

Review

Concepts and Terminologies in 4D Printing

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Abstract: 4D printing (4DP) is a promising technology that enables additive manufactured parts to be programmed for actuation, reducing the need for external power or electromechanical systems. As this area of research has grown exponentially, this review paper aims to define and establish fundamental concepts and terminologies used in the field of 4DP. The objective is to encourage researchers to adopt a more consistent approach and a standardized set of vocabulary associated with this emerging field. Even though the paper covers the most widely used definitions, the multidisciplinary nature may mean that certain words could be used interchangeably or have a different meaning in another context.

Keywords: 4D Printing; additive manufacturing; shape memory material; shape change behaviour; terminologies; definitions; ontologies; standards

1. Introduction

4D Printing (4DP) is an emerging technology that offers unique advantages over conventional additive manufacturing (AM). 4DP parts can be programmed for actuation without the need for external power sources or electromechanical systems, minimizing the probability of failure and reducing the number of components. The field of 4DP has grown rapidly with more research and published material being produced over the last five years. A Scopus search on publications related to 4DP showed 86 unique publications in 2017, followed by 115 publications in 2018, and 189 publications in 2019. As a result, there is an urgent requirement for a better and a more unified understanding toward fundamental concepts and terminologies used in 4D Printing. This paper builds upon existing work regarding the ontologies for additive manufacturing, including, but not limited to, International Organization for Standardization (ISO) TC261) and the American Society for Testing and Materials (ASTM International) F42's efforts toward the standardization of terminologies [1], the work of the National Institute of Standards and Technology (NIST) [2], and other publications [3,4]. Due to the limits of space, this paper will define and establish the fundamental concepts and terminologies used in 4DP. The objective is to encourage a more consistent approach and a standardized use of vocabulary associated with this emerging field. Although this paper aims to offer a comprehensive coverage of definitions, the multidisciplinary nature of this field may mean that some terms could still be used interchangeably or have a different meaning in another context. As this technology continues to evolve, these definitions should be continually revised and updated.

2. Fundamental Concepts

The section introduces important underlying concepts associated with 4D printing (4DP). The term “4D Printing” was originally proposed by a research group at the Massachusetts Institute of Technology [5], who defined this as the ability of additively manufactured objects to change in shape or structure when exposed to external stimuli over time [6–8]. This enables products that can “self-assemble”, where the components are able to autonomously structure and reorganize without

human involvement, such as performing self-folding actions for assembly [9]. This self-folding phenomenon is associated with “shape change behaviour”, or “shape-shifting behaviour”, where 4DP structures have the ability to morph in a controlled way, over a determined period of time. A review by [10] classified basic shape change behaviours to include folding, bending, rolling, twisting, helixing, buckling, curving, topographical change, expansion, and contraction.

Complex shape-changing behaviors consist of multiple deformations by extending an earlier form of a basic shape or as a completely multi-faceted form. This includes complicated structures, such as waving and curling. Multiple shape-changing behaviors can also be combined in a single component. The term behaviour is associated with the “morphing” or “transformation” occurring in the structure or displaying changes to the shape and size of the construct. At a systems level, “hierarchical structures” combine various forms of shape-changing components into a series. These hierarchical self-morphing structures involve concurrent, sequential, and multi-stage shape-changing behaviours at both the system (global) and component (local) level.

4DP parts can be selectively programmed to enable different locations to behave at predetermined intervals of time [11]. In terms of behaviour, the term “shape change effect” (SCE) is used to describe the property of switching between one state when exposed to external stimuli and returning to another state when that stimuli is removed or when exposed to a different stimuli (Figure 1) [7]. To describe this state of change, the “shape memory effect” (SME) is the result of programming that determines both the extent of the state of change and the desired shape when external stimuli is applied [12]. In particular, “shape memory” is a characteristic of a unique class of materials that are capable of being programmed into a temporary shape and able to return to the original shape when exposed to external stimuli [7].

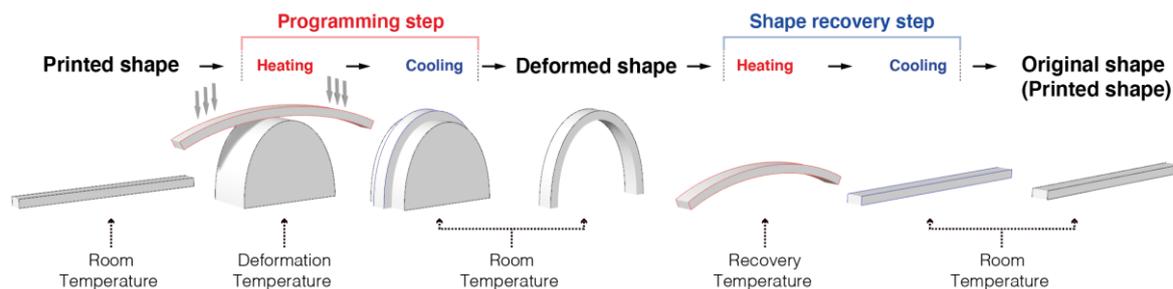


Figure 1. The processes involved in the shape change effect.

Broadly, there are three classifications for the shape memory effect. A “one-way shape-memory effect” occurs when an original shape is recovered, and a new programming step is needed to re-create a temporary shape [8]. A “two-way shape memory effect” occurs when a material can remember two different shapes in a reversible manner, where one shape is memorized at low temperatures, and another memorized at a higher temperature [13]. “Multiple-shape memory effect” involves composites that have more than one reversible switching transitions and can remember one or more shapes in addition to its original permanent shape. This requires multiple thermomechanical programming steps [14]. Multiple-SME is generally meant to have additional intermediate shapes between the permanent and temporary shape. In certain instances, only a couple shapes are reversible and one step-programming is also possible.

Recent studies have investigated applications for 4DP, usually through the principle of “actuation”, which is a mechanical action based on hydraulic, pneumatic, electric, thermal, or mechanical means. An actuator is a device that moves or controls a mechanism. A 4DP actuator can be designed to be capable of mechanical movement or shape change behaviours, such as bending, twisting, or folding. “Self-actuating” is said to refer to the automated actuation process in response to an external stimulus [15]. At the macro level, “origami engineering” refers to origami principles being used in engineering applications, enabling reconfigurable structures that can fold and unfold upon

demand [16]. Within the 4DP context, “active origami” describes the principle of achieving a flat-pack structure, in which an object can self-fold or self-unfold. Examples include a self-assembling box and a pyramid that self-assemble from a flat shape.

Other researchers have constructed “bistable” structures that support two stable states, where reversible switching between stable configurations can be achieved under controlled mechanical actions. Bistable structures do not require additional energy to maintain their stable states and can work as mechanical switches, or to simplify actuation and motion control [17]. Other researchers have explored “auxetic structures” where the structures or materials have a negative Poisson ratio. These structures are thicker, contrary to the force being applied when extended. This is due to the internal design and the way they deform when the samples are uniaxially loaded [18].

3. Properties of 4D Printed Parts

This section describes the properties of 4DP, the factors that influence the 4DP process, as well as the materials that are currently investigated and often used in the literature. In terms of the key properties, the shape fixity, shape recovery and repeatability are often used to describe the characteristics of 4DP parts that are attributed to the quality of the components. In some instances, “repeatability” is an important aspect of 4DP parts, having the ability to repeat the entire cycle without pronounced fracture or significant change to the permanent shape [19]. The “shape fixity” is the extent of a temporary shape being fixed for a “shape memory polymer” (SMP) [20]. The material properties of SMPs are described in Section 3.2. The “shape fixity ratio” (R_f) is defined as the ability of a 4DP part to instill a mechanical deformation applied during the programming process (1). The calculation is derived from the ratio of the strain ϵ_{unload} measured upon removal of the load after the cooling process, to the strain ϵ_{load} that was subject at a specific temperature above the T_g [21].

$$Rf (\%) = \frac{\epsilon_{\text{unload}}}{\epsilon_{\text{load}}} \times 100. \quad (1)$$

“Shape recovery” is defined as the ability of a polymeric material to memorize the original shape from a temporary shape [20]. This “temporary shape” is also known as the “deformed shape”. SMPs can be deformed and fixed in a temporary shape, in which they recover to their original, permanent shape when exposed to a stimulus. The “shape recovery ratio” (R_r) is defined as the ability of a material to be able to remember its permanent shape (2). This is a measure of how much applied strain can be recovered upon being subject to a stimulus. This is the ratio of the difference between the initial strain (ϵ_i) applied at the deformation step, and the final strain (ϵ_f) measured after end of the recovery step to the initially applied strain.

$$Rr (\%) = \frac{\epsilon_i - \epsilon_f}{\epsilon_i} \times 100. \quad (2)$$

The “Shape recovery time” (t) is the duration that it takes for a material to reach its recoverable strain at T_r (3). This characterizes how fast an SMP responds to the stimulus. This parameter is mostly reported by studies that video-record real-time shape changes [22]. The “shape-recovery rate” specifically refers to the speed of the material to recover its original shape. V_m represents the maximum shape-recovery rate, S_i indicates the recovery deflection, and t represents the time interval [23].

$$V_m = \max[(s_{i+1} - s_i)/t]. \quad (3)$$

3.1. Factors Influencing 4D Printing

Acknowledging the earlier work by Momeni et al. [8], an extended conceptual framework for 4D Printing was proposed by Nam and Pei [10] (shown in Figure 2) that demonstrated the important factors of 4DP, comprising the AM process, interaction mechanism, mathematical modelling, stimuli responsive material, and the stimuli that influence the result. These factors contribute toward the type of shape change behaviour, the position of the deformation, and the time taken. Repeated cycles of shape change effects may lead to the “degradation” of 4DP parts, causing cracks and delamination

of the material. If a heat source is involved, then the term “thermomechanical degradation” is used. Optical microscopy and Scanning Electron Microscope (SEM) can be used to evaluate the morphology of the material microstructure and to assess the severity of degradation.

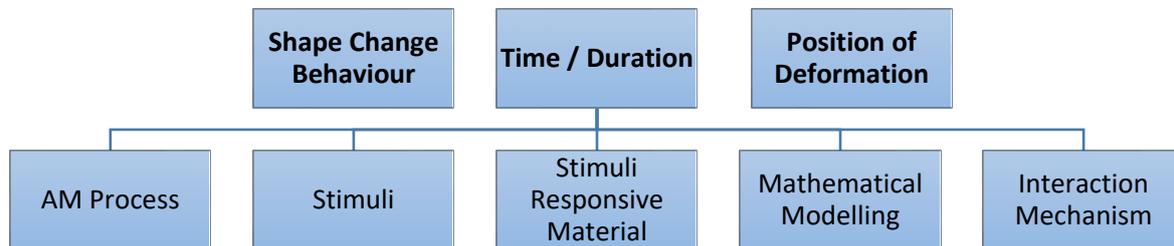


Figure 2. Conceptual framework for 4D printing (4DP) [10], adapted from Momeni et al. [8].

There are seven AM processes described in ISO-ASTM 52900:2017 [1] that establish the processes used in AM technology. Although, in principle, all seven AM processes could be adapted for 4DP, most studies are related to polymers and composites that primarily use material extrusion, material jetting, and vat photopolymerization. Studies related to the use of metals often involve selective laser melting, which is classed as a laser powder bed fusion (L-PBF) process. The definitions of AM processes frequently used in 4DP include “material extrusion”, in which material is selectively dispensed through a nozzle or orifice; “material jetting”, in which droplets of build material are selectively deposited; “powder bed fusion”, in which thermal energy selectively fuses regions of a powder bed; and “vat photopolymerization” in which liquid photopolymer in a vat is selectively cured by light-activated polymerization [1].

Within the 4DP framework, the “interaction mechanism” is also known as the “programming” process, in which the material is deformed into a temporary shape. When heat is involved in the programming process, the term “thermomechanical programming” or “constrained-thermo-mechanics” is used. In this approach, the stimulus is heat and the smart material retains the shape memory effect. The compressive strain during the loading process fixes the programmed state. The use of jigs, forming templates, and fixtures are required during the application of the mechanical load [23]. According to Momeni et al. [8], this constitutes a four-step cycle. First, the structure is deformed by an external load at a high temperature; second, the deformation (strain and constrain) is maintained; third, the structure is unloaded at the low temperature and the desired shape is achieved; fourth, the original shape can be recovered by reheating the structure. “Deformation” refers to the shape of an object being changed by applying force, and pressure known as “loading”. The deformation of the object depends on the force, the material, and whether the material is in an elastic deformation or a plastic deformation.

In certain studies, “integrated programming” is used to refer to programming steps that are instilled during the fabrication process, thereby eliminating the need for separate thermomechanical programming [24]. Studies described that the AM process may influence the 4DP effect, leading to a phenomenon known as “pre-strain”. Bodaghi [25] reported that, for material extrusion, the print speed and nozzle temperature may impose a significant effect on the deformation. The hotter the nozzle temperature, the less the pre-strain. Increasing the print speed increases the pre-stain value and consequently influences the bending deformation. The molten polymer is subject to stress, ‘with the deformation energy being stored elastically’. When the molten polymers are extruded, ‘stresses are relaxed and the elastically stored energy is released, leading to radial expansion of the melt’, which could influence the pre-strain (ibid).

“Mathematical modeling” is used to compute and predict the material distribution and the structure needed to achieve a desired change of shape. This can provide engineers with information regarding the shape-shifting as a function of time, the prevention of collisions between components of the structure during self-assembly operations, and can reduce the number of trial-and-error experiments [8]. “Stimuli-responsive materials” are substances that can receive, transmit, or process

a stimulus. They respond by producing a useful effect, such as an actuation mechanism or a signal that the material is acting upon [6]. This is associated with the term “stimuli” (plural) or “stimulus” (singular), which is a trigger that initiates the shape change of a 4DP part. Examples of stimuli that researchers have used include water, heat, UV light, pH, magnetic sources, or a combination of multiple stimuli. The type of stimuli depends on the material that has been selected as well as the intended application. In some literature, the term “trigger” or “triggering mechanism” can refer to the stimulus.

Within the inner core of the 4DP conceptual framework, the “position” or “location” specifies where the shape change behaviour will take place within the printed part; and at a specified period of “time”, which is the starting point as well as the entire duration that the 4DP behaviour is expected to occur. “Shape change behaviour” has been described in the earlier section to include 12 different types of deformation [10]. Lauff et al. [16] provided similar definitions of behaviours, such as “bending”, which is a distributed curvature caused by the deformation of a material along the deflected area that creates a curvature; and “folding”, which is a specific and localized deformation of a material along a crease pattern to create a sharp shape.

3.2. Material Properties and Materials Associated with 4D Printing

The earlier section described stimuli-responsive materials that respond by producing a useful effect, such as actuation mechanisms or a shape change behaviour. In general, most stimuli-responsive materials used in 4DP are “shape memory materials” that are a subdivision of multi-phase smart materials. They can recover from a deformed state to their initial shape, known as the “shape memory effect”. Shape memory materials exhibit a plastic deformation (temporary shape) when an external stimulus is applied, and they are able to recover to their original shape [12,26]. “Shape changing material” also refers to the reversible material properties that can spontaneously change shape on the application of stimuli and have the ability to regain their original shape upon the removal of stimuli [27].

Other classes of matter include “smart materials”, which are substances that are able to couple or convert energy between various physical domains, such as converting thermal energy from heat into mechanical work [28]. In particular, “thermo-responsive materials” are those that are sensitive to temperatures. The temperature is transferred to the material through a medium, such as water, UV light, or other sources. “Metamaterials” refer to a category of multi-scale structures that widely exhibit thermomechanical characteristics not usually found in nature, and instead utilize properties from the design of the structure and geometry [29]. “Tunable materials” are substances that efficiently adapt to a dynamic environment. For example, Yang et al. [30] demonstrated mechanically tunable lightweight metamaterials utilizing 4D printing produced using photo-crosslinkable and temperature-responsive SMPs. Bodaghi and Liao [31] also introduced tunable metamaterials with reversible thermo-mechanical memory operations. The term “functionally graded materials” (FGMs) is used to describe substances that are designed and fabricated for specific functions and applications, by means of varying the composition and the structure gradually over the volume, resulting in changes in the material properties. FGMs fabricated by additive manufacturing are known as Functionally Graded Additive Manufacturing (FGAM), and examples include the varied densification of concrete [32].

The term “digital materials” is also cited in literature, often making reference to the material jetting process from Stratasys Objet Connex machines that work by mixing two base materials, made up of specific ratios of rigid plastic VeroWhite as an “inactive material” that remains inert, and a rubbery TangoBlack as the “active material” that responds to stimuli to achieve the desired shape memory behavior [33]. These customized materials are also known as “programmable materials” whereby the properties can be modified using a software such as GrabCAD to specify the desired properties of a material [34]. Voxel-based modeling editors such as Autodesk Monolith can be used to represent a three-dimensional bitmap, represented as a “voxel” that contains a singular material. When an array of different voxels made up of distinct materials in a design part are combined, a multi-material structure can be achieved. The layer wise deposition of material enables engineers to control the density and

directionality of material deposition as well as to combine various materials to produce a seamless monolithic component.

According to Loh et al. [35], complex matter can be produced through (1) variable densification within a homogeneous composition; (2) heterogeneous composition through simultaneously combining two or more materials through a gradual transition; or (3) using a combination of variable densification within a heterogeneous composition. A “homogeneous” composition consists of a single material in a single component; a “heterogeneous” composition consists of a mixture of two or more materials in a single component. They can be a combination of smart or conventional materials or using various smart materials that react differently under various stimuli [36].

“Hydrophilic materials” are sometimes used in 4DP applications. These substances are easily wetted and have more thermodynamically favorable interactions with water. Certain hydrophilic materials, when exposed to water, will lead to absorption that can increase in its volume substantially, evoking a shape change effect through linear expansion. “Hydrogels” belong to the group of hydrophilic polymers that can swell in water and hold a large amount of liquid while retaining the structure. 4DP hydrogels are responsive to hydration and temperature in which the hydrogel swells, and a transformation is initiated. Examples of natural polymer-based hydrogels include Dextran, Chitosan, and Collagen; synthetic polymer-based hydrogels include Poly (vinyl alcohol).

The “swelling ratio” is defined as the fractional increase in the weight of the hydrogel (or a similar hydrophilic material) due to water absorption. The “porosity” of the material, which is the measure of void spaces can also significantly improve the response speed of the shape change effect [37]. Another characteristic in 4DP applications is “anisotropy”, which refers to the physical properties that are directionally dependent. Within the AM process, there is a certain degree of anisotropy in the mechanical properties, due to the layer wise method of fabrication. In a 4DP context, the mechanical properties in terms of the tensile strength in a glassy state and its elongation in a rubbery state are important factors to consider when programming shape memory materials. These two properties dictate the capability of shape fixity and the recovery, which are consequential for the function of SMPs [21].

Other research studied the use of “printed active composites”, which are typically soft composites made up of glassy SMP fibers and the stacking of laminas to reinforce the elastomeric matrix. These laminates undergo thermomechanical programming to achieve shape change behaviours. Other forms of “elastomeric matrices” have been described in Lee et al. [38] where they described elastomers as having more free volume than other polymers, which accelerates the rate of swelling. The swelling induces internal stress, in which in the recovery step converts the stored potential energy into elastic energy that acts as an actuation device. A paper by Akbari et al. [39] discussed the use of “Joule heating”, which is a controlled process in which passing an electric current through an electrical conductor produces localized thermal energy in the form of heat. This is also known as resistive, resistance, or Ohmic heating. Localized Joule heating can be used to initiate active hinges in 4DP parts and constantan, a copper-nickel alloy, has been used for such purposes.

In terms of the materials commonly used for 4DP, shape memory alloys and shape memory polymers are the most widely studied. “Shape memory alloys” (SMAs) are popular smart materials that possess two phases: A martensite phase (low temperature) and austenite phase (high temperature). These two phases allow SMAs to alter their shapes and return to the original shape when exposed to high temperatures [40,41]. SMAs are thermomechanical alloys, in which, once treated to acquire a specific shape, they have the ability to indefinitely recover from large strains without permanent deformation and to remember their original geometry.

For example, after undergoing a physical deformation, an SMA wire can be heated through resistive heating to its final transformation temperature (A_f) and regain its original shape [42]. This is mostly applicable to NiTi as others, such as Cu based wires, have low electrical resistance and cannot be easily joule heated. “Nickel-titanium” (NiTi) is a class of SMA that exhibits excellent shape memory behaviour including a high percentage of shape recovery, a large super-elastic strain, and recovery

stress [43]. Studies have shown that NiTi parts can be additively manufactured using selective laser melting (SLM) [44,45]. “Copper-aluminum-nickel-manganese” (Cu-Al-Ni-Mn) is another class of Cu-based SMAs that could be potentially produced using SLM, promising good shape recovery, high transformation temperatures, and suitability for industrial applications [46].

In terms of polymeric materials, “shape memory polymers” (SMPs) can be programmed to memorize a less-constrained shape or configuration, and subsequently assume a strained temporary shape, and then revert to the memorized configuration upon being triggered by an external stimulus [47]. Although most polymeric materials have a heat/chemo-responsive SME, the polymeric materials used in additive manufacturing have a creeping and relaxation problem. “Polylactic acid” (PLA) is a common SMP that is widely available in a filament form and regarded as suitable for 4DP. Generally, most PLA materials have a glass transition temperature of 60 °C and an elastic modulus of 3.5 GPa in the glassy state. The group of “thermoplastic elastomers” (TPE) that exhibit properties of SMPs include thermoplastic (TP), styrenic block copolymers (TPS), polyolefin (TPO), polyurethanes (TPUs), copolyester (TPE), and polyamides [48]. “Thermoplastic polyurethane” (TPU) contains characteristics of rubbers and plastics with excellent biocompatibility and flexibility. TPU has been widely used in medical devices. TPU has high elongation, moderate tensile strength, a moderate Young’s modulus, and strong abrasion and tear resistance [49].

For SMPs, the “glass transition temperature” (T_g) or “transition temperature” (T_{trans}) is used to describe the temperature beyond which a polymer changes from a hard, glass-like state to a rubber-like state. For the ease of comparison with conventional polymers, a single T_g value taken from the midpoint of a broader transition is often used in literature. However, since the width of glass transition may have a profound impact on the shape memory performance of SMPs, most studies report the width of the transition along with the midpoint T_g value, or the onset glass transition temperature ($T_{g\ onset}$) and the ending glass transition temperature ($T_{g\ end}$) along with the midpoint T_g value [22]. The “shape deformation temperature” (T_d) is recognized as the working temperature in which the SMP is strained to a temporary shape. The relationship of T_d is relative to T_{trans} and it has a significant impact on the shape memory performance of the SMP. The “temporary shape fixing temperature” (T_f) is the working temperature at which the temporary features of the SMP are fixed. T_f is often lower than T_{trans} . Lastly, the “shape recovery temperature” (T_r) is the working temperature that the SMP is triggered at in order to recover from a fixed temporary shape. Usually the T_r is higher than (T_{trans}) and is often chosen to be the same as T_d (ibid).

3.3. Experimental Measurements Associated with 4D Printing

An “extensometer” is used to measure changes in the length of components, suitable for stress–strain measurements and tensile testing. Examples include contact-type measurement extensometers, non-contact video extensometers, and non-contact laser extensometers (laser interferometry). The “gaussian curvature” measures the curvature, based on the distances measured on the surface, since 4DP parts are able to produce patterns of spirals, grids, and combinations of spirals and grids that produce negative Gaussian curvatures, positive Gaussian curvatures, and a combination of positive and negative Gaussian curvatures, respectively [50]. A “strain–temperature curve” is a graphical representation that indicates the strain (%) achieved in the Y-axis, and the temperature recorded in the X-axis (°C). The strain is a measurable quantity of force and the strain on an object strictly depends on the external force being applied. “Poisson’s ratio” (ν) is the ratio between the lateral contractile strain and the longitudinal tensile strain in the longitudinal direction of a tensioned material. It indicates how much thinner a material is when it is stretched [18]. A “force displacement graph” can be useful to study the effect of a shape change behaviour and an activation force by parameterizing the associated force (N) with a varying length (L). These graphs are particularly useful for developing the design of actuators [51]. “Young’s modulus” or the elastic modulus is used in studies to measure an object or substance’s resistance to being deformed elastically when a stress is applied on it.

4. Future Work

This paper provided an extensive review from the state-of-art literature, clarifying key terms and concepts, with the main concepts and terms summarized in Table 1. As the field of 4D printing continues to evolve, our understanding of the interaction mechanism, new forms of mathematical modelling, better characterization of stimuli responsive materials, and newly discovered stimuli will enable improved control of 4DP. As such, the domains surrounding aspects of testing, part inspection, and new applications will continue to grow. As the technology continues to evolve, the terms and definitions will need to be updated.

Table 1. Summary of the concepts and terminologies for 4D Printing.

Stage	Start	Programming Step		Temporary Shape	Shape Recovery Step		End
Process	Printed Shape	Subject to Stimuli, e.g., Heating	Removal from Stimuli, e.g., Cooling	Deformed Shape achieved	Subject to Stimuli, e.g., Heating	Removal from Stimuli, e.g., Cooling	Return to original shape (Printed Shape)
Phenomena	-	Shape change effect			Shape memory effect		-
Activity	-	Interaction mechanism		Loading	Shape change behaviour/Shape-shifting behaviour		-
Material and Associations	Shape memory material	Glass transition temperature; Shape deformation temperature	-	Temporary shape fixing temperature	Shape recovery temperature		-
Experimental Measurement	-	-	Shape fixity: Shape fixity ratio	-	-	Shape recovery: Shape recovery ratio; Shape recovery time	Repeatability; Degradation

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