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Adjustable and negative thermal expansion from multilayered systems

Joseph N. Grima[*, 1]**, Ludovica Oliveri**[2]**, Brian Ellul**[1]**, Ruben Gatt**[1]**, Daphne Attard**[1]**, Gianluca Cicala**[2]**, and Giuseppe Recca**[2]

[1] Faculty of Science, University of Malta, Msida MSD 2080, Malta

[2] Faculty of Engineering, Department of Physical and Chemical Methodologies for Engineering, University of Catania, Viale A. Doria n°6, 95125 Catania, Italy

[*] Corresponding author: e-mail joseph.grima@um.edu.mt, Phone: +356 2340 2274, Fax: +356 2540 1091

A construct in the form of a multi-layered system built from conventional materials having different mechanical and thermal properties is proposed. This construct can exhibit pre-determined values of thermal expansion which could also be negative. It is shown that for maximising negative thermal expansion (NTE) there should be a combination of thin layers of stiff materials having a high positive coefficient of thermal expansion (CTE) bonded with thicker layers of a soft material having low CTE values and, more importantly, having Poisson's ratio as high as possible.

The extent to which systems (materials or structures) change shape when subjected to changes in temperature is a subject of great practical importance and has been studied for many years. It is well known that miscalculations of the temperature effects in the design of objects which are subjected to significant temperature changes can lead to disastrous consequences. This led to various studies aimed at designing, analysing, manufacturing and/or testing of materials having very particular coefficients of thermal expansion [1–18] (including materials exhibiting negative coefficients of thermal expansion, i.e. materials which contract when heated [2–17]). Of particular interest are composites having pre-determined coefficients of thermal expansion which are already being used in some niche applications, although there is still an ongoing need to develop simple and non-expensive methods for achieving the same effect on any scale.

Here we propose and discuss a multi-layered system constructible at any length scale from conventional materials having different mechanical and thermal properties which may be combined to form systems which exhibit pre-determined values of thermal expansion which could also be negative. In particular we show that to optimise negative thermal expansion (NTE) characteristics, the systems must be constructed based on what is illustrated in

Fig. 1, i.e. by combining thin layers of stiff materials having a high positive coefficient of thermal expansion (CTE) characteristics bonded together through thicker layers of a soft material having low CTE values and, more importantly, having Poisson's ratio as high as possible, in analogy to work presented earlier [15–17].

To simplify our discussion we discuss first the optimal requirements for one of the most basic systems operating through the mechanism we are proposing which can be tailored to exhibit NTE, i.e. the system in Fig. 1(a). As illustrated in Figs. 1(a) and 2, this system may be described as a sandwich structure made from three layers of two different materials A and B having different thicknesses, thermal and mechanical properties which are perfectly bonded together.

Let us assume that this sandwich is in the form of a cuboid of overall dimensions $x \times y \times z$ at temperature T where the z dimension is given by $2z_A + z_B$, z_A and z_B being the thicknesses of layers A and B at temperature T, respectively. Let us also assume that (i) materials A and B are isotropic; (ii) that material A is much stiffer than material B, i.e. $E_A \gg E_B$, where E_A and E_B are the Young's moduli of materials A and B, respectively, and (iii) that necking effects may be ignored. Given these assumptions, if the system is subjected to a change in temperature ΔT,
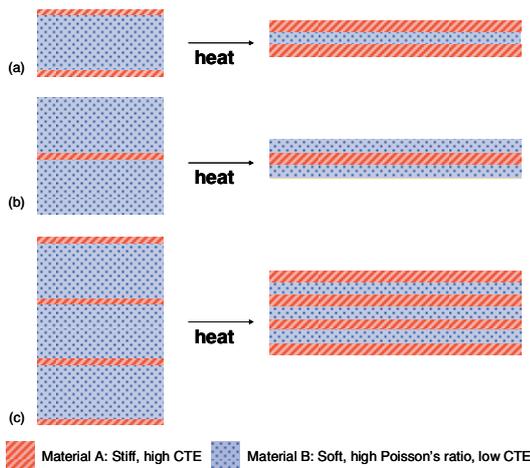
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Figure 1 (online colour at: www.pss-rapid.com) An illustration of the concept proposed here illustrated for systems having three or more layers. Note that this system is highly anisotropic and the observed NTE is only exhibited in the direction orthogonal to the layers (linear NTE) and not overall (volumetric NTE) as is observed in $Y_2Mo_3O_{12}$ or ZrW_2O_8.

then the x and y dimensions of the two layers must remain equal to each other even if materials A and B have different thermal expansion coefficients α_A and α_B, respectively, since we are assuming that the layers are perfectly bonded together. In fact, given that we are assuming that $E_A \gg E_B$, the thermal expansion in the x and y dimensions is dictated primarily by the expansion of material A and may be approximated by $\Delta x = x\alpha_A \Delta T$ and $\Delta y = y\alpha_A \Delta T$. In contrast to this, the thickness in the third dimension will change by an amount

$$\Delta z = 2 \Delta z_A + \Delta z_B , \tag{1}$$

where Δz_A is the change in thickness of each of the A layers and is simply given by

$$\Delta z_A = z_A \alpha_A \Delta T , \tag{2}$$

whilst Δz_B, the change in thickness of the B layer, is caused by both the normal thermal expansion effect and also by deformations in material B so as to accommodate the mechanical constraint that the x and y dimensions remain the same as those of material A to which material B is perfectly bonded to. Thus, since material B is isotropic,
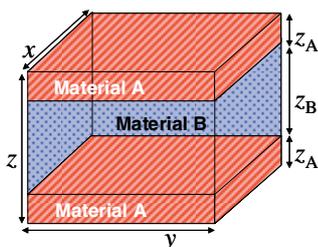

Figure 2 (online colour at: www.pss-rapid.com) Dimensions of the basic system discussed here.

Δz_B is given by [19]

$$\Delta z_B = z_B \left[\alpha_B \Delta T - \frac{2\nu_B \varepsilon_{AB}}{1 - \nu_B} \right], \tag{3}$$

where ν_B is the Poisson's ratio of material B and ε_{AB} is the strain caused by material A on material B as a consequence of materials A and B having different thermal expansion coefficients which is given by

$$\varepsilon_{AB} = (\alpha_A - \alpha_B) \Delta T. \tag{4}$$

By combining Eqs. (1)−(4), it becomes clear that

$$\Delta z = \left[2z_A \alpha_A + z_B \alpha_B - 2z_B \nu_B \frac{\alpha_A - \alpha_B}{1 - \nu_B} \right] \Delta T , \tag{5}$$

which will be negative for a positive ΔT (heating) if

$$2z_B \nu_B \frac{\alpha_A - \alpha_B}{1 - \nu_B} > 2z_A \alpha_A + z_B \alpha_B . \tag{6}$$

For example, if our system is heated and material A expands more than material B, then provided that $\nu_B > 0$, the 'mechanical' term $-2z_B \nu_B (\alpha_A - \alpha_B)(1 - \nu_B)^{-1} \Delta T$ will have a negative magnitude, i.e. the in-plane expansion of the A layers causes a shrinkage in the out-of-plane (z) dimension of the B layer. If this shrinkage is large enough to nullify the 'thermal' terms $[2z_A \alpha_A + z_B \alpha_B] \Delta T$, there will be an overall contraction in thickness (z) dimension when the system is heated, i.e. negative thermal expansion.

From these equations, it is clear that to magnify the NTE, in addition to having material B being much softer than material A, one should also have material B with as high a positive Poisson's ratio as possible (i.e. material B must be a rubber-type material). In fact, it is this Poisson's/rubber effect which is the essential feature for this mechanism to operate and generate NTE. Also, the CTE of material A must be as large as possible relative to that of material B so as to set the Poisson's effect in operation. However, so as to ensure that expansion in A caused by its high CTE does not nullify the contraction in B caused by the Poisson's effect, the A layers should be as thin as possible when compared to the B layer. In other words, as stated above, for maximum NTE, one should combine thin layers of stiff materials having high positive CTE characteristics bonded together through thicker layers of a soft material having low CTE values and more importantly, having Poisson's ratio as high as possible. Note that as illustrated in Fig. 1, similar properties will be obtained by systems having different numbers of layers, etc. (provided that care is taken so that their construction avoids bending caused by the well known bi-material strips effect).

To obtain further evidence that what is proposed here can indeed result in negative thermal expansion in the out-of-plane direction, we used the Finite Elements (FE)
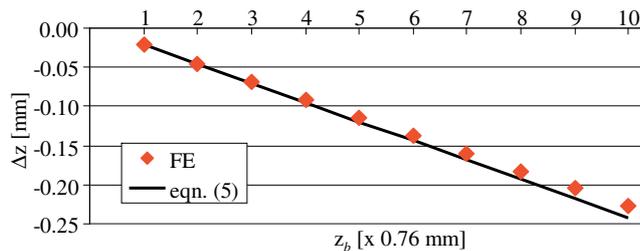
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Figure 3 (online colour at: www.pss-rapid.com) Change in thickness as predicted by the analytical expression in Eq. (5) and by the FE simulations as a function of the thickness of layers of the soft material when the system is subjected to a change in temperature of 100 K. Note that these systems all exhibit NTE the magnitude of which may be controlled by the thickness z_b.

software package ANSYS Academic Research V.12.0 to construct a multilayered structure consisting of alternating layers of two elastic materials A and B which were perfectly bonded together and meshed using the 3-D, 20-node, coupled-field SOLID226 element. These simulations confirmed that it is indeed possible for this mechanism to operate. For example, when we performed simulations varying z_B having $x = y = 200$ mm, $z_A = 0.076$ mm, $E_A = 0.552$ GPa, $E_B = 0.000517$ GPa, $\alpha_A = 1.9 \times 10^{-4}$ K^{-1} and $\alpha_B = 1.90 \times 10^{-5}$ K^{-1}, $\nu_A = 0.333$, $\nu_B = 0.499$ we found that NTE would be exhibited, this being in close conformity with what was predicted by Eq. (5) (see Fig. 3). Note that material A has the properties of a typical polyvinylidene chloride and material B has the properties of a typical silicone rubber [20]. Here it should be emphasised that the simulations presented here are only meant to act as a 'proof of concept' and more detailed studies need to be carried out before choosing the materials (e.g. the material used in the simulations for the layer A has the limitation of being brittle, has low flex resistance and flakes off easily). One should also be careful to use appropriate ratios for x/z and y/z to ensure mechanical stability as otherwise, one may deviate from the requirement that the geometry of the layers is flat.

Before we conclude it is important to highlight that the mechanism proposed here is not only of use to generate NTE but also as a mechanism of producing systems with pre-desired mechanical properties. In fact, it should be emphasised that the exact magnitude of the thermal expansion coefficient in the thickness dimension may be adjusted not only through careful choice of materials A and B, but also through control of the relative thickness of the layers. It should also be noted that what is proposed here is a scale independent mechanism, i.e., it could theoretically be possible to implement this effect at any scale of structure provided the right components can be made available, as is normally the case with models for negative Poisson's ratio [8] or negative compressibility [21].

We hope that given the simplicity of the proposed system and its adjustability, the proposed construct or variations of it will be manufactured and find use in practical applications.

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