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Improved metrics for assessment of immortal materials and products

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Abstract

An emergent vision for industrial sustainability is moving beyond the circular economy into the possibility of "immortal" products. This requires development of methods to enable the reliable and scalable production of novel products and systems that possess the inherent ability to sense and repair damage enabling in service healing and immortality. In the literature, this is mostly described by the self-healing property of the materials (polymers, metals, composites, ceramics and bio and non-bio hybrid systems). Self-healing systems are generally classified according to material type and self-repairing autonomy. This paper presents a brief review of existing immortal products and current methods of assessment. The paper presents an amalgamation of published research in the form of new process windows for the selection of self-heling materials and systems. Another contribution is made by the development of new metrics for assessing self-healing capability, with a vision to produce healing metrics that incorporate technical performance, as well as social, environmental and economic impact.

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Keywords: Self-healing, Sustainable manufacturing, Immortal products, Healing efficiency, Healing metrics

1. Introduction

The development of self-healing technologies is enabling smart materials [1] that can sense damage and autonomously heal. However, while novel materials may offer some energy savings and/or functional benefit [2], it is unclear of the social, environmental and economic impact these technologies might offer. Therefore, this paper argues that improved metrics must be considered if we are to develop immortal products and materials, with better understanding of the wider implications of integrating this technology into industrial applications. This paper will firstly, locate self-healing opportunities within Circular Economy, secondly, present a review of self-healing materials, their properties and their methods of assessment, and thirdly propose improved metrics for self-healing materials for a Circular Economy.

Nomenclature

η - Healing efficiency
e_h - Healing effectiveness
P_v - Critical property for the virgin product/material
P_d - Critical property after damage
P_h - Critical property for the healed product

1.1 Definition of self-healing and immortality

One of the main objectives in manufacturing nowadays is to extend life and functionality in applications. Particularly for products that are very common in everyday life, healing of functionality is vital. Healing is implemented by reversing failure of a property whether this corresponds to functionality or aesthetics or both. Self-healing is initiated "automatically" upon failure of a property and/or dis-function. Diesendruck et al [3] referred to healing of products as a reversal of failure

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and a substantial restoration of product impaired functionality and/or integrity. He claimed that healed products should also qualify for a manufacturer warranty as per new parts. Nosonovsky and Rohatgi [4] described self-healing as a thermodynamically expensive process that triggers a restoring thermodynamic force. In all cases, healing can be intrinsic or extrinsic, the healing materials can be synthetic or bio and healing can occur autonomously or semi-autonomously when a stimuli (e.g. heat, radiation, pressure, etc.) is applied [5].

1.2 Circular Economy and Self-Healing Assessment

The Circular Economy is a model that recommends industry and society transition from the traditional linear process of, make-use-dispose, to a practice that is more circular and regenerative by design [6, 7]. The main principles and intentions of a Circular Economy are to:

- *Preserve and enhance natural capital* - by ensuring materials and components can be separated and flow within either the technosphere or biosphere.
- *Facilitate effective use of resources* - by keeping materials, components and products cycling longer to extract the maximum value while in use.
- *Foster system effectiveness* – by designing out waste and developing adaptable, resilient systems that ensure the retention of value within the system [7].

The main aim of the approach is to keep products, components and materials at their highest utility and value at all times, while distinguishing between technical and biological cycles [8]. So, products, components and materials are reused, remanufactured or recycled and fed back into the system, reducing further extraction of resources. Self-healing materials and development of immortal products could assist in this endeavor and could potentially prolong the service life of many products and components through self-healing cycles. In Figure 1, the small circles in the maintain loop represents the self-healing cycles within the use phase of a product. However, self-healing technology and concepts are still emergent [9] and the wider environmental and circular implications have yet to be deliberated; therefore, in order to uncover what potential impacts self-healing technologies might have upon the circular system, a review of circular factors was conducted to understand which metrics for circularity of a new material or product are relevant.

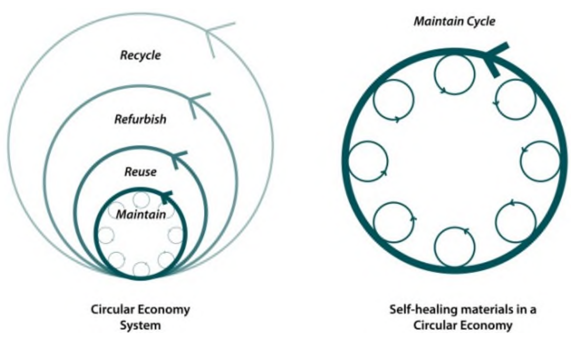

Fig. 1 Framework for self-healing applications in the circular economy system.

In the past, the vast majority of the developed self-healing materials/applications were validated by assessing the healing efficiency obtained. This was described as the recovery of usually a property or a degree of functionality. The most common way in literature to describe the healing efficiency is by Eq. 1:

$$\eta(\%) = \frac{P_h}{P_v} * 100 \qquad (1)$$

where η is the healing efficiency of a material property calculated by comparing the property status of the virgin (P_v) and the healed (P_h) material. For this evaluation a critical property linked to the damage has to be identified. Diesendruck et al [3] expressed healing efficiency as a function of the material property recovery over the deterioration of that property. This takes also into account the degree of damage before assessing the degree of self-healing. In this paper to avoid confusion in relation to equation 1, we define this as efficacy or healing effectiveness as expressed by the Eq. 2:

$$e_h(\%) = \frac{P_h - P_d}{P_v - P_d} * 100 \qquad (2)$$

where e_h is the healing effectiveness of a healing agent, P_h is the critical property for the healed product, P_d is the property after damage, and P_v is the property for the virgin first generation product.

Equation 2 is more relevant for assessing the effectiveness of different healing systems because it models the degree of damage and damage healing. However, both metrics in Equation 1 and 2 are applicable to a single type of damage, and model self-healing as restoration of a single material property. The metrics do not deal with cases of materials impaired to more than one property. From a damage perspective, for instance when a crack is healing, the healed material is tested and the material tensile strength might be restored. However there might be other properties like flexural or impact strength of the material that haven't returned to the original functionality. This could prove crucial in applications designed for multiple utilities. There is thus a need for research on metrics for evaluating healing for multiple failure modes on incorporating constraints for other important properties.

2. Process Windows for selecting self-healing materials

There are a number of papers within the literature on self-healing materials that discuss the type of induced damage, the healing conditions, and the evaluation of healing methods as well as the best efficiency achieved.

2.1. Self-healing in polymers

There are several methods of self-healing polymers in the literature classified either as intrinsic, where there is an internal design and/or chemistry that triggers healing without any external interaction, or extrinsic methods that require an added stimuli to be provided in the system. Example healing triggers include thermal [10, 11] and electrical [12] treatment, the use of water [13, 14] or a photo induced approach [15-17], acid [18] or multi-pH [19] or even salt responsive [20] healing systems. Intrinsic mechanisms mainly involve supramolecular polymers [21-23], hydrophobic interactions in hydrogels [24], covalent bond gels [25], host-guest monomers or polymers [26] and metallo-gels [27] among others. There are also hybrid systems in which both extrinsic and intrinsic healing can occur [28-31].

2.2. Self-healing in polymer composites

The repair of damage-induced crack in polymer composites has been well discussed for both intrinsic and extrinsic strategies [3, 32-34]. Some of the healing mechanisms in composites involve healing agent filled capsules dispersed in the matrix [35, 36]. The healing agent can be a hardener that fills and heals cracks or can re-establish anticorrosive surface properties [37], conductivity [38] and others. Additionally, there are hollow-fiber reinforced polymer composite technologies with combined embedment of healing agent and hardener [39]. Vascular networks enable a system of vessels to deliver the healing agent to larger and multiple damaged areas [40, 41].

2.3 *Other self-healing systems*

Many other self-healing applications can be found in literature expanding the scope of self-healing in metals and in particular in anticorrosive metal coatings [43] or in food packaging and underwater applications [44]. Soldering using

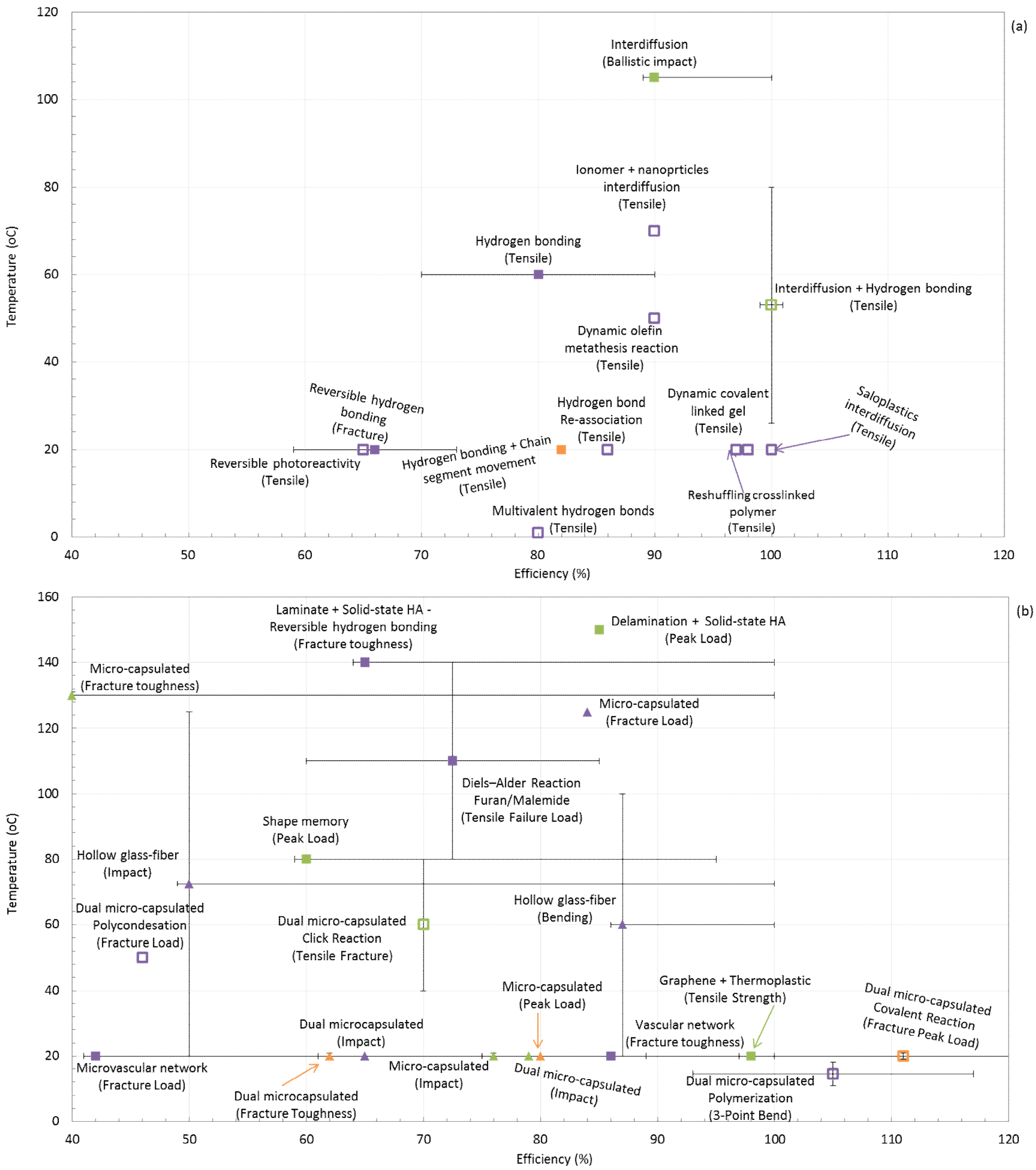

Fig. 2 The self-healing efficiency plotted against the healing temperature for different (a) polymer and (b) polymer composite systems for which self-healing is repeatable (solid square), may be repeatable (non-solid square) or is non-repeatable (triangle) process. The colour coding subdivides the healing process time into scales of ⩽1 hour (green), ⩽ 24 hours (purple) or > 24 hours (orange). ***Process window developed from review data in*** [42].

polymeric capsules has been used to repair conductivity [45] and shape memory alloys have been used as reinforcement and as tool to minimize crack volume on both polymers [46] and low melting temperature alloys [47]. Additionally, the electrochemical healing approach has been investigated by electroplating structural metals [48].

Figure 2 presents process windows for healing efficiencies achieved as a function of the healing temperature for different polymer (Figure 2a) and polymer composite (Figure 2b) self-healing systems as informed by the literature. The scale bars represent the reported temperature range of healing and the range of healing efficiencies achieved. The different self-healing systems are also classified based on the time scale of healing and whether healing is applicable for several cycles or not. The following information was inferred:

- Polymer composites can achieve higher healing efficiencies than polymer healing systems. In fact, there were cases that the healing strategy led to enhanced functionality.
- A number of polymer healing systems reported in literature can self-heal more than once (Figure 2a).
- There are cases where the healing efficiency is a dependent property that is evaluated and, for the same polymer composite the recovery varies for different mechanical properties (Figure 2b).
- Time of healing could be a critical factor for an immortal product for healing in-situ under operating conditions.
- Reported polymer systems mainly require a day or less to repair while the healing times recorded for polymer composites are more varied.

2.4 *Circularity metrics for immortal products and materials*

Even though the Circular Economy has developed into an established area of interest for industry and academic research, thus far, very few methods, metrics or factors have been proposed that measure the circularity of product or material [49-52]. However, of those developed, three pieces of research were identified as relevant to this study, each proposing these different sets of factors. Firstly, developed by the United Kingdom, Waste and Resources Action Programme (WRAP) [53], suggest that when developing any new material or product for closed-loops systems, these factors need to be considered:

- *Lifecycle analysis* which helps identify the key lifecycle impacts of the materials, product or service.
- *A closed-loop approach* which should be deliberated from the start.
- *Strategic analysis* of future scenarios where resources are tightly constrained.
- *Business models for resource efficiency* that can adapt and best fit these future scenarios.
- *Design of the product*, i.e. design for modularity, remanufacture or disassembly and material choice impact analysis.
- *Consideration of manufacturing and end-of-life* to ensure whole life cycle is analysed.

Whereas, the Cradle to Cradle Product Innovation Institute [54] analyse and certify products according to these criteria:

- *Material Health* which determines whether the product is manufactured using optimised non-toxic materials and chemicals.
- *Material Reutilization* which evaluates the recovery potential for materials and components so they can be safely and efficiently be returned to flow within their respective cycles i.e. Technosphere and Biosphere.
- *Renewable Energy and Carbon Management* which quantitatively measures the percentage of renewable energy utilized in the manufacture of the product, as well as the embodied energy associated with the product from cradle to gate.
- *Water Stewardship* which qualitatively and quantitatively evaluates the water usage and waste as a result of the manufacturing process.
- *Social Fairness* which qualitatively assesses the impact of a products manufacture upon people, communities and the general environment.

While these two sets of factors outline a comprehensive approach for developing circular product systems and provide specific and measurable metrics to consider, there is still opportunity to explore more detailed and quantifiable elements which is what the 'Material Circular Indicators' offers. Developed in partnership between Granta and the Ellen MacArthur Foundation, this tool measures the 'restorative material flows of a product or company' [51], to help designers and companies assess how circular their products or offering could be. Firstly, it assesses a material within two main parameters: **Feedstock** (where it comes from) and **Destination** (where it goes after use), and measures how much of the material can be reused and recycled, while also considering the efficiency of the recycling process as well. Then secondly considers, the **Lifespan** (measured against the industry standard) and **Functional units** (measured as the number of uses during its lifetime). Within this model, the two factors that are most relevant to the vision of immortal products and aims of the paper are, namely Lifespan and Functional units. Essentially these represent the 'Utility' of the material or system [51, 52]. These two factors provide a means through which to describe the potential positive and negative outcomes that could occur in relation to the healing efficiency, which will be discussed in the next section of the paper.

3. Discussions and Concluding Remarks

The identified metrics that can be linked between the technical assessment of immortal materials and their sustainability dimensions are presented in Table 1. This table shows how these two sets of metrics, (self-healing and circular) can converge and be potentially explored as one model. The ability to assess not only the circularity, but the impact and value of immortal products is an important step in the development of self-healing, as this will help assist this technology be more widely considered for implementation in industrial systems in the future. The next steps for this research would be to explore how these metrics can converge to form an assessment tool for immortal products. Table 1 shows how the factors are related.

Table 1 A list of comparable factors between the self-healing and circular economy metrics.

Self-healing	Context in self-healing	Circular Economy Comparator
Healing efficiency	Expresses the extent to which the functional property is restored. This gives a direct measure for design.	Could relate to the warranty that can be offered for a remanufactured product compared to a virgin product.
Healing efficacy or effectiveness	The healing efficacy and takes into account the size of the damage that had been inflicted and the resulting extent of recovery.	Could relate to the overall lifespan of the product and no. of functional uses it could achieve.
Healing Cycle time	Can be critical; depending on the application, lost functionality and risk assessment.	Relates to the time needed to repair, remanufacture which affects viability
Healing Repeatability	This will directly affect the overall lifespan and number of functional uses a product can have	Relates to no. of repair and remanufacture cycles that can occur

Aside from the more detailed metrics presented in review for examination, there are wider factors of sustainability assessment that can be considered, some of which include; long term social and environmental impact; an understanding of how these materials will flow within the system; effectiveness of energy needed to self-healing over the embodied energy; material health of the components used; cost and user convenience. While all the factors listed within this paper are relevant, Table 1 provides a starting point for developing of an immortal product assessment tool and represents a more detailed and improved perspective on the metrics for self-healing technologies. Taking also into account Figure 1 the immortal product can be considered as a super enabler of a circular economy that retains the highest value in products for subsequent use.

For the future it is necessary to develop a framework for assessing the extent to which self-healing is environmentally beneficial and under which conditions self-healing materials are preferable over materials without this property. A life cycle assessment based on the self-healing factors presented in Table 1 could help evaluate the environmental performance of self-healing materials. An option is to consider the lifetime impact over an infinite period of assessment on a value based judgment. The assessment is based on comparing the impact of a single life product to that of a multiple life product over a defined period of service. This will be addressed in a future paper dedicated to the development of life cycle assessment and case studies for immortal products.

The future outlook and challenges are to design and develop materials and products with the fastest healing cycle according to the criticality of the application. There is also a need to develop a holistic healing technology that is able to heal more than the critical damage i.e. not compromising other important properties or functionality. Developing mathematical models for the healing will enable optimization and the inferred evaluation of impacts of immortal products. This technology is vitally important and has already found applications in concrete [55], fuel tanks [56] and deep sea cables [57] among others. It is therefore timely to consider the performance metrics and sustainability assessment of such technology.

Data Statement

This is a review, synthesis and framework development paper, and therefore all data underlying this study is cited in the references.

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