text{O}_2 (\text{g}) \rightarrow 4\text{CO}_2 (\text{g}) + 6\text{H}_2\text{O}$
Combustion of butane
$2\text{C}_4\text{H}_{10} (\text{g}) + 13\text{O}_2 (\text{g}) \rightarrow 8\text{CO}_2 (\text{g}) + 10\text{H}_2\text{O}$
Combustion of propane
$\text{C}_3\text{H}_8 (\text{g}) + 5\text{O}_2 (\text{g}) \rightarrow 3\text{CO}_2 (\text{g}) + 4\text{H}_2\text{O}$

Table 7. Thermal decomposition reactions for gas evolution^[108]

Thermal decomposition reactions
Decomposition of sodium bicarbonate
$2\text{NaHCO}_3 (\text{s}) \rightarrow \text{Na}_2\text{CO}_3 (\text{s}) + \text{H}_2\text{O} (\text{g}) + \text{CO}_2 (\text{g})$
Decomposition of sodium azide
$2\text{NaN}_3 (\text{s}) \rightarrow 2\text{Na} (\text{s}) + 3\text{N}_2 (\text{g})$
Decomposition of mercury oxide
$2\text{HgO} (\text{s}) \rightarrow 2\text{Hg} (\text{l}) + \text{O}_2 (\text{g})$

**Figure 5.** Vaporization diagram. Showing the migration of molecules from an aqueous phase to a gas phase placed over a heat source and the enthalpy of vaporization formula^[110].

Although several interesting designs allow full actuation, the vacuum pump requires a large (and heavy) power source and, similar to positive pressure systems, a series of connectors, microcontrollers, and batteries^[115]. Since negative-pressure-driven actuation is limited by the strength of the materials used for the fabrication of the actuators, these systems offer a safer operating mechanism than those driven by positive, usually high, pressures via an inherent fail-safe mechanism^[116]. For the use of GCRs, the safety considerations need to be assessed according to the nature of the used reactants.

A notable example of this approach is the combustion of hydrocarbons, where localized elevated temperatures are achieved. As a result, the use of appropriate personal protection equipment (including high-temperature gloves) is advised. It is worth mentioning that when the temperature is controlled, combustion reactions result in a gas molecules deficit, hence its consideration as both GER and GCR. The mechanism for the combustion of methane (first reaction, Table 8) requires two molecules of O_2 as a precursor for every CO_2 molecule created, meaning it uses three equivalents of gas in the system (one CH_4 and two O_2) for every equivalent of gas produced [Table 8]. As described by Heisser *et al.*, a momentary vacuum or negative pressure is generated by the rapid cooling of the system when powering soft pneumatic

Table 8. GCRs previously used in soft robotics

Type	Reaction	Gas consumed	Ref.
Methane combustion	$\text{CH}_4 (\text{g}) + 2\text{O}_2 (\text{g}) \rightarrow \text{CO}_2 (\text{g}) + 2\text{H}_2\text{O} (\text{l})$	O_2	[88,89]
Oxidation	$4 \text{Fe} (\text{s}) + 3\text{O}_2 (\text{g}) + 6\text{H}_2\text{O} (\text{l}) \rightarrow 4\text{Fe}(\text{OH})_3 (\text{s})$	O_2	[31]
Metal hydroxide	$\text{KOH} (\text{s}) + \text{CO}_2 (\text{g}) \rightarrow \text{KHCO}_3 (\text{aq})$	CO_2	[31]

GCRs: Gas consumption reactions.

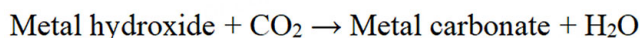
actuators using combustion reactions^[89]. The acid-base reaction of potassium hydroxide (KOH) with CO_2 , forming KHCO_3 and H_2O , was the first neutralization reaction successfully employed to produce negative pressures for driving pneumatic soft systems^[31] [Figure 6]. To the authors' knowledge, the use of GCRs for negative pressure-driven soft pneumatic actuators has only been reported in the aforementioned study. According to the reported results, the use of GCRs and their coupling with GERs for the oscillating actuation of soft pneumatic systems represent a significant opportunity for the soft robotics community to produce precise and compliant systems. These reactions are just a few within a broader category of similar chemical processes that will be further discussed in Section "GCRs".

GCRS

Hydroxides and CO_2 reactions

Acid-base (or neutralization) reactions are a fundamental class of chemical reactions that involve combining an acid and a base to form a salt and H_2O . According to the work of Brønsted and Lowry, acids are defined as proton donors and bases as proton acceptors. The reactions of CO_2 with various hydroxides are typical examples of acid-base neutralization reactions, which have been extensively studied^[117]. These reactions not only play a significant role in distinct industrial processes but also significantly influence the natural environment, such as in the carbonate-bicarbonate system in oceans and lakes. They are also known as double displacement reactions or metathesis reactions. In the case of the acid-base neutralization reactions involving CO_2 , the acid and base react to form a salt and H_2O , with the salt often in solid form. An example is the reaction between CO_2 and calcium hydroxide [$\text{Ca}(\text{OH})_2$], which forms CaCO_3 as a precipitate and H_2O ; this reaction plays a significant role in the formation and dissolution of CaCO_3 in natural systems such as coral reefs and shells^[118,119] [Table 9].

The production of carbonates, such as CaCO_3 , is an example of a precipitation reaction, i.e., to form an insoluble solid product. The overall balanced equation for a typical reaction producing carbonate is expressed as:



Oxidation reactions

Oxidation reactions use O_2 as a reactant and are classified into two categories depending on whether they occur at room temperature or require specific conditions. The reactions that occur at room temperature involve, for example, the rusting of Fe, the reaction of Cu and mercury (Hg) with O_2 , the reaction of tin (Sn) with O_2 , the reaction of lead (Pb) with O_2 , and the reaction of ethanol ($\text{C}_2\text{H}_5\text{OH}$) with O_2 . For soft robotics, oxidation reactions are of interest due to the loss of gas (O_2), which can be harnessed to generate negative pressures and, therefore, movement or change in shape. The speed of these reactions and the temperatures at which they occur are crucial as they directly influence the performance and functionality of soft robotic systems^[120]. On the other hand, the reactions that require elevated temperatures or specific conditions to occur include the reaction of aluminum (Al), iron sulfide (FeS_2), MnO_2 , carbon (C), sulfur dioxide (SO_2)

Table 9. Hydroxide-based GCRs

CO₂ absorption reactions
$2\text{NaOH (aq)} + \text{CO}_2 \text{ (g)} \rightarrow \text{Na}_2\text{CO}_3 \text{ (aq)} + \text{H}_2\text{O (l)}$
$\text{Ca(OH)}_2 \text{ (aq)} + \text{CO}_2 \text{ (g)} \rightarrow \text{CaCO}_3 \text{ (s)} + \text{H}_2\text{O (l)}$
$\text{Mg(OH)}_2 \text{ (aq)} + \text{CO}_2 \text{ (g)} \rightarrow \text{MgCO}_3 \text{ (s)} + \text{H}_2\text{O (l)}$
$\text{Sr(OH)}_2 \text{ (aq)} + \text{CO}_2 \text{ (g)} \rightarrow \text{SrCO}_3 \text{ (s)} + \text{H}_2\text{O (l)}$
$\text{Ba(OH)}_2 \text{ (aq)} + \text{CO}_2 \text{ (g)} \rightarrow \text{BaCO}_3 \text{ (s)} + \text{H}_2\text{O (l)}$
$2\text{Al(OH)}_3 \text{ (aq)} + 3 \text{CO}_2 \text{ (g)} \rightarrow \text{Al}_2(\text{CO}_3)_3 \text{ (s)} + 3 \text{H}_2\text{O (l)}$
$\text{Fe(OH)}_2 \text{ (aq)} + \text{CO}_2 \text{ (g)} \rightarrow \text{FeCO}_3 \text{ (s)} + \text{H}_2\text{O (l)}$
$\text{Cu(OH)}_2 \text{ (aq)} + \text{CO}_2 \text{ (g)} \rightarrow \text{CuCO}_3 \text{ (s)} + \text{H}_2\text{O (l)}$
$\text{Ni(OH)}_2 \text{ (aq)} + \text{CO}_2 \text{ (g)} \rightarrow \text{NiCO}_3 \text{ (s)} + \text{H}_2\text{O (l)}$
$\text{Zn(OH)}_2 \text{ (aq)} + \text{CO}_2 \text{ (g)} \rightarrow \text{ZnCO}_3 \text{ (s)} + \text{H}_2\text{O (l)}$
$\text{Pb(OH)}_2 \text{ (aq)} + \text{CO}_2 \text{ (g)} \rightarrow \text{PbCO}_3 \text{ (s)} + \text{H}_2\text{O (l)}$

GCRs: Gas consumption reactions.

with O₂, and the reaction of Fe with O₂, HNO₃ and H₂SO₄ (see Section “Oxidation reactions that do not occur at room temperature”).

Oxidation reactions that occur at room temperature

The reaction between solid Fe, gaseous O₂, and H₂O results in the formation of solid iron (III) oxide hydroxide [Fe(OH)₃] at room temperature^[121,122]. These reactions can occur at any temperature above 0 °C [Table 10].

Oxidation reactions that do not occur at room temperature

As mentioned at the beginning of Section “GCRs”, other oxidation reactions occur at high temperatures or specific pressure conditions to be energetically feasible; the listed reactions [Table 10] have been reported to occur at temperature ranges from 400 to 1,150 °C^[123-125]. However, these high-temperature reactions are not typically relevant to the field of bio-inspired soft robotics. This is primarily because the materials used in these fields, such as elastomers and hydrogels, are designed to operate under ambient conditions and can degrade or lose their functionality under high temperatures^[126,127]. Therefore, understanding and controlling reactions at such elevated temperatures is not a primary concern in these research areas.

Hydrogenation

Hydrogenation reactions are a type of chemical reaction in which hydrogen is added to double-bond containing (i.e., unsaturated) compounds, such as alkenes and alkynes, to convert them into saturated compounds. However, hydrogenation can be applied to other types of compounds as well under a range of conditions, including nitriles, aromatic compounds, CO₂, ketones, nitro compounds, carboxylic acids, and esters [Table 11].

These reactions have been widely used in the chemical and food industries to modify the properties of various compounds^[128]. Hydrogenation reactions can occur at RTP, but the reaction rate is typically low. To increase the reaction rate, catalysts are usually added, such as nickel (Ni), Pd or Pt^[129,130]. Moreover, the reaction conditions, such as temperature, pressure, and H₂ to unsaturated compound ratio, can also affect the reaction rate and selectivity.

COUPLING POSITIVE AND NEGATIVE PRESSURES

The use of chemical reactions to actuate pneumatic soft actuators is an area that has seen limited research,

Table 10. Oxidation-based gas consumption

Oxidation reactions
Room temperature
$4 \text{ Fe (s)} + 3 \text{ O}_2 \text{ (g)} + 6 \text{ H}_2\text{O (l)} \rightarrow 4 \text{ Fe(OH)}_3 \text{ (s)}$
$2 \text{ Cu (s)} + \text{ O}_2 \text{ (g)} \rightarrow 2 \text{ CuO (s)}$
$2 \text{ Hg (l)} + \text{ O}_2 \text{ (g)} \rightarrow 2 \text{ HgO (s)}$
$2 \text{ Sn (s)} + \text{ O}_2 \text{ (g)} \rightarrow 2 \text{ SnO}_2 \text{ (s)}$
$2 \text{ Pb (s)} + \text{ O}_2 \text{ (g)} \rightarrow 2 \text{ PbO (s)}$
$4 \text{ Al (s)} + 3 \text{ O}_2 \text{ (g)} \rightarrow 2 \text{ Al}_2\text{O}_3 \text{ (s)}$
High temperatures
$2 \text{ Fe (s)} + 3 \text{ O}_2 \text{ (g)} \rightarrow 2 \text{ FeO (s)}$
$2 \text{ FeS}_2 \text{ (s)} + 7 \text{ O}_2 \text{ (g)} \rightarrow 2 \text{ FeO (s)} + 4 \text{ SO}_2 \text{ (g)}$
$2 \text{ MnO}_2 \text{ (s)} + \text{ O}_2 \text{ (g)} \rightarrow 2 \text{ MnO}_3 \text{ (s)}$
$2 \text{ C (s)} + \text{ O}_2 \text{ (g)} \rightarrow 2 \text{ CO (g)}$
$2 \text{ SO}_2 \text{ (g)} + \text{ O}_2 \text{ (g)} \rightarrow 2 \text{ SO}_3 \text{ (g)}$

Table 11. Hydrogenation GCRs

Hydrogenation reactions
Unsaturated fatty acids
$\text{C}_{17}\text{H}_{33}\text{COOH (l)} + \text{H}_2 \text{ (g)} \rightarrow \text{C}_{17}\text{H}_{35}\text{COOH (l)}$
Ketones
$\text{R}_2\text{C=O} + \text{H}_2 \text{ (g)} \rightarrow \text{R}_2\text{CH-OH}$
Nitro compounds
$\text{RNO}_2 + 3 \text{ H}_2 \text{ (g)} \rightarrow \text{RNH}_2 + 2 \text{ H}_2\text{O (l)}$
Carboxylic acids
$\text{C}_{17}\text{H}_{35}\text{COOH (l)} + \text{H}_2 \text{ (g)} \rightarrow \text{C}_{17}\text{H}_{35}\text{CHO (l)} + \text{H}_2\text{O (l)}$
Esters
$\text{RCOOR}' + \text{H}_2 \text{ (g)} \rightarrow \text{RCH}_2\text{OH} + \text{R}'\text{OH}$
Haber-Bosch
$\text{N}_2 \text{ (g)} + 3 \text{ H}_2 \text{ (g)} \rightarrow 2 \text{ NH}_3 \text{ (g)}$

GCRs: Gas consumption reactions.

even more so for integrating both positive and negative pressure sources within soft systems. Fatahillah *et al.* demonstrated that soft grippers simultaneously combining positive and negative pressure-driven actuation can exert larger forces at lower bending angles than actuators driven by positive pressure only^[115]. These actuators consist of two independent channels, enabling them to combine contraction and extension forces through positive and negative pressures, respectively. Such soft grippers have shown the capability to securely hold objects weighing up to 4 kg with a negative pressure of only 200 mbar and a positive pressure of 600 mbar. This review has identified specific chemical reactions, as detailed in Section 3, which can meet the power and pressure requirements. Integrating these chemical reactions into an actuation system offers a promising avenue for fulfilling system requirements for soft grippers.

Interestingly, if GERs and GCRs are operated in separate reaction systems, they can be simultaneously incorporated into systems similar to that reported by Fatahillah *et al.* [Figure 7A]^[115]. Additionally, arranging these reactions serially within the same pneumatic system and pairing of a GCR and GER that reciprocally utilize the same gas as a product and reactant will facilitate oscillating motion via positive-negative pressure coupling^[31].

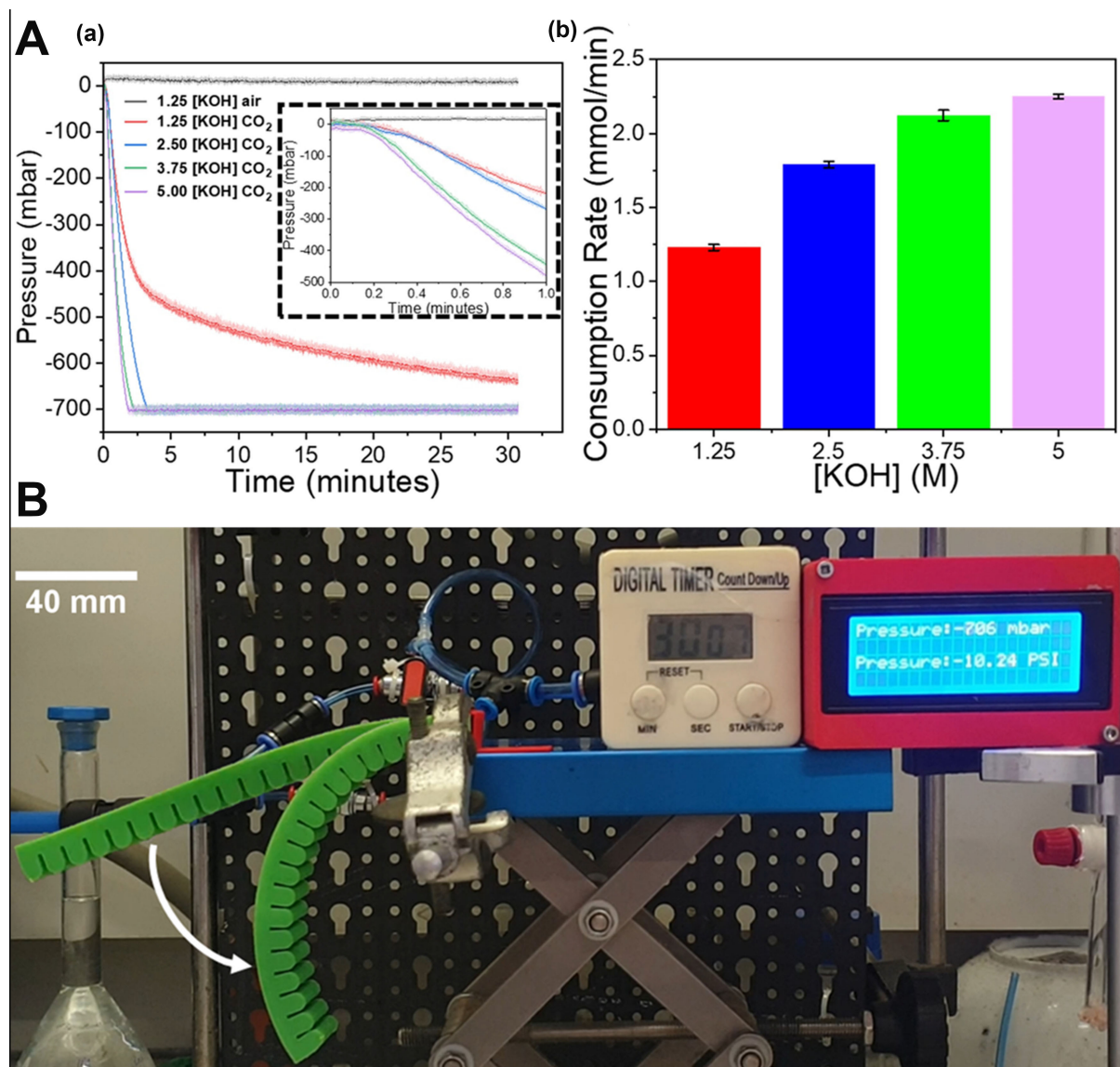


Figure 6. GCRs as a power source for negative pressure-driven actuation. (A) The reaction rate of the GCR of KOH with CO₂; (B) showing the effect of the GCR on the soft actuator when $P < 0$. Reproduced with permission^[31], Copyright 2023, Mary Ann Liebert Inc. GCRs: Gas consumption reactions.

Towards continuous actuation: coupling GER and GCRs

Chemical and safety considerations

As chemical reactions produce new products, the potential energy available in the resulting materials and systems can present safety challenges. This energy can be rapidly released as heat or pressure, determined by the kinetics and thermodynamics of the reaction^[131], underlining the need for detailed knowledge prior to application. The main experimental dangers arise from mischarging reactants, insufficient studies of the thermo-chemistry, inadequate pressure or temperature control, unsuitable agitation, under-rated or inadequate reaction chambers and containers, and human factors^[132]. Once the risks have been identified and assessed, the system involving the assessed chemical reaction must incorporate design measures to prevent those dangers.

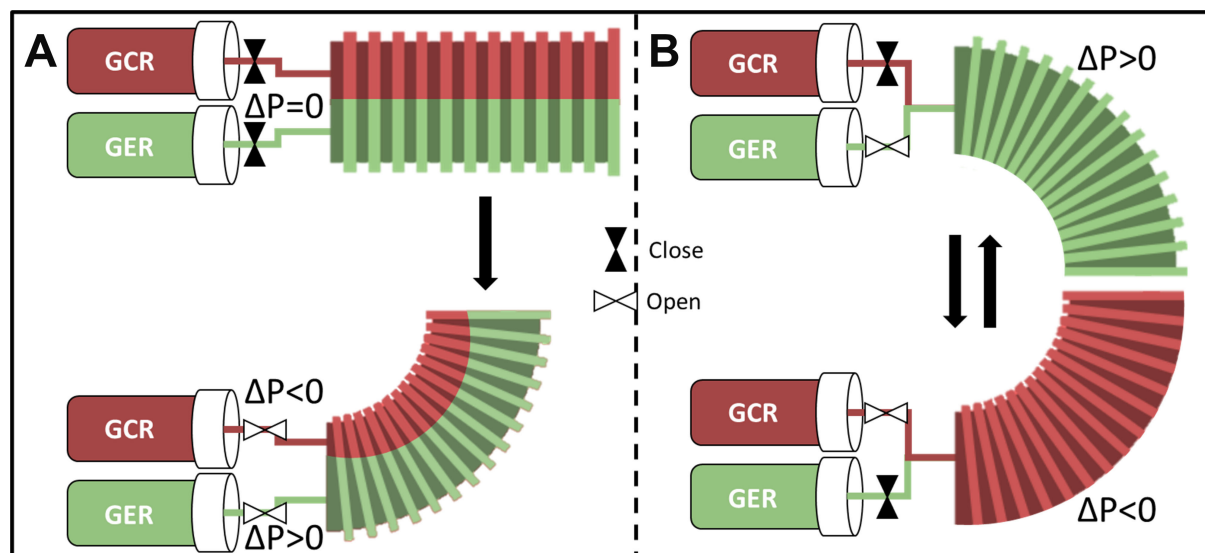


Figure 7. Coupling of GER and GCR as power sources. (A) Parallel configuration: Both GER and GCR are activated or opened simultaneously, resulting in actuation from the rest state with the GERs and GCRs valve closed state, as described in ref^[195]; (B) Serial configuration: the GER and GCR work independently and are alternated to achieve actuation in different directions. In this configuration, one power source is activated or open while the other remains inactive or closed, and the roles are reversed to change the direction of actuation. GER: Gas evolution reaction; GCR: gas consumption reaction.

There are a number of challenges when considering using chemical reactions for continuous actuation. The most significant factor is the precursor-product link between the GERs and GCRs. Further essential aspects to consider include response times (the time it takes for the actuator to react and perform the intended action) and compatibility (the materials used to fabricate the actuator must be compatible with the solvents or species used or generated in the reactions). Even if there is a precursor-product link between chemical reactions, by-products can interfere, compete, and dangerously interact with any involved species. For example, the reaction of chlorine and ammonia produces nitrogen chloride and hydrogen chloride. This reaction can produce various other by-products, including phosgene, a highly toxic gas. Phosgene can react with the desired products, nitrogen chloride and hydrogen chloride, to produce other toxic gases. This can lead to serious health problems or even death. Therefore, it is paramount to conduct a complete risk assessment of the intended reactions. Depending on the chemicals involved, they would exhibit different mechanisms of toxicity in humans. For instance, reactions where metal ions are produced can include side products such as highly reactive radicals, which are harmful and toxic to human cells^[133]. The chemical reactions highlighted in this review for energy generation in soft actuators were meticulously chosen for their safety and non-toxic nature, except where specifically indicated otherwise. Among these reactions are water splitting, acid-carbonate reactions, combustion, thermal decomposition, displacement, vaporization, and fermentation, all used for producing GERs. In contrast, GCRs employ methods such as hydroxide-based reactions, oxygenation, and hydrogenation. For most of these reactions, both the reactants and products are considered safe and non-toxic. For example, the water splitting reaction, while primarily a safety concern due to its explosive potential, produces non-toxic gases. However, appropriate safety measures must be strictly adhered to when utilizing this reaction.

One of the greatest concerns when designing a suitable container for a GER is thermal runaway. In the worst case, an exothermic reaction is uncontrollably accelerated by the increase in temperature produced by the ongoing reaction. The accelerated reaction and increase in temperature can create secondary decomposition reactions with higher energy and are significantly more hazardous than the original reaction.

Having a good understanding of the kinetics, the rate of the reactions, the potential for side reactions, and the stability of the products attained is essential for a successful and safe coupling of GERs and GCRs. Most importantly, the safety considerations to mitigate the potential hazards of GERs, including pressure build-up, fire and explosion, and toxic gas release, must be carefully discussed and assessed before designing and carrying out safe GERs and GCRs^[134-136].

Once risk assessments are completed, and any safety concerns and potential hazardous outcomes of a GER and GCR coupling have been addressed, additional variables such as temperature, volume, pressure and stability of the reactants and products should be considered. As these variables are connected by the ideal gas law, the size and construction of the materials in contact with the reagents and products are fundamental factors that require a careful design process. By carefully selecting the volume of the container and the amount of reactant used, it is possible to create conditions where the GERs and GCRs can proceed efficiently without being significantly influenced by temperature changes. A good example of this size consideration can be found in our previous work^[31] where a GER and GCR were successfully combined. The latter was set by carefully studying the selected chemical reaction and tailoring the reaction chamber to it, which allowed the use of low amounts of reactants to develop a safe and appropriate coupling of GERs and GCRs without noticeable temperature variation.

Design and control

Working with matter in its separate phases simultaneously is common in the chemistry field. The combination of and reaction between solids, liquids and gases are determined by surface interactions at two or more phases. Since applying chemical reactions in soft pneumatic systems relies on the interaction of gases with other reactants, a good understanding of such interactions is needed^[11]. The primary concern to create coupled GERs-GCRs is to enable gas interaction (formed from the GER) with GCR reactants. To this end, the focus is set on detailing actuator designs that could allow this type of interaction in various ways. Since most of the produced pneumatic soft systems focus on the actuator design, the design of the power sources is neglected (apart from miniaturization). When having two separate but compatible power sources, e.g., GER and GCR, their design and correct coupling is crucial to obtaining the maximum and most effective actuation response. As shown in [Figure 7](#), the configuration, isolation, and integration of both GER and GCR in a unified system offer initial variations and options to satisfy varied actuation mechanisms.

A parallel and isolated configuration [[Figure 7](#)] is desired when the actuation and functioning of a system rely upon the simultaneous application of positive and negative pressures. As detailed by Fatahllilah *et al.*, positive-negative pressure (PNP) actuators contain two separate pneumatic actuators; when actuated simultaneously, via pressure increase or decrease, they achieve a larger bending force at lower bending angles^[115]. Most of the soft pneumatic systems reported in literature work with individual pressure stimuli (positive or negative) comprising single or interconnected fluid chambers. For this type of individual pressure actuation, a serial and interconnected configuration of GER and GCR is desired. Both PNP and individual pressure actuation systems, with the appropriate configuration, would need a cascade arrangement of chemical reactions to allow an oscillating and cyclic performance akin to those found in electronically or electromechanically controlled systems.

Typically, actuators driven by chemical reactions exhibit varying response times and actuation forces, primarily influenced by the cycles of gas evolution and consumption reactions^[31,84,88,137]. Compared to conventional pneumatic systems, these chemical reaction-driven actuators may demonstrate slower response times and reduced force generation. For example, standard pneumatic systems, comprising a compressed air source, connection lines, flow-controlling valves, and regulated air receivers, can reduce rise

times to the millisecond range^[138] and offer tunable force output depending on the intended application^[139]. In contrast, the response time of chemically powered pneumatic actuators is typically within the seconds to minutes range^[31,70], and the force generated directly depends on the amount of gas produced and the internal volume of the soft actuators. It is important to note that while chemical reaction-driven actuators might not match the rapid response or high-force generation of conventional systems, they offer unique advantages in terms of design flexibility, miniaturization, and suitability for specific environments or applications.

In investigations where the coupling of GER and GCR has been evaluated for soft actuators^[31], the reaction chambers comprise over 95% of the total system volume (including the actuator). The volume of reactants, for both GER and GCR coupling, required only about 5% of the total volume of the system. As mentioned earlier, surface interactions are crucial for the effective completion of the GER and GCR involved, with interfacial areas of the systems and gas solubility playing a crucial role on the reaction kinetics for this type of reaction^[140-142]. Challenges, including small gas-liquid interface areas and poor gas miscibility, can be addressed by reducing the volume allocated for the power source and carefully designing reaction chambers with larger surface area for optimal chemical interactions. Furthermore, scaling these systems to larger robots introduces additional challenges, such as maintaining effective reaction kinetics at increased scales. Such challenges will necessitate innovative design and material selection approaches for larger-scale soft actuators.

To achieve independence from electrical systems, the soft and compliant properties of materials used in soft system fabrication have been leveraged to create integrated and non-electric fluidic circuits with logic-operating principles similar to those found in electronic circuits^[143,144]. Analogous to electronic circuits, a fluidic “switch” can be considered a basic form of control, allowing, or blocking the signal propagation. Microfluidic boards composed of check-valves and switch-valves, which are autoregulated by geometric design, can function as charging capacitors as pressure builds up and bends the soft deformable layer between microchannels^[145] [Figure 8A and B]. In this manner, system pressure dynamically autoregulates the soft controller, allowing for frequent oscillations under specific conditions. Constraints in channel and component sizes are the main limitations for optimal performance in these systems. Soft valves have been developed by exploiting the instability of elastomeric membranes, enabling them to switch, in a binary fashion, between pressures.

By selectively adjusting the printing or manufacturing density of materials, it becomes possible to create reaction chambers within the actuator, provided the reactants are safe for interaction^[146]. In addition to reaction chambers, a flexible power chamber, such as an expanding bladder made from non-extensible materials, can also be employed. However, it is important to ensure that the volume change under pressure does not influence the robot actions^[147]. Developing suitable containment systems is crucial for enhancing efficiency and reducing the number of reactants needed per reaction. Furthermore, miniaturization and portability of power sources are essential for creating optimal, efficient, and fully autonomous soft systems.

The pursuit of autonomous soft systems promotes developing systems capable of executing routines with minimal human intervention. Depending on the desired routine, power input in soft pneumatic systems is typically regulated by the passive operation of electrical switches, regulators, or valves that periodically supply the actuators.

Wehner *et al.* demonstrated control methods with GERs^[70]. Oscillating pneumatic actuation has been achieved by transporting H₂O₂ through microfluidic control boards into catalytic chambers, generating O₂

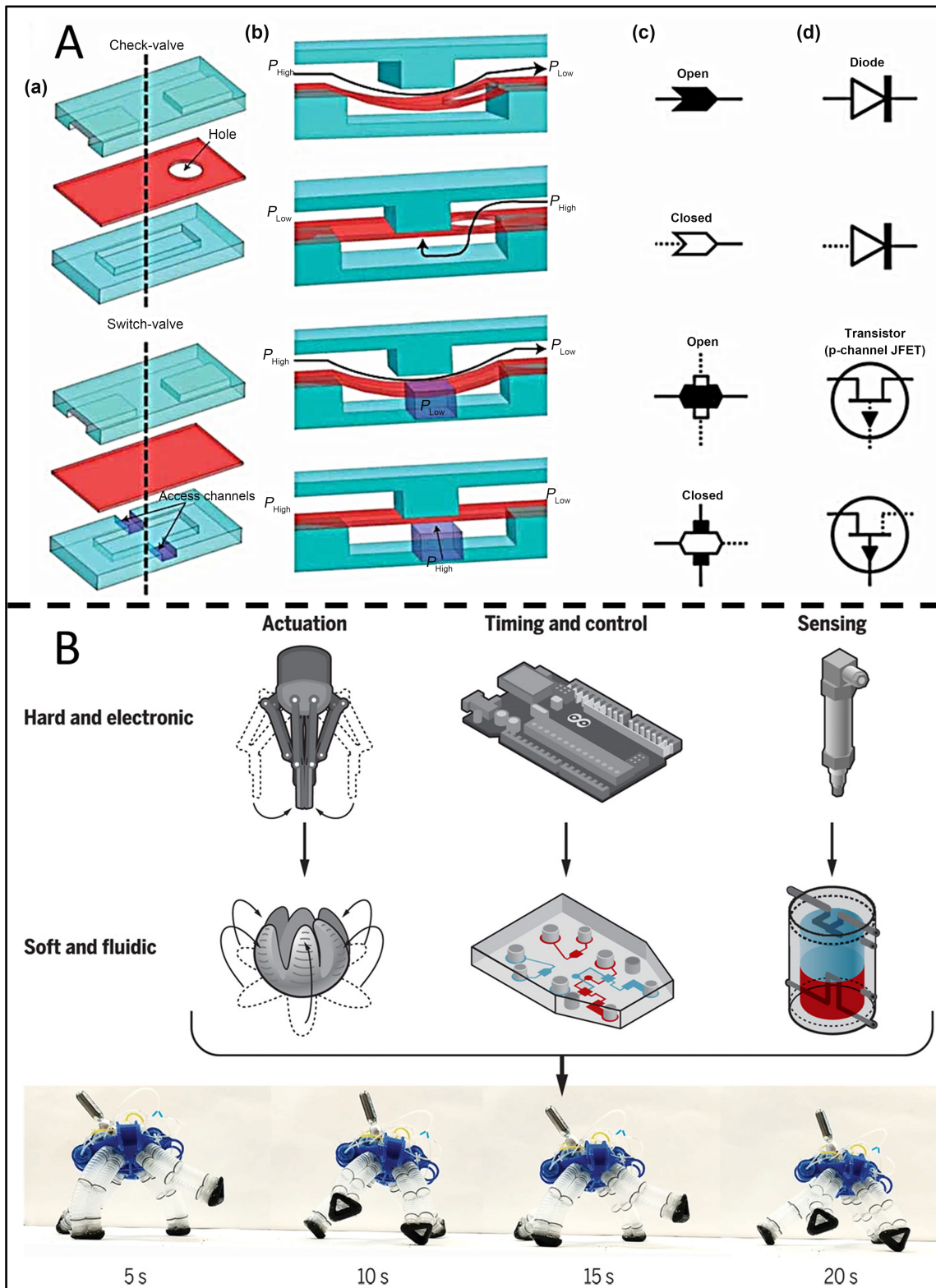


Figure 8. Fluidic-based soft control systems (A) A check-valve and switch-valve in both the open and closed state based on differential pressure and their corresponding component state symbol along with the analog diode and p-channel JFET transistor components. Reproduced with permission^[145]. Copyright 2010, Springer Nature; (B) Showing the different components for actuation, control and sensing to create autonomous crawling systems combining bistable valves, soft logic gates and soft pneumatic actuators^[143,144]. Reproduced with permission^[143]. Copyright 2021, AAAS. JFET: Junction field-effect transistor; AAAS: American Association for the Advancement of Science.

and pressurizing the system. Although not yet proven with GER-GCR coupling systems, the working principle based on fluid pressure seems compatible and suitable for achieving oscillating and cycling actuation via the coupling of GER and GCR [Figure 9A]. The working principle of bistable flexible membranes in rubber “poppers” allows these valves to harvest power only while transitioning between open and closed configurations [Figure 9B and C]. Carefully selecting and tuning the mechanical properties of membranes ensure that the switch remains in its selected position without any applied force and will not switch its configuration until the necessary force to “snap” the membrane back is achieved.

CONCLUSIONS AND OUTLOOK

Efficiency and control

One of the most attractive features of chemo-driven pneumatic actuation is that it can be used to create autonomous actuators, with the long-term goal of finding self-sustaining (oscillating) coupled chemical reactions to negate the requirements for an external power source^[70]. This makes chemo-driven pneumatic actuation well-suited for applications where power is limited or unavailable. However, achieving fully oscillating pneumatic systems presents a significant challenge due to thermodynamic constraints^[150]. For instance, certain fully oscillating chemical reactions, such as the Belousov-Zhabotinsky (BZ) reaction, are known but are very slow and complex^[151]. Additionally, advantages and disadvantages of chemical-based pneumatic actuation are generally related to the type of GER and GCR used, such as the actuation speed and the achievable amplitude motion. It is noteworthy that other factors related to the materials selection for fabricating the embedded reaction chambers and pneumatic actuators must be considered to avoid issues related to material fatigue or degradation.

As this review focuses on using various chemical reactions for actuation in soft robotic systems, it is important to highlight that the “ideal” chemical reaction highly depends on the specific application. In medical applications, biocompatibility and precision are paramount, necessitating safe and controllable reactions^[152]. For underwater robotics, reactions efficient in low-oxygen environments are preferred^[153]. In industrial settings, factors such as speed, scalability, and cost take precedence^[154]. This diversity in requirements underscores the need for tailored approaches in selecting chemical reactions for soft robotics, a theme that has recurred throughout our discussions on GERs and GCRs.

Looking ahead, there is an unexplored opportunity in exploiting bio-based reactions such as enzyme-based reactions for chemo-driven pneumatic actuation^[155]. The use of bio-catalysts could potentially offer more sustainable and environmentally friendly alternatives while maintaining high efficiency at low temperatures and pressures.

Furthermore, chemo-driven pneumatic actuation can also benefit from external agents or specific conditions to boost its efficiency and control. In this context, materials science has made significant strides by developing diverse materials that can include embedded or encapsulated catalysts for powering reactions that drive actuation^[156-158]. These materials not only serve as catalysts to augment GERs and GCRs, thereby reducing the energy requirements for reactions to proceed, but also offer gas selectivity and affinity for superior control over production and consumption ratios.

However, as mentioned, one of the primary challenges when coupling GERs and GCRs lies in exerting control over gas capture and storage^[10]. To address this issue, a range of smart materials showing promising potential for integration into chemo-driven pneumatic soft actuators has been introduced^[159]. One promising strategy for controlling gas capture and storage involves employing porous materials^[160-163]. Characterized by an intricate network of interconnected pores, these materials can be tailored to selectively trap and release gases. This intrinsic and hierarchical porosity positions them as highly effective candidates for gas storage and separation.

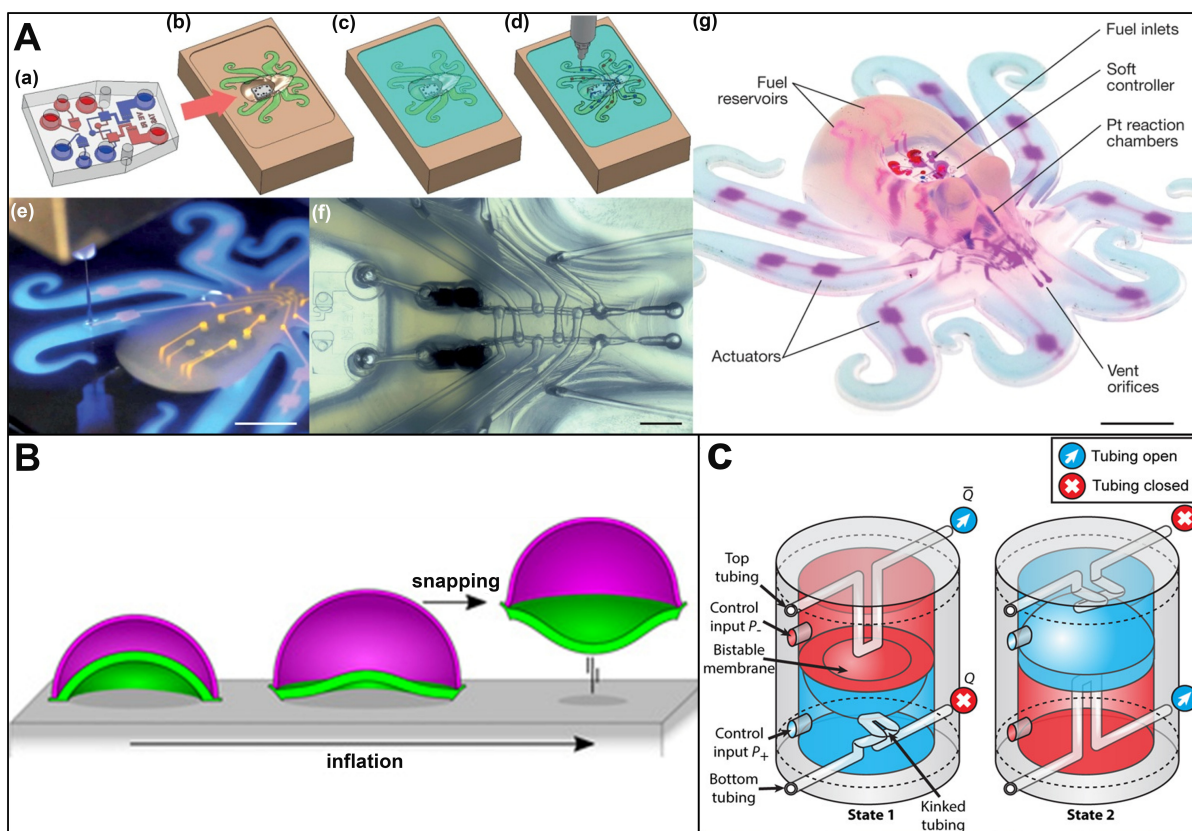


Figure 9. Fluidic-based control technologies in soft systems. (A) The “Octobot” featuring a series of microfluidic channels to manage the pneumatic actuation of the octopus-like tentacles. Reproduced with permission^[70]. Copyright 2016, Springer Nature; (B) A jumping actuator inspired by rubber popper toys, combining gas pressurization and the snapping mechanism of a bistable membrane for actuation. Reproduced with permission^[148]. Copyright 2020, Science Robotics; (C) Bistable valve utilizing the snapping mechanism of a bistable membrane to regulate fluid flow. Reproduced with permission^[149]. Copyright 2018, Science Robotics.

While porous materials and liquids are indeed smart materials solutions based on innovative chemistry, their ability to convert into gas or remove gas brings them into the realm of chemical approaches. For instance, hierarchical pressure-driven systems could potentially be developed if a material absorbs and desorbs gases within a pressurized system^[164,165]. Giri *et al.* have successfully explored the concept of porous liquids^[166], where gas molecules are trapped within the molecular cages of a liquid, introducing volumetric vacancies to the liquid. This approach could be extended to designing hierarchically porous systems that utilize both porous materials and porous liquids to enhance gas capture, storage and on-demand release^[167].

A simple approach to obtain the efficiency of the presented chemical reactions is by yielding the products obtained. This strategy will depend not only on the reactants but also on the physical characteristics or architectures of the reaction chambers, the distribution of the reactants, and the mass transfer of the products to further favor the reaction process (see Section “THEORETICAL CONSIDERATIONS”). In a more general fashion, the efficiency could be expressed as the ratio of the output energy to the supplied (or input) energy. This concept would lead to calculations of the energy demands (energy yields) of the reactions conducted under the different conditions and architectures of the reactor or reaction chamber^[134,168].

Developing more efficient and controllable chemo-driven pneumatic soft actuation systems and materials could revolutionize soft robotics and have a wide range of practical applications in fields such as biomedical engineering and advanced manufacturing.

Design and manufacturing

By exploring and integrating various geometries and material densities, the rapidly growing field of soft pneumatic actuation has unlocked a broad spectrum of achievable motions (i.e., linear, bending, and twisting)^[169]. Nevertheless, it is paramount to start considering integrating power sources within the design of chemo-driven pneumatic actuators. Bio-inspiration is already widely accepted and exploited for soft robotics and has been a primary source of inspiration for motion and material design^[35,170]. For example, chambers for storing chemicals can be found in many biological systems^[171,172]. It is envisaged that a significant step toward achieving the autonomy of chemo-driven pneumatic systems can be made by assigning a dedicated compartment for storing reactants and gases.

Safety

As mentioned in Section “Towards continuous actuation: coupling GER and GCRs”, the safety of new systems that generate pressure or vacuums needs to be carefully understood to inform the design of future systems. Many of the systems suggested can be controlled via feed ratios, and the products and reactants are selected to be biocompatible in many cases. Although future work will pave the way for introducing physical barriers (such as flow gates) to prevent uncontrolled chemical reactions, consideration has to be given to the chemical degradation of the materials used over time, embedded reaction chambers and their degradation and the ability of the system to vent in a safe fashion when potentially overpressurized.

Sustainability

The concerns regarding sustainability come under two areas. Firstly, it is crucial for future systems to focus on regenerating starting materials for both GERs and GCRs. Achieving sustainability may involve creating closed loops by coupling these reactions, although the number of effective cycles will be inherently limited. An innovative approach is to generate starting materials from biological sources, such as harnessing gas production from gut bacteria or leveraging natural pH variations within biological systems, to maintain the viability of these meticulously designed systems. Secondly, when considering the response of the actuator to these reactions, it is essential to address both the degradation of the systems and the potential loss of functionality. To ensure compatibility with biological systems, comprehensive testing over repeated cycles is necessary, particularly focusing on resolving issues related to the reservoirs and power sources of actuators. To address the concern of gas loss through material fatigue (and degradation) with prolonged use, recent advances in self-healing materials offer a promising solution. For instance, a study by Terryn *et al.* demonstrates the development of self-healing elastomers, thereby mitigating the risk of gas leakage and prolonging the functional lifespan of actuators^[173]. Incorporating such self-healing systems within the makeup of actuators could significantly enhance the sustainability and durability of soft pneumatic actuators, especially in long-term application settings.

In conclusion, we believe that significant opportunities exist, in carefully chosen application areas, to expand the use of chemo-pneumatic soft actuation. Although this field is in its early stages of development, noteworthy pathways already exist related to a) the application of bio-inspired design and b) the rules for safe and long-term sustainable solutions to soft actuation. The hope is that this overview of the field, the detailed discussion of all aspects of efficiency and control, design, safety, and sustainability, as shown in the roadmap presented in [Figure 10](#), will lead to innovation in the research, development and application of chemo-driven pneumatic actuators.

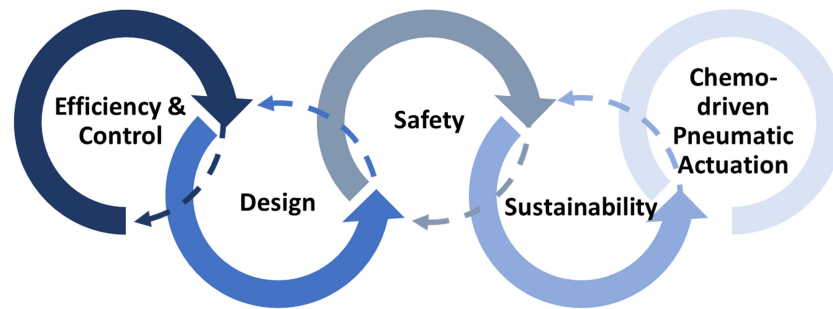


Figure 10. Roadmap towards chemo-driven pneumatic actuation. Crucial areas of consideration for the development of reliable, sustainable, and compliant chemo-driven pneumatic soft actuators. Dashed arrows represent the feedback loops present on each of the crucial areas to allow for the optimization of chemo-driven actuators.

DECLARATIONS

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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