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# Temperature sensing and actuating capabilities of polymeric shape memory composite containing thermochromic particles

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# Abstract

**Purpose** – The study reported here intended to create and to study multifunctional shape memory polymer (SMP) composite having temperature sensing and actuating capabilities by embedding thermochromic particles within the polymer matrix.

**Design/methodology/approach** – The multifunctional materials were fabricated following a process consisting of blending (of the thermochromic particles and the SMP at various ratios), mixing, degasing, moulding and thermal curing, thus incorporating thermochromic particles within the polymer. The effect of the thermochromic particles on the thermomechanical properties and thermally responsive shape memory effect (SME) of the resulting multifunction SMP composites were characterised and interpreted.

**Findings** – It was found that exposure of the composites to temperatures above 70°C led to a pronounced, reproducibly reversible change of their colour that was recorded by the thermal and electrical actuation approaches. It was also found that the colour of the composites was independent of the mechanical state of the SMP. Such effects enabled monitoring of the onset of the set/release temperature of the SMP matrix. Furthermore, the combination of thermochromic additive and the SMP resulted in significantly improved thermomechanical strength, absorption of infrared radiation and temperature distribution of the SMP composites.

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**Research implications** – The temperature sensing and actuating capabilities of the polymeric shape memory composites developed through this study will help to extend the field of potential applications of such composites to fields including sensors, actuators, security labels and information dissemination, where colour indication is an advantageous feature.

**Originality/value** – The shape memory polymer (SMP) composites capable of temperature sensing and actuating are novel.

**Keywords** – Shape memory polymer, Thermochromic particles, Sensing capability, Actuating capability, Multifunctionalisation

Paper type Research paper

#### Introduction

Shape memory polymers (SMPs) have attracted extensive attention among smart materials and structures research community as an exciting class of stimulus responsive materials, yet having the ability to memorise a permanent shape [i.e. shape memory effect (SME)] (Mather et al., 2009; Xie, 2011; Sun et al., 2012). SMPs could be manipulated and "fixed" to a temporary shape under specific conditions of temperature and stress, and subsequently relax to the permanent state upon application of external stimuli, such as heat, light, magnetic field, or electrically resistive heating (Lendlein and Langer, 2002; Meng and Hu 2009; Liu et al., 2009; Lu and Gou 2012; Hu et al., 2012; Zhao et al., 2012; Lu et al., 2013; Lu et *al.*, 2014a). The mechanism behind shape memory capability lies in their molecular network structure, which contains at least two separate phases or domains (Rousseau 2008; Nguyen et al., 2008; Xie 2010; Huang et al., 2010). The phase or domain showing the highest thermal transition is responsible for the permanent shape. The other is the switching phase or domain that has, on the other hand, the ability to soften and is responsible for the temporary shape (Lendlein and Kelch 2002; Dietsch and Tong 2007; Lu and Huang 2013b; Lu and Huang 2013a; Lu et al., 2013; Sun et al., 2014; Lu and Du 2014). SMPs can retain two or sometimes three shapes, and cover a wide property ranging from stable to biodegradable, from soft to hard, and from elastic to rigid (Tobushi et al., 2001; Miaudet et al., 2007; Sun and Huang 2010). These unique characteristics enable the SMPs to be used in a myriad of fields from aerospace engineering to medical applications, since they can change shape and adopt properties in response to external stimuli (Maitland et al., 2002; Gall et al., 2004; Paik et al., 2006; Lan et al., 2009; Liu et al., 2009).

Much previous research work has focused on structure-property relationships of various SMP materials based on chemically or physically cross-linked polymer systems (Kim *et al.*, 1998; Gunes *et al.*, 2008; Ma *et al.*, 2013). Most of the current research in the field of SMPs is focused on the intrinsic function of the SME, functional composite, the exploitation of different stimuli as triggers and demonstration of potential application (Ma *et al.*, 2013; Bellin *et al.*, 2006; Tobushi *et al.*, 2008; Lu *et al.*, 2009; Lu *et al.*, 2010; Luo and Mather 2010; Lantada *et al.*, 2010; Le *et al.*, 2011; Lu *et al.*, 2011b; Lu *et al.*, 2011a; Fejos *et al.*, 2012; Lu 2012a; Lu 2012b; Fejos and Karger-Kocsis 2013; Lu *et al.*, 2013; Lu *et al.*, 2013c; Lu *et al.*, 2014b).

In this paper, we describe a novel SMP composite that incorporates thermochromic particles, which serve as an intrinsic sensor thus providing additional functionality. Thus, the composite contains chromogenic sensing colour-changing microcapsule particles that provide a low-cost, intrinsic and effective signal indicating the onset of set/release temperature. Thermochromism is the property of materials to change colour in response to a change in temperature. Thermochromic colour-changing microcapsules are based on mixtures of a leuco dye and, most often, an acid and a solvent for both the dye and the acid. At a lower temperature, all three ingredients exist in three separate solid phases. At an elevated temperature, all three ingredients melt allowing both the dye and the acid to be dissolved in the solvent causing the dye to react with the acid and subsequently changing from coloured to colourless. Such a mechanism dictates that the three ingredients need to be closely adjacent to each other in order to allow the colour change to occur reversibly and reproducibly as a result of temperature change. Consequently, the three ingredients are often necessarily encapsulated in the form of microcapsules to ensure that the ingredients are in the close vicinity of each other.

In this study, a novel SMP composite has been fabricated by employing thermochromic microcapsules to achieve temperature-sensing capability via visible colour change. It was found that exposure of the composites to temperatures above 70°C led to a pronounced, reproducibly reversible change of their colour that was induced by the thermal and electrical actuation means. It was also found that the colour of the composites was independent of the mechanical state of the SMP. Such effects enabled monitoring of the onset of the set/release temperature of the SMP matrix. Furthermore, the combination of thermochromic additive and the SMP resulted in significantly improved thermomechanical strength, absorption of infrared radiation and the temperature distribution of the SMP composites. As a result, SMP composites having thermal sensing and actuating capabilities, as well as being responsive to external thermal and electrical stimuli, have been achieved.

# Experimental

# Materials

### Thermochromic microcapsules

The thermochromic material employed was temperature-sensitive, colour-changing microcapsules in a powder form having grain diameters ranging from 2 to  $7\mu$ m (see Figure 1), supplied by New Prismatic Enterprise Co. Ltd. When exposed to temperatures above 70°C, the material goes from coloured (black) to colourless. It will return to its original colour as the temperature goes down. Therefore, these thermochromic microcapsule powders have a built-in ability to sense changes in temperature.

(Take in Figure 1)

# SMP matrix material

The epoxy SMP matrix used in the study reported here was an in-house fabricated twocomponent thermosetting resin.

# Methods of preparation

The colour-changing microcapsule powders were blended into the SMP resin at different weight fractions of 5wt.%, 10wt.%, 15wt.% and 20wt.%. The mixture was mechanically stirred at a speed of 600rad/min for 30min. The resulting mixture was then degassed in a vacuum oven to completely remove air bubbles. A resin transfer moulding process was used to fabricate the SMP composite samples under a constant pressure of 6bar. After the mould was filled, curing was done upon heating from room temperature to 100°C at a ramp of approximately 1°C/min and then kept for 5 hours before the temperature was ramped to 120°C at a heating rate of 20°C per 180 min. Finally, the temperature was ramped to 150°C at 30°C per 120 min to obtain the final SMP composite.

# Methods of characterisation

# Characterisation of the morphology and structure of colour-changing microcapsule in SMP

Scanning electron microscopy (SEM, VEGA3 TESCAN) was employed to characterise the morphology of colour-changing microcapsule particles in SMP composite.

# Characterisation of glass transition of SMP composite

 $T_g$  plays an essential role in influencing the shape recovery behaviour of the epoxy-based SMP. Therefore, it was necessary to characterise the effect of colour-changing microcapsule powder on the  $T_g$  prior to the investigation of the shape recovery behaviour of this epoxy-

based SMP. Differential scanning calorimetry (DSC 204F1, Netzsch, Germany) measurements were carried out in a nitrogen environment within a temperature range of 25 to 120°C at a constant heating rate of 10°C min<sup>-1</sup>.

#### Dynamic mechanical thermal analysis

Dynamic mechanical thermal analysis (DMA 242C, Netzsch, Germany) was conducted on the SMP composites to characterise the elastic modulus and tangent delta as a function of temperature. All tests were performed in three-point bending mode at a constant heating rate of 10°C·min<sup>-1</sup> and an oscillation frequency of 1Hz from 25 to 150°C. Specimens with dimensions of 9.0×1.90×1.85mm<sup>3</sup> were initially locked into a deformation of 0%.

### Characterisation of chemical structures and infrared absorbing efficiency

The chemical structures and infrared absorbing efficiency of coloured dye particles and SMP composite were determined by Fourier transform infrared (FTIR) spectroscopy (Nicolet AVATAR 360) in a transmittance mode.

#### Characterisation of the flexural strength of SMP composites

The flexural strength of SMP nanocomposites was measured via three-point bending using a Zwick/Roell servo-mechanical testing frame with a series of digital controllers. An Instron clip-on extensometer was used for strain measurement in three-point bending mode and a forced air convective environmental chamber was employed for elevated temperature tests. The static bending tests were performed at a loading speed of 2mm·min<sup>-1</sup> with a support span of 30mm, where the dimensions of the tested samples were 50×2.9×1.86mm<sup>3</sup>. The relationship between flexural strength and filler content in SMP composites was investigated at a temperature of 25°C.

#### Thermal/electrical actuation with temperature sensing

As a demonstration to show simultaneous optical response, the colour change of an epoxybased SMP composite filled with colour-changing microcapsule powder was stimulated by thermal and electrically resistive Joule heating, respectively, in the temperature from 25°C to above 70°C. Furthermore, an infrared video camera was used to record and monitor the temperature distribution in the SMP composite incorporated with colour-changing microcapsules and carbon fibre mat. Nine snap-shots of the tested SMP composite sample were presented to characterise the effect of colour-changing microcapsule powder on the thermally conductive property of SMP. A " $\Pi$ " shaped SMP composites sample with dimensions of 50×4×2mm<sup>3</sup> was bent into a "U" shape at 110°C. Images were taken with a digital camera at a constant frame rate of 30Hz, and with an appropriate visual range to detect the sample's curvature.

### **Results and discussion**

#### Morphology and structure of colour-changing microcapsule in SMP

Figures 2 (a) and (b) show the typical morphologies of colour-changing microcapsule particle at an accelerating voltage of 20.00keV. As can be seen, the colour-changing microcapsule particles had a diameter ranging from 1 to  $10\mu$ m, and were homogeneously dispersed into the polymer matrix. No large aggregates of microcapsule particles were found. It is expected that such microcapsules would endow the transparent matrix with a black colour, and would enable the SMP composite to change colour in response to external stimuli.

(Take in Figure 2)

#### Chemical structures and infrared absorbing efficiency

Figure 3 reveals the difference of infrared absorbing efficiency between coloured dye particles and SMP composite in the wavenumber range of 4000 to 400cm<sup>-1</sup>. It can be seen, from Figure 3, that SMP composite has lower absorption (less than 20%) over the whole wavenumber range, while coloured dye particles show consistently stronger (from 10% to 85%) absorption over the whole test wavenumber range. It was thus considered that the presence of the coloured dye particles within the SMP matrix would significantly increase the infrared absorbing capability of the resulting SMP composite. The reason is largely due to that most of the emitted energy is transmitted by SMP matrix, which is transparent, while most of the emitted light is absorbed by the coloured dye particle since it is black and opaque. Experimental result revealed that there was no chemical interaction between the coloured dye particle and the SMP matrix within the resulting composite. It was thus concluded that the chemical structures of SMP matrix and coloured dye particles in the composites were not influenced.

(Take in Figure 3)

#### **Glass transition temperature**

Since glass transition always occurs within a temperature range, the midpoint of the temperature range shown in the differential scanning curve was defined as the  $T_g$  of a tested SMP sample in this study. The change in heat flow as a function of temperature is presented in Figure 4.  $T_g$  was therefore determined as 48.83°C, 49.02°C and 50.43°C for the SMP

composites filled with 5 wt.%, 15 wt.% and 20 wt.% colour-changing microcapsule powder, respectively. It can be seen that the glass transition is shifted to a higher temperature range with an increase in the weight concentration of colour-changing microcapsule powder. On the other hand, in order to avoid the effect of water or moisture on the test results, the DSC measurements were conducted with two heating/cooling cycles. The  $T_g$  of the SMP composites determined by the second heating curve is presented in Figure 4 (b), while that determined by the first heating curve is revealed in Figure 4 (a). Experimental results indicated that the  $T_g$  was altered little as the weight concentration of colour-changing microcapsule powder increased from 5wt.%, 15wt.% to 20wt.%. It is expected that there is no strong chemical interaction between the polymer matrix and the microcapsule filler. That is to say that the effect of colour-changing microcapsule powder on the thermal property of the SMP matrix is not significant.

(Take in Figure 4)

#### **Flexural stress**

As shown in Figure 5, the flexural stress of the SMP composites with 5wt.%, 15wt.% and 20wt.% colour-changing microcapsule powder is 26.62, 26.32 and 26.39N, respectively. These results suggest that the mechanical strength of the composite specimens is not depressed with increasing colour-changing microcapsule powder content. The deformation of the composites might be contributed to the occurrence and propagation of cracks initiated by the debonding between matrix resin and filler, which is the major failure characteristic of the composite specimens. Also, the cracks might not propagate easily since the viscous matrix resin became hard to flow and the composite materials became brittle. With the content of the colour-changing microcapsule powder further increased, the maximum fracture strains of the composites did not always increase due to the contingency factors of cracks propagation.

(Take in Figure 5)

### Dynamic mechanical thermal analysis

The storage modulus and tan  $\delta$  data of the SMP composites were plotted as a function of temperature in Figure 6 (a). The data show that the storage modulus is 2237, 1984 and 2054MPa for the SMP composites at 25°C, respectively. Experimental results imply that the addition of colour-changing microcapsule powder has little negative effect on the storage modulus of composites over the whole temperature range. Note that the typical size of colour-changing microcapsule is similar to that of the macromolecular chains (He *et al.*,

2002). Therefore, the mobility of macromolecular chains would be restrained by the colourchanging microcapsule powder. Such experimental results were further supported by another experiment for elastic modulus, as shown in the Figure 6 (b), where the elastic modulus ( $E_o$ ) of SMP composites was determined by,

$$E_e = \sqrt{E_s^2 + E_l^2} \tag{1}$$

where,  $E_s$  is the storage modulus and  $E_l$  is the loss modulus.

(Take in Figure 6)

#### Thermal/electrical actuation with temperature sensing

Figures 7 (a) and (b) show the colour change of the tested sample in response to thermal and electrical stimuli with an increase in temperature. One sample is shown to give an example of thermally and electrically induced colour change. When the sample was heated to above 70°C, the sample regained its original shape after having been deformed into a temporary shape. Beside this characteristic SME in SMP, the composite could also alter its colour in response to the change of temperature. As a result, exposure of the SMP to heat leads to both shape recovery and a pronounced colour change by thermal and electrical actuation means.

(Take in Figure 7)

The synergistic effect of colour-changing microcapsule powder and carbon fibre mat on the electrically induced shape recovery is revealed in Figure 8. The SMP composites contained 5wt.% colour-changing microcapsule powder and 4.2wt.% carbon fibre mat, respectively. A constant 10V DC voltage was applied onto the SMP composite. It took 48s to complete the shape recovery. The composites showed very little recovery ratio during the first 6s, but then exhibited faster recovery behaviour until 42s. Finally, the SMP composite sample regained its original shape. And it was found that the temperature distribution was uniform, likely resulting from the synergistic effect of colour-changing microcapsule powder and carbon fibre mat.

(Take in Figure 8)

### Conclusions

In conclusion, a novel type of shape memory material with reversible temperature sensing capability has been developed by incorporating colour-changing microcapsule powder into SMP materials. A series of experiments had been conducted to study the effects of colourchanging microcapsule powder on the SMP composites. The temperature sensing capability of such composite material assemblies has been demonstrated by thermal and electrically resistive Joule heating. The electrically driven recovery behaviour was characterised at an electric voltage of 10 V. And temperature distribution of the SMP composite incorporated with colour-changing microcapsule powder and carbon fibre mat was monitored in the recovery process by electricity. The exposure of this microcapsules to temperatures above 70°C causes dissolution of the pH indicator dye and the acid in a solvent, resulted in colour changes. The colour was clearly visible and was independent of the mechanical state of the SMP and thus the effect allowed monitoring of the onset of the set/release temperature of the material. In view of the numerous fields that shape memory materials target, extensive applications of these chromogenic shape memory systems can be expected.

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# **Conflict of Interest**

The authors declare no conflict of interest.

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**Figure 1** The morphology of colour-changing microcapsule particles at a scale of (a)  $10\mu m$  and (b)  $20\mu m$ , respectively



**Figure 2** The morphology of colour-changing microcapsule particles in SMP matrix at a scale of (a) 2µm and (b) 20µm, respectively



**Figure 3** FTIR spectra of coloured dye particles and SMP composite in the absorbance mode



**Figure 4**  $T_g$  of the SMP composites determined by the DSC testing. (a) DSC scans of the first heating cycle; (b) DSC scans of the second heating cycle



**Figure 5** Stress-strain curves of SMP composites filled with various weight concentrations of colour-changing microcapsule powder in a triple-point bending mode



**Figure 6** (a) Storage modulus and tan  $\delta$  curves of the SMP nanocomposites at an oscillation frequency of 1 Hz; (b) Elastic modulus of the SMP nanocomposites as a function of temperature



**Figure 7** Thermochromic SMP composite induced by increasing temperature (a) Thermal actuation of SMP composite incorporated with colour-changing microcapsule powder; (b) Electrical actuation of SMP composite incorporated with colour-changing microcapsule powder and carbon fibre mat



**Figure 8** Snap shots of shape recovery and temperature distributions of SMP composite incorporated with colour-changing microcapsule powder and carbon fibre mat. The tested SMP composite was driven by loading an electric voltage of 10 V.

<b>0 s</b>	<mark>6 s</mark>	<mark>12 s</mark>
- 103 -	- <u>A</u>	<u>. 11</u>
<sup>*</sup> <mark>18 s</mark>	24 s	30 s
. 6 .	<u>.</u>	π
· · ·		
30 S	42 S	48 S
· 25 -		