Composites with needle-like inclusions exhibiting negative thermal expansion: A preliminary investigation

Joseph N. Grima a,⁎, Brian Ellul a, Daphne Attarda, Ruben Gatta a, Michael Attard b

⁎ Corresponding author.
E-mail address: joseph.grima@um.edu.mt (J.N. Grima).

1. Introduction

It is a well known fact that most materials which we encounter in everyday life expand on heating and contract when cooled, a property which may be explained by looking at interatomic distances [1–2]. This subject of thermal expansion is one of great practical importance and has been studied for many years since miscalculations of the temperature effects may lead to disastrous consequences.

Expansion in a material may be one of two forms: isotropic or anisotropic. In isotropic expansion, the material expands by the same extent in any direction (isotropically) upon heating whilst if the extent of expansion is dependent on the particular direction where the measurement is taken, then the expansion is referred to as anisotropic expansion. To quantify thermal expansion in some particular direction one may make use of the linear coefficient of thermal expansion, henceforth referred to as CTE which may be defined as \( \varepsilon \) which relates the resultant strain \( \varepsilon \) as a result of a change in temperature of \( dT \) through \( \varepsilon = \alpha T \).

As noted above, in most cases, materials expand when heated, i.e., \( \varepsilon \) is positive for a positive \( dT \) (i.e. as \( T \) increases) with the result that the CTE is positive. However, it should be noted that materials which defy common expectation and contract when heated do exist [3–19]. It should also be noted that for anisotropic systems, different values for the CTE will exist, depending on the direction of measurement and it is possible that a system exhibits negative thermal expansion (NTE) in some but not all directions.

Over the years, there have been various studies aimed at designing, analysing, manufacturing and/or testing of materials and structures having very particular CTE values, in particular, studies looking at systems, including composites exhibiting NTE [3–21]. Some of these studies have considered the possibility of generating very low or negative thermal expansion in structures constructed from conventional materials. These systems, which are constructible at any lengthscale, include those proposed by Clegg et al. [17–20] and Lakes [21] and have the advantage that they are relatively easy to construct at reasonable costs and thus have excellent potential for commercialisation. Alderson et al. [22] have also proposed a method for achieving these effects using auxetic materials (i.e. materials exhibiting negative Poisson’s ratio). Normally, such systems also offer the added advantage that they can be tailor-made to exhibit any desired thermal expansion properties (positive, negative or near zero) through careful choice of the geometric parameters and/or materials used in the construction.

Furthermore, such systems are also of interest in view of the fact that they can be engineered to exhibit other interesting macroscopic properties, e.g. negative Poisson’s ratio [23–25] with the result that one achieves multifunctional systems with obvious added value.

In this paper we propose and discuss a novel system constructible at any lengthscale including the microscale from conventional components having different mechanical and thermal properties which may be combined to form composite systems which exhibit any desired CTE values, in particular negative ones (NTE).
2. The concept

For simplicity, to illustrate how systems operating through the proposed mechanism can be designed to exhibit NTE, we shall first consider a simple and basic system and predict the optimal requirements for it to exhibit this effect. The system which is shown in Fig. 1 consists of a cylindrical rod of length \( l \) and radius \( r \) made of a material B which is embedded inside another cylindrical shell of thickness \( t \) made from a material A having different thermal and mechanical properties with all dimensions being measured at a reference temperature \( T \).

Assuming that materials A and B are perfectly bound to each other at the interface and that they are isotropic with respect to their thermal and mechanical properties, when the system is subjected to a change in temperature \( \Delta T \), both materials expand or contract in volume accordingly, each at a different rate. Since the materials are bound to each other, they cannot expand freely and as a result, each of them exerts forces on the other. In particular, the material with the higher CTE exerts a tensile force in the longitudinal direction on the material having the lower CTE and conversely the latter exerts a compressive force on the former resulting in a mechanical strain. This strain in turn gives rise to an additional strain in the radial direction, the magnitude of which is dependent on the Poisson’s ratio (henceforth referred to as Poisson’s effect). This causes a contraction (or an expansion) in this direction and unless the Poisson’s ratio of the materials is zero, this Poisson’s effect may have significant contributions to the overall radial strain and therefore should not be neglected.

In addition, to further simplify our analysis, we shall also assume that:

1. The inclusion has a needle shape, i.e. the length is much larger compared to its thickness so that changes in dimension arising from the Poisson’s effect due to expansion in the radial direction can be ignored since they are negligible when compared to that resulting from a longitudinal expansion;
2. Young’s modulus of the outer cylinder (i.e. the matrix) is much smaller than that of the inclusion and thus additional mechanical strains in the radial direction arising due to mismatch of the CTEs can be neglected;
3. No necking effects occur during deformation.

Thus, in the analytical model it is being assumed that the strain at the interface is only dependent on the thermal expansion and Poisson’s effect of material B, i.e. the inner circumference of material A expands accordingly to accommodate the changes in the outer circumference of material B thereby satisfying the boundary condition of the interface (and therefore eliminating gaps or unrealistic overlaps).

In this way, it is possible to derive a simple expression for the radial strain by taking into consideration only the radial thermal strain and the additional radial mechanical strain arising from Poisson’s effect due to expansion in the longitudinal direction. Taking first the longitudinal strain \( \varepsilon_l \) into consideration, it can be quantified through the following equation:

\[
\varepsilon_l = \alpha_a \Delta T + \frac{F_A}{\alpha_a E_A} = \alpha_a \Delta T + \frac{F_B}{\alpha_b E_B}
\]

where \( \alpha \) is the CTE of the material, \( E \) is Young’s modulus, \( a \) is area, \( F \) the force exerted by the other material and the subscript refers to the material (materials A and B). At equilibrium, \( F_B = -F_A = F \), so that solving for \( F \), the following expression is obtained:

\[
F = -\frac{\alpha_a E_A E_B (\alpha_a - \alpha_b)}{\alpha_a E_B + \alpha_b E_A} dT
\]

The mechanical strain on material A arising from this force is therefore:

\[
\varepsilon_A = -\frac{F}{\alpha_a E_A} = -\frac{\alpha_B E_B (\alpha_a - \alpha_b)}{\alpha_a E_B + \alpha_b E_A} dT
\]

so that the induced radial strain arising from Poisson’s effect can be given by:

\[
\varepsilon_A = \frac{\nu_a \alpha_a E_a (\alpha_a - \alpha_b)}{\alpha_a E_B + \alpha_b E_A} dT
\]

and similarly for material B:

\[
\varepsilon_B = -\frac{\nu_b \alpha_b E_B (\alpha_a - \alpha_b)}{\alpha_a E_B + \alpha_b E_A} dT
\]

Therefore, the total strain \( \varepsilon_t \) in the radial direction, taking into account the thermal expansion of both materials can be given by:

\[
\varepsilon_t = \frac{2\nu_a \alpha_a E_a (\alpha_a - \alpha_b)}{E_A} \frac{(t + r)^2 - t^2}{r^2} = \frac{[\nu_a \alpha_a E_a (\alpha_a - \alpha_b) / (t + r)]^2}{(t + r)}
\]

Thus defining \( \alpha_a \) as the thermal expansion coefficient in the radial direction, we note that:

\[
\alpha_a = \frac{\nu_a \alpha_a E_a (\alpha_a - \alpha_b)}{E_A} \left[ \frac{1}{(t + r)^2} - \frac{1}{r^2} \right]
\]

Since \( \nu_a, E_a, \) and \( r \) are all positive quantities, it follows that the \( \alpha_a \) and the radial strain is negative if:

\[
\frac{\nu_a \alpha_a E_a (\alpha_a - \alpha_b)}{E_A} \left[ \frac{1}{(t + r)^2} - \frac{1}{r^2} \right] < 0
\]

This inequality suggests that for a system with \( E_A < E_B \) as in our earlier assumptions the NTE effect can be enhanced by decreasing \( \alpha_a / \alpha_b \). This ensures that thermal expansion in the radial direction of the thicker material is minimal so that the main contribution comes from the Poisson’s effect which can be enhanced by using a soft matrix with a high Poisson’s ratio and a much stiffer inclusion with a low or negative Poisson’s ratio. In this way, the matrix has very little effect on the inclusion, i.e. the degree by which material A is stretched is much higher than that by which B is compressed. This in turn means that the decrease in thickness of A is much larger than the increase in thickness of B (if \( \nu_B > 0 \)) making NTE possible. If the inclusion is auxetic, the change in its radial dimension due to compression will further contribute to a more negative thermal expansion (and also have the additional advantage of a larger pull-out resistance [26,27]). For example for a system with \( r = 0.5 \text{ mm}, t = 50 \text{ mm}, E_A = 0.01 \text{ GPa}, E_B = 200 \text{ GPa}, \nu_A = 0.49, \nu_B = 0.3, \alpha_A = 15 \times 10^{-6} \text{ K}^{-1} \) and \( \alpha_B = 324 \times 10^{-6} \text{ K}^{-1} \) the strain

![Fig. 1](image-url)
in the radial dimension for a 100 K increase in temperature is predicted to be $-8.09 \times 10^{-3}$ and $-8.15 \times 10^{-3}$ if an auxetic inclusion ($v_A = -0.3$) is used. It is also interesting to note that zero thermal expansion is also predicted if $v_A = 0.091$.

3. Simulations

In an attempt to obtain further evidence that what is proposed here can indeed result in a method for controlling the thermal expansion in the radial direction which could also lead to NTE, we used the finite elements (FE) software package ANSYS Academic Research V. 12.0 to construct this most basic system. In the simulation, we used two elastic materials A and B which were perfectly bonded together and meshed using the 2-D, 8-node, coupled-field PLANE223 plane element with axisymmetric behaviour where the $z$-axis is the axisymmetric axis while symmetric boundary conditions were applied along the $r$-axis (Fig. 2). The nodal degrees of freedom (DOF) are translations in the $z$- and $r$-directions and temperature where Table 1 lists the nodal constraints. As regards loading, a uniform temperature rise of 100 K was applied on all the nodes.

Two sets of simulations were performed. In the first, this system was solved linearly for the geometry corresponding to $l = 500$ mm, $t = 50$ mm and $r = 0.5$ mm. The material was modelled as perfectly elastic and isotropic with $E_A = 0.01$ GPa, $E_B = 200$ GPa, $v_A = 0.3$, $\alpha_A = 15 \times 10^{-6}$ K$^{-1}$ and $\alpha_B = 324 \times 10^{-6}$ K$^{-1}$ while the Poisson’s ratio of the matrix ($v_A$) was varied from $-1.0 < v_A < 0.5$ subjected to a temperature rise of 100 K. In the second set, $v_A$ was set at 0.49 whilst the thickness $t$ was allowed to vary from $100 > t > 0$ with all the other parameters set as before. It should be emphasised that although the simulations performed here are in the mm range, the effect is scale independent and can also be exhibited at smaller or larger scales.

As post-processing, for both sets of simulations, the radial strain $\varepsilon_r$ was recorded as $v_A$ or $t$ were varied from which we calculated the thermal expansion coefficient $\alpha_r$ in the radial direction as $\alpha_r = \varepsilon_r / \Delta T$. The results of these two sets of simulations are shown in Figs. 3 and 4 which compare the FE results with those predicted by the analytical model.

These results clearly show that in accordance to what is predicted by the analytical model, the systems presented here can exhibit a wide range of thermal expansion properties which can be varied by varying either the types of materials used (something which may be limited by the availability of materials having the required properties) or the geometric parameters, such as the thickness of the outer material. In particular, the simulations confirm that in the radial direction, the system can indeed exhibit not only negative linear thermal expansion, a property which is exhibited best when the outer material (the matrix) has high positive Poisson’s ratios but also zero linear thermal expansion. This property is highly desirable in applications where a system has to maintain its structural integrity while subjected to significant changes.

Table 1

<table>
<thead>
<tr>
<th>Line</th>
<th>DOF constraints</th>
<th>$U_r$</th>
<th>$U_z$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td></td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td></td>
<td></td>
<td>0</td>
</tr>
<tr>
<td>3</td>
<td>Coupled$^a$</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>4</td>
<td></td>
<td></td>
<td>Coupled$^a$</td>
</tr>
<tr>
<td>5</td>
<td></td>
<td></td>
<td>Coupled$^a$</td>
</tr>
</tbody>
</table>

$^a$ The DOF is equal for all the nodes.

Fig. 2. (a) Line numbers showing the respective nodes. (b) 3/4 of the composite in 3D.

Fig. 3. Plot of the radial CTE $\alpha_r$ versus Poisson’s ratio of the matrix $v_A$ for both the FE and analytical results.

Fig. 4. Plot of the radial CTE $\alpha_r$ versus the matrix thickness $t$ for both the FE and analytical results.
an increase in the thickness

tems which could be modified to exhibit these effects include

ded inside the matrix in a random but aligned manner. Other sys-

the highly expanding and hard needle shaped inclusions are moul-

Fig. 5 which shows a cross-section of a possible composite where

complex system based on the same mechanism is illustrated in

thermal expansion of composites (which may not necessarily have

presented here may be used in the design and manufacture of real

material, which thinning has not been accounted for in the analyt-

in the circumference will cause an additional thinning of the outer

thickness $r$ which optimises the magnitude of NTE.

The main discrepancies between the analytical and FE models

seem to occur at low values of $r$ and these are evident in the plots

of $\alpha_r$ against the thickness $t$ of the surrounding material where at

low values of $t$, the analytical model underestimates the extent

of NTE. The most likely reason for this underestimation is due to

the fact that at small values of $t$, Poisson’s effects as a result of an

increase in the thickness $r$ (and hence the circumference) of the

needle shaped inclusion are not insignificant. Such increase in the circumference will cause an additional thinning of the outer

material, which thinning has not been accounted for in the analytical

model. All this suggests that although there is a need to fine-
tune the analytical model so as to take into account this effect

(something which unfortunately will decrease the simplicity of the analytical model), the analytical model as presented here can

already provide a reasonable estimate of the thermal expansion properties of such systems.

Before we conclude, it is important to note that the concepts

presented here may be used in the design and manufacture of real

composites exhibiting such NTE properties or for controlling the

thermal expansion of composites (which may not necessarily have
to exhibit NTE). For example, an easily constructible but more

complex system based on the same mechanism is illustrated in

Fig. 5 which shows a cross-section of a possible composite where

the highly expanding and hard needle shaped inclusions are moul-
ded inside the matrix in a random but aligned manner. Other sys-
tems which could be modified to exhibit these effects include

fibre-reinforced composites which if constructed using the right

component materials would also be able to exhibit these unusual properties. In such systems, one would assume that the CTE will

also be affected by other factors such as the degree of perfection

in the alignment of the needle shaped inclusions and the packing,

but the principles which may lead to the unusual thermal effects

presented in this paper, including the ability to exhibit NTE, remain

the same. It is also important to note that although materials

which have the thermal and mechanical properties required to

achieve this NTE effect in a very significant manner are not cur-
rently in abundance, the effect may be obtained by building micro

mechanical systems (e.g. using syringe-like structures filled with a

highly expanding fluid embedded within a soft material) or having

the components themselves being man-made composites which

are tailor-made to have the required properties.

4. Conclusion

This work presented a concept for controlling the thermal

expansion through the introduction of highly expanding and hard

needle shaped inclusions into a soft matrix. In particular, we de-

rived an analytical model for a simple cylindrical system contain-

ing a needle-like inclusion which can be tailor-made to exhibit

any pre-desired positive or negative thermal expansion coefficient

in radial directions. In fact, the derived expression for the radial

strain showed that by the correct combination of the materials’

thermal and mechanical properties, NTE and zero thermal expan-

sion in the radial direction are possible, with the NTE effect being

further enhanced by the use of auxetic inclusions. The validity of

the model under the made assumptions has also been verified

using finite element analysis.

We hope that the models presented and discussed here will

encourage experimentalists to manufacture and commercialize

new materials which can be tailor-made to have properties to fit

particular practical applications based on the concepts presented

here. Given the simplicity of our systems and their adjustability,

we envisage materials based on what is proposed should find

extensive use in many practical applications where negative or

zero thermal expansion is required, or where the thermal expan-

sion needs to be controlled in a cost-effective manner.

Acknowledgments

The financial support of the Malta Council for Science and Tech-

ology (MCST) through their 2006 R&I programme and of the Mal-
ta Government Scholarship Scheme (Grant Number ME 367/07/17

awarded to Daphne Attard) is gratefully acknowledged.
References