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A review of humidity-responsive actuation in liquid crystal elastomer films: Controlling reversibility and irreversibility through hydrogen bonding

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Abstract. Humidity-responsive actuation refers to the phenomenon where certain materials alter their shape, size, or color in response to fluctuations in humidity levels. This behavior is reminiscent of natural processes observed in plants, where seeds and flowers react to moisture. The potential applications of humidity-responsive materials span various fields, including soft robotics, smart sensors, biomimetic devices, and anti-counterfeiting measures. Liquid Crystal Elastomers (LCEs)are particularly noteworthy in this context, as they possess a unique combination of properties derived from both liquid crystals and elastomers. These materials exhibit significant changes in shape in response to external stimuli such as temperature, light, electric fields, and humidity. As a result, this paper focuses on the humidity-responsive properties of Liquid Crystal Elastomer (LCE) films, with a specific emphasis on the role of hydrogen bonding in this process. And this review covers the synthesis methods, underlying mechanisms, performance characteristics, challenges, and future prospects of humidity-responsive Liquid Crystal Elastomer (LCE) films.

Keywords: Humidity-responsive Actuation, Liquid Crystal Elastomers, Humidity

1. Introduction

Humidity, as an omnipresent environmental factor, significantly influences how a lot of materials behave. Some materials exhibit a remarkable sensitivity to humidity, undergoing changes in shape, size, or even color in response to its fluctuations. This phenomenon, known as humidity-responsive actuation, is not limited to laboratory settings but is also observed in nature.

For instance, the ice plant (Mesembryanthemum crystallinum) has seed pods that open and close with humidity changes, aiding in seeds dispersal, while the African Daisy (Osteospermum ecklonis) has petals that move in response to the moisture, protecting its reproductive organs from rain and dew [1] [2-4]. The appeal of humidity-responsive materials and devices to scientists and engineers lies in their versatility, safety, and wireless operation. These materials can enable the development of gentle robots, color-changing sensors for humidity detection, drug-delivery systems triggered by moisture, and even anti-counterfeiting labels that only become visible in humid conditions, akin to QR codes or patterns that only show up when it is humid [5].

Various types of materials exhibit humidity-responsive behavior, including inorganic materials, polymers, and biomaterials. However, one standout class is LCEs, which blend liquid crystals with rubbery materials. LCEs are capable of changing shape in response to different stimuli such as temperature, light, and humidity. This shape-shifting ability is attributed to the dynamic behavior of liquid crystal segments within the polymer chains, enabling controlled and programmable responses. In addition, LCEs can exhibit diverse colors and patterns based on their interaction with light [6].

Therefore, this review zeroes in on humidity-responsive LCE films, particularly emphasizing their response to humidity through hydrogen bonding. And this bonding, which occurs between hydrogen and other atoms like oxygen or nitrogen, especially in the presence of water, is crucial for the reversible changes in shape observed in these films. The review will discuss the latest advancements in the fabrication of these films, their underlying mechanisms, and performance characteristics, as well as the challenges and future prospects associated with humidity-responsive LCE films. [7].

2. Fabrication of Humidity-Responsive Liquid Crystal Elastomer Films Through Hydrogen Bonding

Making humidity-responsive LCE films with hydrogen bonding sounds complex, but it can be broken down into two major steps: cooking up the LCE precursors and then curing the LCE films. The precursors are like a special recipe that contains four ingredients: liquid crystal monomers (the base and structure of LCEs), cross-linkers (they tie everything together), initiators (the start button for polymerization), and hydrogen bond donors or acceptors (they bring in the hydrogen bonding magic).

Curing the LCE films is a bit of a balancing act. The curing process requires the right temperature and humidity, and everything must be just right. This alignment can be done in a few ways, such as stretching, rubbing, using magnetic or electric fields, or photoalignment. The curing has got two phases: pre-curing, where a partly cross-linked network is obtained, and post-curing, where all the material is locked into place.

Regarding the types of humidity-responsive LCE films, there are two main varieties: the reversible and the irreversible. The reversible type uses a hydrogen bond donor or acceptor as cross-linkers. These can make and break links with water or other hydrogen bond partners, allowing the LCEs to change shape back and forth in response to humidity changes. The irreversible type, on the other hand, uses these hydrogen bond players as additives. They interact with the LCEs in a way that once the shape changes due to humidity, either making the material more flexible or locking it in place.

3. Mechanism of Humidity-Responsive Actuation of Liquid Crystal Elastomer Films Through Hydrogen Bonding

Humidity-responsive LCE films operate through a sophisticated interplay of molecular interaction, best understood through the Flory - Rehner theory. And this theory provides insights into how cross-linked polymers react when they undergo swelling. It delineates that the whole game of swelling changes in LCEs is down to three kinds of energy: mixing energy (how LCEs and water get along), elastic energy (how stretchy the LCEs are), and surface energy (the tension at the interface between LCEs and water).

In the context of humidity-responsive LCE films, the critical balance lies in optimizing the interplay between the mixing and elastic energies. The mixing energy decreases as hydrogen bonds form between the LCEs and water, making them more compatible. Conversely, the elastic energy increases as the LCEs swell or shrink, causing them to stretch or compress. These films aim to minimize the total energy change when they take in or lose water.

For the reversible type of LCE films, the presence of reversible cross-links acts as a built-in on-off switch. These cross-links can either snap open or lock shut. When it gets humid, these links break open, making the LCEs more relaxed and causing them to swell and bend. But when the air dries out, the links close back up, making the LCEs stiffen and go back to their original shape.

In contrast, the irreversible type exhibits a one-way change in behavior. This is achieved by making the LCEs either more flexible or more rigid through plasticization or antiplasticization. This changes the behavior of the liquid crystal mesogens inside the LCEs. When humidity goes up, the interaction with hydrogen bond donors or acceptors makes the LCEs more pliable, allowing the mesogens to move to a

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more relaxed state. But when the humidity drops, this interaction lessens, making the LCEs more rigid and locking the mesogens in place.

4. Performance of Humidity-Responsive Actuation of Liquid Crystal Elastomer Films through Hydrogen Bonding

Judging how well these humidity-responsive LCE films work with hydrogen bonding is a bit like rating a high-tech performance. There are a few key things to watch out for, like the actuation range (how much the LCE films can change shape), the actuation speed (how quickly they react to humidity changes), the stability (how well they hold their shape under steady humidity), and the durability (how many times they can change shape and still work well).

The performance of these LCE films is not just random; it is influenced by a bunch of factors. Think of it like a recipe – the ingredients (composition of LCEs), the cooking method (structure and morphology), and the kitchen conditions (humidity and environment) all play a part. The composition affects the hydrogen bonding, liquid crystal order, and elasticity. The structure and morphology control how the liquid crystal mesogens line up, how the LCEs swell or shrink, and how water gets in and out. The humidity condition is the driving force behind the whole actuation show, and the external environment can either help or hinder the performance.

Improving these films is a bit like tweaking a high-performance engine. One can play around with the composition to get better bonding, order, and elasticity. They can design the structure and morphology for better alignment, swelling, and water interaction. Adjusting the humidity conditions can give one more control over the actuation. And do not forget about adding other elements to the mix, which can either boost or balance the humidity response.

5. Challenges and Opportunities for the Future Development of Humidity-Responsive LCE Films Through Hydrogen Bonding

The field of humidity-responsive LCE films using hydrogen bonding is like an uncharted territory that is getting more and more explorers each year. But, it is not all smooth sailing; there are some big challenges and exciting opportunities ahead.

First up, the whole mechanism behind how these LCE films respond to humidity is kind of a mystery. We are talking about the nitty-gritty details of how LCEs, water, and hydrogen bond players interact at the molecular level. To crack this puzzle, we need some next-level experiments and theories that can zoom in on these interactions across different scales.

Then there is the performance of these films. Right now, it’s like they are in the early stages of training – not quite Olympic material yet, especially in terms of how much they can change, how fast they react, how stable they are, and how long they last. To up their game, we need to come up with new materials, play around with their structures and shapes, fine-tune the humidity conditions, and maybe throw in some other stimuli to see what happens.

And let us not forget about actually using these films in real life. Right now, their application is very limited. To really make the most of these films, we need to think outside the box. This could mean designing smart devices that adapt on the fly, combining sensing and moving functions, or even trying to mimic natural systems.

6. Conclusion

In this review, a thorough exploration has been conducted into the current landscape pertaining the development, understanding, and advancement of humidity-responsive LCE films through the strategic utilization of hydrogen bonding. The analysis has not only shed light on the existing state of affairs but has also cast a spotlight on the myriad challenges and opportunities that lie ahead in this dynamic field.

Indeed, the exploration has revealed that humidity-responsive LCE films represent a promising avenue for scientific and technological advancement. These materials, with their unique blend of liquid crystal and elastomer properties, exhibit remarkable responsiveness to environmental changes,

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especially in humidity levels. Besides, the paper aims to provide a thorough and thought-provoking look at this topic, hoping that it sparks even more curiosity and innovation in this fascinating area of study.

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