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# Review Article

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# **Green Materials**





# Sustainable feedstocks for 4D printing: biodegradable polymers and natural resources

#### Leena V. Bora

Department of Chemical Engineering, Institute of Technology, Nirma University, Ahmedabad, India (corresponding author: leena.bora@nirmauni.ac.in)

### Kevin S. Vadaliya

Department of Chemical Engineering, Institute of Technology, Nirma University, Ahmedabad, India

#### Nisha V. Bora

Department of Mechanical Engineering, Lalbhai Dalpatbhai College of Engineering, Ahmedabad, India

Additive manufacturing is an emerging technology supported by 'Industry 4.0'. When combined with a stimulus-responsive behavior, incorporating the fourth dimension of time, it results in a manufacturing technique known as four-dimensional (4D) printing. 4D-printed entities have the distinctive characteristic of property transformation, under the influence of a drafted stimulus, which can be cleverly engineered for the desired application. The conventional stimulus-driven smart printing inks deployed in 4D printing are non-biodegradable polymers that pose a challenge to sustainability. Hence, it is imperative to appraise the utilization of sustainable raw materials, composed of natural and synthetic biodegradable polymers, as feedstocks for 4D printing. Natural resources have fluctuating properties, which make them receptive toward intelligent engineering. This review is an effort toward the implementation of sustainable feedstocks as printing inks for 4D printing, for eventual environmental benignity. It covers several sustainable raw materials for 4D printing and the strategies to use them in conjunction with conventional inks in order to bring down the volume of non-biodegradables. This review can serve as a reference for designers and engineers wishing to use sustainable inks for 4D printing, thereby boosting the momentum needed to consolidate this next-generation technology in line with the UN Sustainable Development Goals.

Keywords: 3D printing/4D printing/biodegradable/green polymers/natural materials/stimuli/sustainable/time

## 1. Introduction

'Industry 4.0' (a term used interchangeably with 'the Fourth Industrial Revolution') is professed to be technology driven, which enables higher efficiency and productivity.<sup>2,3</sup> It involves increased automation through software algorithms for realization of cyberphysical transformations. Additive manufacturing (AM) constitutes one of the nine pillars of Industry 4.0,4 which aids in accurate and controlled printing and morphing, catering perfectly to the intended application. The history of four-dimensional (4D) printing begins with the inception and development of three-dimensional (3D) printing, one of the manufacturing techniques encouraged by 'Industry 3.0'. After a substantial amount of research on 3D printing with respect to design, printing techniques, algorithms and applications, it was in the year 2012, during a TED conference at the Massachusetts Institute of Technology, that Tibbits<sup>5</sup> introduced the concept of 4D printing. One of the definitions that perfectly explains 4D printing is '4D printing is the evolution in the shape, property, and functionality of a 3D printed structure with time when it is exposed to heat, light, water, pH, etc.' (p. 2<sup>6</sup>). Since then, research in this direction has started and, owing to its remarkable features, gained quick momentum and popularity.7 On account of the characteristic attribute of 4D printing of controlled property morphing

over time, <sup>8,9</sup> 4D constructs can be cleverly designed to adapt the desired property for the required application. For example, 4D printing has recently begun to find practical applications in renewables, <sup>6,10</sup> textiles, <sup>11</sup> electronics, <sup>12,13</sup> biomedicines, <sup>12</sup> agriculture, <sup>14,15</sup> aerospace, <sup>16–18</sup> purification <sup>19</sup> and so on.

Figure 1 shows the four basic attributes of 4D printing – namely, (a) the AM processes used for printing; (b) the smart materials (SMs), which undergo a user-defined transformation (of shape, color, volume, flavor etc.) by establishing communication with their environment (stimulus); $^{6,20-22}$  (c) the stimuli bestowed to actuate the printed material; and finally, (d) the potential applications.

The technological revolution supported by Industry 4.0 also motivates the use of renewable resources and encourages sustainability.<sup>23,24</sup> Fortifying the circular economy,<sup>25,26</sup> vitalizing biodegradable feedstocks in the printing technique and devising sustainable constructs encourage the possibility of implementation of 4D printing in the sustainable domain. Providentially, natural resources exhibit fluctuation in properties<sup>27</sup> and thus can be engineered intelligently for a specific application.