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Engineering Polyimide Materials for Moon Exploration Missions: Intrinsic Self-Healing and Shape Memory Properties via Supramolecular Interactions

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Abstract. Polyimides are a well-established class of high-performance polymers known for their excellent thermal and chemical stability, mechanical robustness, low outgassing, and radiation resistance. These properties have made polyimides essential in aerospace applications, both for systems operating in low Earth orbit and for those used in long-term exploration missions. In recent years, polyimides have attracted growing interest for planetary exploration, particularly for lunar missions. The critical challenges of these missions include mechanical abrasion from regolith dust and the extreme temperature fluctuations. These conditions can damage exposed surfaces and compromise system performance. To overcome these challenges, self-healing materials represent a promising strategy, enabling polymers to autonomously repair mechanical damage and thereby enhance the reliability and operational lifetime of spacecraft components.

Introduction

Among the different classes of high-performance polymers, polyimides (PIs) are recognized as one of the most reliable options for aerospace applications. Their combination of thermal and chemical stability, mechanical strength, low outgassing, and resistance to radiation has made PIs widely used in many aerospace systems, from satellites in low Earth orbit (LEO) to components for future lunar missions [1, 2]. For LEO applications, PIs are commonly used as insulating layers in multi-layer insulation systems and as protective coatings against atomic oxygen erosion, which can severely degrade polymeric surfaces [3]. This class of polymers also finds application in flexible solar arrays, solar sails, and high-temperature adhesives [3]. In recent years, PIs have attracted growing interest for their applications in planetary missions, especially for lunar exploration missions. The critical challenges of these missions, such as mechanical abrasion from regolith dust, demand materials that can withstand harsh environments while maintaining structural integrity. Indeed, the surfaces of exposed materials may suffer progressive damage, for instance due to the accumulation and adhesion of abrasive lunar dust [4, 5]. This phenomenon can degrade the surface, and compromise the functionality of systems, posing a risk to mission success.

Over the last decade, self-healing materials have gained increasing interest in the aerospace field, thanks to technological advances that allow the development of systems able of repairing mechanical damage [6]. Integrating these materials into the design of spacecraft components can enhance structural integrity, safety, and mission reliability, extending the operational lifetime of space systems [7, 8]. Based on this ability, self-healing mechanisms are classified as intrinsic or extrinsic [9]. Extrinsic self-healing refers to systems in which the healing agents are embedded

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within the material structure, typically stored in microcapsules networks that release the agent upon damage [10]. Intrinsic self-healing mechanisms do not require healing agents. They are typically based either on dynamic covalent chemistry, such as Diels–Alder reactions, dynamic urea and imine bonds, or on supramolecular interactions, including H-bonding, van der Waals forces, hydrophobic interactions, π–π stacking, and interdiffusion mechanisms [11]. According to the type of recovery process, self-healing materials are autonomous and non-autonomous [9]. Non-autonomic self-healing materials require an external trigger, like heat. Autonomic self-healing materials do not require any additional external trigger, but the damage itself is the stimulus for the healing [9].

In this context, the development of PI with self-repair capabilities and shape memory behaviour is a promising strategy to enhance the resilience and longevity of components during long-term exploration missions. This work focuses on the design, synthesis and characterization of flexible supramolecular PI materials with non-autonomous intrinsic self-healing and shape-memory properties. These results highlight the strong potential of polyimide systems engineered via supramolecular interactions to act as multifunctional materials for future space missions.

Experimental section

Polyimide synthesis. Three types of PIs were synthesized under an argon atmosphere via the polycondensation reaction between the same aromatic fluorinated dianhydride and two different diamines (one aliphatic and one aromatic). The corresponding diamine(s) were dissolved in a mixed DMAc/IPA solvent under mechanical stirring, and a stoichiometric amount of dianhydride was subsequently added to the solution. The reactions were carried out for 24 h at room temperature. After reaction, the solutions were heated in oven to evaporate the solvent. Finally, a thermal imidization process was optimized to obtain homogeneous PIs membranes (thickness ~ 300 μm).

Characterization methods. Density was measured by the hydrostatic weighing method in ultrapure water using the Sartorius YDK01 kit [2]. The wettability was assessed using the DataPhysics OCA15 PRO tensiometer, and the surface free energies (SFE) was calculated following the Owens–Wendt method [2]. In order to assess the self-healing behaviour of materials, optical images were acquired using a Leica polarized light microscope (DMLP series, Leica Microsystems, Germany). The sample surfaces were mechanically scratched with a scalpel, simulating external mechanical damage that may occur during exploration missions. Subsequently, the samples were heated at different temperatures (T = 50 °C and 150 °C). The two temperatures were chosen to provide a representation of the thermal conditions encountered on the lunar surface. Images were acquired immediately after the mechanical damage and subsequently after the repair induced by heat exposure.

Results and discussion

The fully aromatic PI is referred to as PI-1 (Fig.1a), the supramolecular semi-aromatic PI containing an aliphatic diamine and the same aromatic dianhydride as PI-2 (Fig.1b), and the supramolecular copolyimide prepared from both an aliphatic and an aromatic diamine with the same aromatic dianhydride as coPI-3 (Fig.1c).

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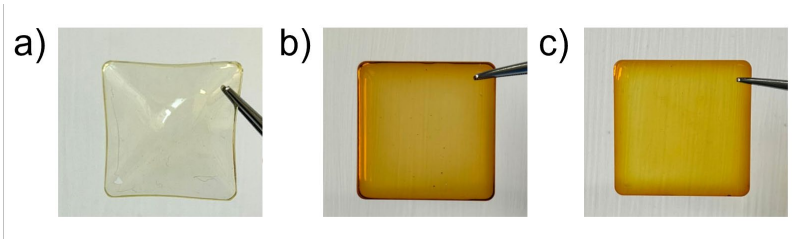


Fig.1 *a) Fully aromatic polyimide PI-1, b) semi-aromatic polyimide PI-2, and c) semi-aromatic copolyimide coPI-3 membranes obtained after thermal treatment.*

The density values of PI-1, PI-2, coPI-3 are reported in Table 1. The analysis indicates that the density decreases from 1.452 g·cm^{-3} (PI-1) to 1.107 g·cm^{-3} (PI-2) and 1.154 g·cm^{-3} (coPI-3) with the introduction of the aliphatic diamine. This reduction can be attributed to the lower packing efficiency and higher free volume associated with the flexible aliphatic segments. The reduction in density contributes to lowering payload mass, a key factor in spacecraft design and mission cost efficiency.

The WCA and the SFE with its dispersive (γ^d) and polar (γ^p) components are reported in Table 1. The wettability analysis unveiled the high level of hydrophobicity for all samples, with WCA ≥ 100°. The SFE showed a noticeable decrease, from 30.08 mJ m^{-2} for PI-1 to 24.69 mJ m^{-2} for PI-2, due to the introduction of the aliphatic diamine. The increase of hydrophobicity and the decrease of surface energy play a crucial role in dust mitigation. The reduction in SFE as it decreases the van der Waals and electrostatic forces responsible for lunar dust adhesion. Consequently, materials with higher hydrophobicity and lower SFE are less inclined to particle accumulation. In addition, hydrophobicity represents a critical requirement for space materials, as it effectively mitigates moisture uptake. Indeed, during ground handling and launch, materials may absorb water from the terrestrial environment, which in orbit can sublimate under specific conditions and generate structural defects.

Table 1. *Density, WCA, SFE with dispersive (γ^d) and polar (γ^p) components, for the aromatic PI-1 and the semi-aromatic PI-2 and cPI-3 samples.*

Sample	Density [g cm^{-3}]	WCA [°]	SFE [mJ m^{-2}]	γ^d [mJ m^{-2}]	γ^p [mJ m^{-2}]
PI-1	1.452 ± 0.024	100.0 ± 0.8	30.08	29.02	1.06
PI-2	1.107 ± 0.021	103.7 ± 2.3	24.69	23.75	0.94
coPI-3	1.154 ± 0.063	101.1 ± 1.6	25.07	24.03	1.04

Fig.2 shows the optical images of the materials surface immediately after the damage and subsequently after the repair induced by heat exposure. For brevity, only two representative samples (PI-1 and PI-2) at one self-healing temperature (T = 150 °C) are shown. The fully fluorinated aromatic PI-1 exhibited no self-healing (Fig.2a). Indeed, even after 40 min, the cut is still clearly visible. The same behavior was also observed at T = 50 °C. Fig. 2b shows the intrinsic self-healing behavior of PI-2 upon heating at T = 150 °C, where the damage disappears immediately.

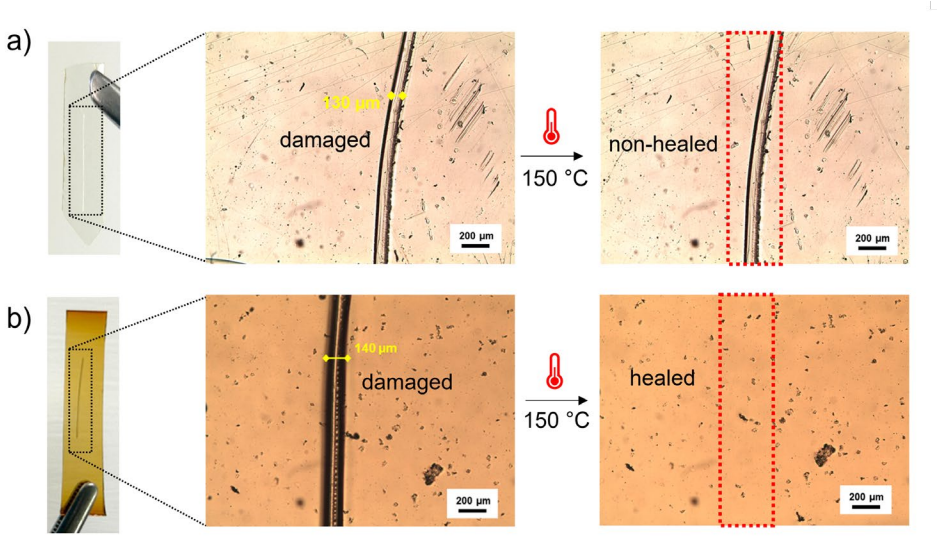


Fig.2 *Optical microscopy images of cut surfaces (left) and their response after heating at 150 °C (right). a) Aromatic PI-1, which does not self-heal even after 40 min, and b) semi-aromatic PI-2, showing rapid intrinsic self-healing within few seconds.*

Overall, all semi-aromatic PIs developed in this work exhibit intrinsic self-healing ability, and the repair process becomes progressively faster with increasing temperature, reaching healing times of only a few instants under elevated conditions (T = 150 °C). An increase in temperature enhances the segmental mobility of the polymer chains, allowing them to diffuse more across the damaged interface. Also, dynamic supramolecular bonds become more reactive, as higher thermal energy facilitates their reversible recombination. At T = 50 °C, PI-2 required approximately 20 min to restore the damaged area, whereas coPI-3 exhibited much faster recovery, completing the healing process in less than 20 s. When the temperature was increased to 150 °C, both materials healed significantly faster, with PI-2 achieving complete recovery in under 1 min and coPI-3 in less than 10 s. These results clearly indicate that higher temperatures enhance chain mobility and dynamic bond exchange, leading to accelerated self-repair. The self-healing rate is a critical factor for space applications, where rapid recovery from mechanical damage is essential to maintain structural integrity and functionality in harsh environments. Indeed, materials exhibiting fast self-healing can significantly enhance the reliability and lifetime of spacecraft components.

The supramolecular semi-aromatic PIs (PI-2 and coPI-3) developed in this work exhibit also shape memory behavior, being capable of recovering their original shape after deformation. Fig. 3 shows a schematic representation of the shape memory behaviour of PI-2. Specifically, the sample is deformed (Fig.3a) and subsequently recovers its original shape (Fig.3b-c), with the recovery time decreasing as the temperature increases.

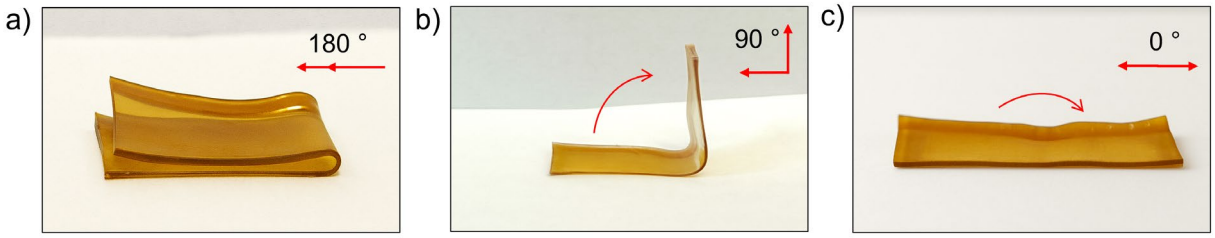


Fig.3 *Representative illustration of the shape memory behaviour of PI-2. a) The sample is deformed to 180°; b) bent to 90°, with a recovery time dependent on temperature; c) recovery of the original flat shape (0°). The images highlight the high elasticity and excellent shape recovery ability of the material after deformation.*

The test was carried out at 50 °C and 150 °C. The fully aromatic sample does not exhibit shape memory behaviour, as it is unable to recover its original shape after deformation. For both supramolecular materials, at T = 50 °C, the restoration process requires approximately 10 s, while at higher temperatures (T = 150 °C) it occurs instantly. This thermally induced response highlights the presence of dynamic reversible interactions within the polymer network, which enable rapid and efficient shape recovery. The shape memory process occurs even at room temperature, although the complete restoration may require up to 20 min for PI-2 and coPI-3. This behaviour is particularly advantageous for space applications, where shape memory capability can be exploited for deployable structures and adaptive components. Moreover, shape memory polymers could be integrated into deployable components, antennas, solar arrays, or dust-mitigation systems, where controlled deformation and recovery are crucial for operation and compact storage during launch.

Conclusions

Fully aromatic (PI-1) and supramolecular semi-aromatic (PI-2 and coPI-3) materials with a thickness of approximately 300 µm were successfully fabricated in DMAc/IPA. All materials exhibited low density, particularly in the case of polyimides containing aliphatic diamine. High hydrophobicity was observed for all materials, while the surface free energy decreased for the supramolecular polyimides. These characteristics are particularly relevant for space applications, as low density contributes to mass reduction, while hydrophobicity minimizes moisture uptake, preventing defect formation during mission operations. Moreover, a low surface free energy helps reduce lunar dust accumulation on the surface. Optical microscopy revealed the ability of the semi-aromatic materials to fully recover their morphology after mechanical damage, with healing efficiency strongly dependent on temperature. Higher temperatures enhanced chain mobility and dynamic bond exchange, leading to faster repair. The semi-aromatic PIs developed in this work also demonstrate shape memory behavior suitable for advanced space applications. Their thermally triggered recovery allows compact storage and autonomous deployment of structural elements, reducing both mass and mechanical complexity in spacecraft design. This functionality, combined with intrinsic self-healing capability, provides a dual advantage for long-duration missions where maintenance and repair are particularly challenging.

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