

Dynamic Mismatch Between Bonded Dissimilar Materials

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In the bonding of dissimilar materials, the coefficient of thermal expansion (CTE) relates to only the static or thermal equilibrium case, and does not represent most actual conditions (i.e., the service and processing temperatures are usually changing rather than fixed). This article outlines an approach that computes the effective, or dynamic, CTE mismatch. This dynamic mismatch varies with the bonded material shapes and sizes, surface characteristics, and heating or cooling conditions and times and may be several times greater than the corresponding static CTE mismatch. Unrelieved, the computed transient or dynamic thermal-strain mismatch may exceed the yield point of the metal, while the transient or dynamic mismatch stress often exceeds the flexural or compressive strength of the ceramic. Understanding transient mismatch phenomena has led to new, unmatched metal-ceramic joints that withstand repeated, rapid thermal shocks and subsequent severe mechanical shocks. The final forced fractures occur outside the bonded regions, indicating defect-free joints.

INTRODUCTION

The joining of dissimilar materials has been used widely to form complex structures with simple standard shapes, often combining the advantages of both materials. Metal-ceramic joining, first developed during World War II, has been used extensively in the electronics industry.¹ However, reliably strong and temperature-resistant joints are still not available worldwide.^{2,3}

Thermally generated stresses and strains are critical factors in dissimilar materials joints. In metal-ceramic joints, differences in coefficients of thermal expansion (CTE) between the metal and the ceramic produce thermal-mismatch stresses and strains. These stresses and strains determine the failure probability of the joint.

According to McDermid et al.^{4,5} and Mehan et al.,^{6,7} a large mismatch in CTE (f) between the metal and ceramic (namely, $\Delta f = 10^{-5} \text{ }^\circ\text{C}^{-1}$), results in failure of the joint at the metal-ceramic interface. These failures are caused by the high stresses generated during cooling from the bonding temperature.

However, such CTE mismatches relate to only the static, or thermal equilibrium, case. They do not truly represent dynamic or transient conditions when the joint is being heated or cooled. Yet

such transient or dynamic conditions always exist during the manufacture or service of the joint.

An important problem with common joining processes is the understanding and control, over a period of time, of dynamic mismatches in temperatures, CTEs, and thermal strain and stress profiles and gradients in the joint region. This article describes such dynamic mismatch phenomena and proposes special, laterally graded composition and/or physical-property profiles of the joint region.

As will be shown, the computed dynamic mismatches in expansion strain may, if unrelieved, exceed the yield points of even the metallic joining materials. The dynamic mismatch stresses also often exceed the flexural or even comprehensive strengths of the ceramic materials. What fails most metal-ceramic joints, or causes most ceramic coatings to crack, peel, flake, or spall, is, therefore, the dynamic, rather than the static, thermal-expansion mismatch. Through this dynamic mismatch approach, one can determine the location, magnitude, and occurrence time of the maximum dynamic-mismatch stresses and strains. One can also devise simple procedures to estimate the joint strength and to reduce these critical stresses and strains on the relatively weak ceramic.

STATIC MISMATCHES

Thermal Strains and Expansion Coefficients

The linear CTE (f) is defined as the thermal expansion per unit length per degree Celsius. It refers to only the static or thermal equilibrium case. For a given material, this coefficient is a constant for a temperature range of interest. Within this range, therefore, the CTE does not depend on the initial and final temperatures, specimen geometries, sizes, diffusivities, surface characteristics, and heating or cooling rates and other conditions. Each material has its own single, unique, static CTE for a given temperature range.

During a cool-down process, the static thermal shrinkage (or negative expansion) strain (e) for a given material is, by definition, the static CTE (f) multiplied by the cooling temperature range (Δu):

$$e = f\Delta u$$

For a steel rod with a CTE of f_s , cooling through a temperature range of Δu_s , the strain is e_s . Similarly, for a rod of Macor (Corning's machinable glass-ceramic)⁸ with a CTE of f_m , cooling through a temperature range of Δu_m , the shrinkage strain is e_m .

Macor is machinable on conventional metalworking machines. The key to its machinability is its two-phase microstructure of randomly oriented mica microcrystals in a glass matrix. During machining, cracks are propagated in the direction of the applied force. These cracks are deflected by the microcrystals to the surface. According to Corning, Macor has a CTE of about $9.35 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$, and can be sealed to 52% nickel alloys, chromium-iron stainless steel, platinum, and other materials by using a special glass frit from Corning.⁸

In the static case, the materials of a steel and Macor joint are always in constant thermal equilibrium. That is, $u_m = u_s$ for all times. At the beginning of cooling (time $t = 0$), both materials are at the same brazing temperature of u_0 . At any time during the cooling after the joining by, for example, brazing, the cooling temperature ranges for steel and Macor are always the same in the static case. Thus:

$$\Delta u_m = u_0 - u_m = u_0 - u_s = \Delta u_s = \Delta u$$

Usually, one assumes that both the steel and Macor are homogeneous, defect-free, and perfectly joined together. In addition, the static thermal-mismatch strain is not relieved, modified, or adjusted in any way. In this simple case, this static mismatch strain between steel and Macor is:

$$\Delta e = e_s - e_m = (f_s - f_m)\Delta u = \text{constant} \cdot \Delta u$$

DYNAMIC MISMATCHES

Temperatures

Dynamic mismatches result from the fact that metals and ceramics have widely different thermal conductivities.⁹ During heating of a metal-ceramic joint, the temperature of the ceramic lags behind that of the metal, often markedly so; during cooling, the opposite is true. This produces different temperature profiles in the metal and ceramic at a particular

time on either heating or cooling. Dynamic mismatches in temperatures, effective CTE, thermal strains (i.e., expansions on heating or shrinkages on cooling), and thermal stresses (strains multiplied by Young's modulus) then result.

In reality, after the actual brazing to produce the metal-ceramic joint, only at the beginning of the cooling ($t = 0$) are the two materials at the same brazing temperature of u_b . At any subsequent cooling time after the brazing ($t > 0$), the ceramic is at a higher temperature than the metal. There is, therefore, a nonzero dynamic temperature differential (Δu).

Consider the special case of a long metal rod joined end-to-end to a long ceramic rod of the same diameter, $D = 2r$. The metal is SAE 1010 carbon steel, while the ceramic is Macor. The joint is brazed at 950°C and is, for a worst-case condition, suddenly air quenched in a 20°C environment.¹⁰

The following assumptions are made in the computation of the dynamic or transient mismatch stresses and strains:

- The steel and Macor cylinders are infinitely long and have only separate and independent, radial heat conduction. There is no axial heat flow from one material to the other.
- Biaxial or triaxial stresses and strains are not considered.
- Only elastic strains and stresses are treated.
- Strain and stress relief through plastic deformation or other mechanisms is ignored.
- Both materials are homogeneous and free of any defects such as pores, voids, microcracks, inclusions, or second phases.
- The two materials do not have elemental interdiffusions, undergo phase changes, or otherwise suffer modifications in physical and chemical properties.
- There are no intervening bonding material layers of different chemical, thermal, and mechanical properties than those of the steel and Macor.

In a metal-to-metal joint, the assumption of pure radial-heat conduction in the cylinders is obviously not valid. However, if one or both cylinders are made of thermally insulating materials such as ceramics, this assumption is a good start. Mainly because of the radial-heat-conduction assumption, the temperature in each cylinder is uniform at a given radius. To provide a more detailed analysis, if any one or more of these assumptions were not made, would be extremely difficult. At this time, the comparative errors from the above assumptions are not known, even qualitatively. Hence, any expensive, time-consuming analysis is not justified.

The Fourier equation for independent radial-heat conduction in long metal and

ceramic cylinders is well known. The solution of the cylindrical heat-conduction problem consists of an infinite series. Each term of the series is a product of a Bessel's function and an exponential function, as given in various textbooks on heat conduction (see, for example, Reference 11). One can thus determine the temperature profiles at different locations (i.e., radial positions $[r]$ in a cylindrical end-to-end joint) at various times. At the critical time (t_c) the critical profile of the temperature differentials and the associated, maximum transient dynamic thermal-mismatch stresses and strains obtain.

Table I gives the step-by-step temperature changes of a 5.08-cm diameter, cylindrical end-to-end steel-Macor joint for the temperatures of steel and Macor, respectively, at the cylindrical axes ($r = 0$) for $t = 0$ to 41,800 s after cooling from the brazing to near-room temperature.¹⁰ The computer simulation results in Table I also give the maximum temperature differential between steel and Macor at the axial center point (i.e., $\Delta u = u_m - u_s$), at different cooling times.

Immediately after brazing ($t = 0$), this differential is zero because both the steel and Macor are at the same brazing temperature of 950°C. Subsequently, the faster cooling of the steel increases this

differential, reaching a maximum of 775°C at $t = 1,000$ s. After both rods are significantly cooled, the temperature differential decreases. Beyond 29,900 s (8.3 h), for example, both rods are near room temperature at 20°C. The maximum temperature mismatch or differential of 775°C at $t = 1,000$ s produces the maximum or critical dynamic mismatch stress and strain, as shown in Figure 1.

By comparison, for a 2.54-cm-diameter steel-Macor joint, the maximum temperature mismatch of 727°C at the axial center occurs sooner (i.e., at 440 s) after cooling.

Thermal Strains and Expansion Coefficients

The dynamic thermal expansion coefficients (f^*) and the resultant dynamic thermal-mismatch strains (Δe^*) and stresses (s^*) strongly depend on the joint materials, geometries, sizes, physical and surface properties, and heating or cooling conditions. Starting with zero strain on cooling from the brazing temperature of 950°C, the dynamic strain in the steel rod is: $e_s^* = f_s \Delta u_s$, where $\Delta u_s = 950 - u_s$, while in the Macor rod, $e_m^* = f_m \Delta u_m$, where $\Delta u_m = 950 - u_m$. Also, $u_s \neq u_m$ and $\Delta u_s \neq \Delta u_m$. The difference in dynamic mismatch strain is:

$$\Delta e^* = f_s \Delta u_s - f_m \Delta u_m$$

Under the pure cylindrical heat-conduction model, the computed dynamic or transient mismatch strain reaches a maximum of about 0.0123 at $t = 1,000$ s, as shown in Figure 2. Such a high strain, if not relieved or reduced, would exceed the yield point of the steel, which is joined to the even more rigid Macor.

The dynamic (or effective) CTE mismatch (Δf^*) can be computed by dividing the dynamic mismatch strain (Δe^*) by the average cooling temperature range [i.e., $\Delta u_{av} = 950 - (u_s + u_m)/2$]. For the 5.08 cm, steel-Macor end-to-end joint cooling from 950°C to 20°C, this dynamic CTE mismatch depends greatly on the cooling time and conditions. A maximum computed dynamic CTE mismatch of about $29.6 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$ occurs at a cooling time of 90 s, as shown in Figure 3. Such a high dynamic CTE mismatch is intolerable, according to McDermid and Mehan.⁴⁻⁷

Figure 4 shows that for the 5.08 cm steel-Macor rod joint cooling from 950°C to 20°C, the computed effective or dynamic CTE mismatch, $\Delta f^* = (f_s^* - f_m^*)_{av}$, is more than two to five times greater than the corresponding mismatches for the static or equilibrium case, for cooling times of 10 s to 6,000 s. This ratio of dynamic CTE to the static CTE reaches a maximum of 5.3 at $t = 75$ s.

Thermal Mismatch Stresses

To compute the dynamic mismatch stresses, one may further neglect the pres-

Table I. Computed Dynamic Temperature Mismatches in a Steel-Macor Joint

t (s)	u_m (°C)	u_s (°C)	Δu (°C)
0.0	950	950	0
6.0	950	947	3
12.0	949	935	14
23.9	949	901	48
35.8	949	867	82
47.8	948	835	113
59.8	948	804	144
89.6	948	731	217
119	947	665	282
239	935	456	478
358	918	316	503
478	901	220	681
598	884	155	729
717	868	112	756
836	851	82	769
956	835	62	773
1,200	804	39	765
1,792	731	23	708
2,390	665	22	643
3,580	551	22	528
4,780	456	21	436
5,980	379	21	358
7,170	316	21	296
9,560	220	21	199
12,000	155	21	134
14,300	112	21	91
19,100	62	20	42
23,900	39	20	19
29,900	27	20	7
35,800	23	20	3
41,800	21	20	1

Data Used in Calculations: Rod diameter $D = 5.08$ cm. Surface heat-transfer coefficient = 0.039 per cm for both steel¹¹ and Macor. Thermal diffusivity = 0.108 cm²/s for steel¹² and 0.0054 cm²/s for Macor.⁸ CTE of steel¹³ = 14.3 × 10⁻⁶ °C⁻¹; CTE of Macor⁸ = 9.35 × 10⁻⁶ °C⁻¹.

ence of the braze and the metallized layers, and use a Timoshenko approach,¹⁴ as follows.

Consider a portion of the steel specimen of unit length and unit cross-section, brazed together with a Macor specimen of equal length and cross-section. At a given time after cooling from the brazing temperature, the temperature of the steel is u_s and $\Delta u_s = 950 - u_s$, while the temperature of Macor is u_m and $\Delta u_m = 950 - u_m$. The steel specimen has thus shrunk from unit length to $1 - f_s \Delta u_s$, while the Macor has shrunk to $1 - f_m \Delta u_m$ (Figure 5). The steel has shrunk more than Macor, since both f_s and Δu_s are greater than f_m and Δu_m , respectively. To maintain joint integrity, the originally stress-free but overshrunk steel must be stretched with dynamic tensile stress s_s^* by the adjoining Macor to length y from length $1 - f_s \Delta u_s$, while the undershrunk Macor must be compressed with dynamic compressive stress s_m^* by the steel to the same length y from length of $1 - f_m \Delta u_m$ (Figure 5).

Hence, the tensile stress in the steel is

$$s_s^* = E_s (y - 1 + f_s \Delta u_s) / (1 - f_s \Delta u_s)$$

where E_s is the Young's modulus of steel (2.11×10^4 kg/mm²).

The compressive stress in Macor (s_m^*) is

$$s_m^* = E_m (1 - f_m \Delta u_m - y) / (1 - f_m \Delta u_m)$$

where E_m is the Young's modulus of Macor (3.52×10^4 kg/mm²).⁸

Apparently, $s_s^* = s_m^*$. Hence,

$$y = [(1 - f_m \Delta u_m) E_m + (1 - f_s \Delta u_s) E_s] / (E_s + E_m)$$

The computed stresses, shown in Figure 6, exceed 37.1 kg/mm², well above

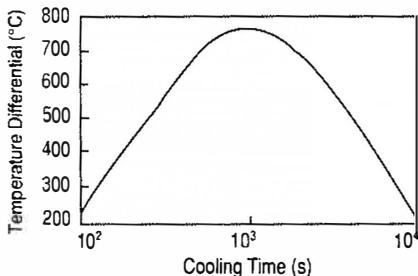


Figure 1. The variation of temperature mismatch with time.

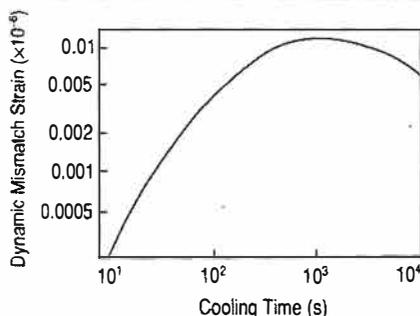


Figure 2. The variation of dynamic mismatch strain ($\Delta \epsilon^*$) with time.

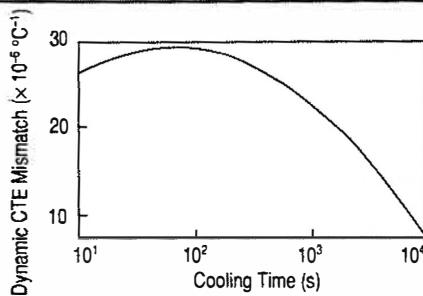


Figure 3. The variation of dynamic CTE mismatch with time.

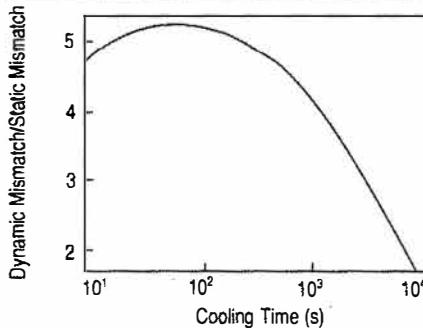


Figure 4. The ratio of dynamic to static CTE mismatch with time.

Macor's flexural strength of 10.5 kg/mm² or even its comprehensive strength of 35.2 kg/mm².

Statically, Macor only marginally "matches" a few low-expansion metals.⁸ Because of the high dynamic mismatches in CTE, strain, and stresses, the inadequate mechanical strength and thermal resistance of most conventional metal-ceramic joints in general, and steel-Macor joints in particular, are not surprising.

MECHANICALLY EQUIVALENT STRESS

Stresses are stresses no matter how they originate. Stresses due to externally applied loads, internal residual stresses, phase transition-induced stresses, thermal-mismatch stresses, and their combinations all cause the weaker ceramic to fail, precisely when the combined stresses exceed a certain fracture strength of the ceramic. This failure always occurs at the moment of maximum temperature differential between the metal and ceramic.

Qualitatively, the higher the allowable air or water-quench temperature or severity, the higher the actual dynamic-mismatch stress and joint strength. From the above analysis, there is even a calculable maximum "mechanically equivalent stress" from each quench treatment.

With standardized cooling or quenching conditions, such as rapid (e.g., within 0.5 s) 20°C air-cooling or ice-water quenching, there is a one-to-one correspondence between the joint strength (at, for example, 20°C) and the allowable initial cooling or quenching temperature. This temperature can thus be a direct measure of the mechanical

strength of the joint with a specific joint configuration (e.g., cylindrical, end-to-end) and size (e.g., 5.08 cm in diameter).

Standard tensile or flexure tests are often difficult for metal-ceramic joints because of the critical jiggling, sample-alignment, and loading requirements. Actual metal-ceramic joints often also have complex geometries, and special material, size, or composition and property profile combinations. All these conditions can make the standard mechanical test results difficult to reproduce and extrapolate to actual service conditions, or to determine if valid specifications have been met.

Yet, a controlled cooling or quenching test is simple and fast. It can be applied to a joint of any practical shape and size. There are no errors due to sample jiggling, aligning, and loading. Nor are there any unknown joint damages due to handling prior to or during the actual testing. The results are often more relevant and immediately useful without extrapolations as to sizes, shapes, joint configurations, and thermal shock environments. It is particularly useful and cost-effective for the following cases:

- Joints of complex geometries and shapes.
- Very large or small samples.
- Joints of combinations of materials with widely different mechanical properties.
- Joints that fail under dynamic cooling or heating conditions, which are difficult to duplicate on standard testing machines.
- Joints of delicate parts that are hard to jig, align, or load. For example, it would be not only very costly but

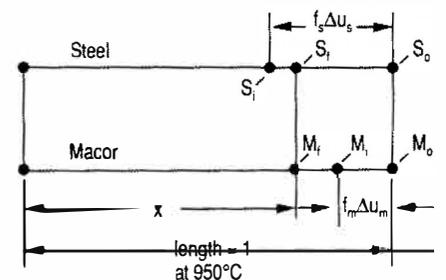


Figure 5. A schematic diagram used in the calculation of dynamic mismatch stresses.

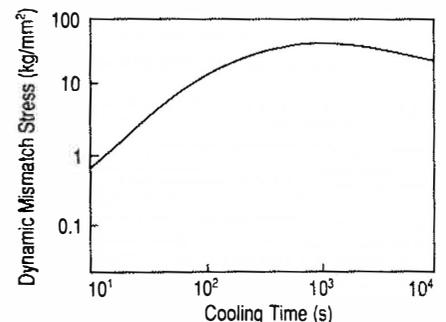


Figure 6. The variation of dynamic mismatch stresses with time.

difficult to develop the necessary equipment and procedure for determining the bond strength of an irregular diamond crystal bonded onto a copper substrate for electronic heat-sink applications.

- Peeling, spalling, microcracking, and adherence to substrates of thin films.

MINIMIZING DYNAMIC MISMATCH

The dynamic-mismatch strains and stresses computed above were unexpectedly high. Therefore, new methods must be developed to minimize the dynamic mismatch stresses on the relatively weak ceramic. The following two methods, used singly or in combination, will reduce these high dynamic-mismatch stresses and strains:

- Radially grading the thermal conductivity, CTE, and Young's modulus of the braze metal, to ensure that the maximum residual mismatch stress, after absorption in the braze, will not exceed the local material strength in the ceramic at any location or time.
- Providing a soft, yieldable braze-metal layer to absorb within this layer much of these mismatch stresses so that the relatively weak ceramic is no longer subjected to high stress.

These methods may involve the use of a composite, radially graded braze disk. Such a disk has a pure copper central core, which is placed inside the opening of an outer 70:30 cartridge brass ring or washer. The CTE of pure copper is $16.5 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$, while that of the cartridge brass¹³ is $19.9 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$. Also, the Young's modulus of the brazing-annealed, dead-soft pure copper is much lower than that of the cartridge brass. The thermal conductivity of the pure copper central core at 0°C is $4.03 \text{ W}/(\text{m}\cdot\text{K})$, while that of the outer cylindrical tube with 30% Zn in Cu is $1.14 \text{ W}/(\text{m}\cdot\text{K})$.⁹

The combination of high thermal conductivity and low CTE and Young's modulus in the core region of the joint achieves the required results. In a steel-ceramic joint, the maximum dynamic mismatches in temperatures, CTE, and thermal strains or stresses occur at the axial centers of the interfacial region. A dead-soft, brazing-annealed, pure copper therefore occupies the core region. This copper has a small Young's modulus and a yield strength less than the fracture strength of the ceramic. It is easily deformable to absorb and relieve much of the dynamic-mismatch thermal strains and stresses. Pure copper also has a relatively low CTE to reduce these mismatch effects in the first place. In addition, the copper is a good thermal conductor, equalizing the temperature between the metal and ceramic to fur-

ther minimize mismatch strains and stresses.

On the other hand, the periphery of the braze disk is made of relatively more expansive but thermally lower-conducting 70:30 brass. At the peripheral region, the mismatch-temperature differentials are relative small. The higher Young's modulus of the cartridge brass is even desirable at the peripheral region to enhance the joint rigidity.

This composite braze disk design will thus provide the radially graded profiles of braze composition, CTE, ductility, and thermal conductivity needed to minimize the critical dynamic-mismatch stresses.

The composite braze metal disks can be made by, for example, multiple printing, metallurgically cladding, or mechanically press-forming a sphere or disk inside a washer, or by slicing concentric metal tubes of graded compositions with a solid pure-metal core.

Elemental interdiffusion during braze manufacture, the brazing operation, or special pre- or post-brazing heat treatments produces a more diffused composition profile in the braze disks and leads to efficient lateral-grading results for a given transverse size of the bonded region. More description of the radially graded seals is given in Reference 15.

If these two methods are still insufficient to prevent dynamic thermal-mismatch failures, the conventional axial elemental grading or composition changes may also be added. Instead of copper (melting point $1,083^\circ\text{C}$), one can select other yieldable metals such as silver (961.9°C), gold ($1,064.4^\circ\text{C}$), tin (232.0°C), zinc (419.6°C), lead (327.5°C), antimony (630.5°C), cadmium (320.9°C), aluminum (660.4°C), magnesium (648.8°C), gallium (29.8°C), indium (156.4°C), thallium (303.5°C), or bismuth (271.3°C), with the appropriate alloying elements needed for grading.¹⁵ Higher-melting braze metals may also be used for high-temperature structural metal-ceramic joints.

NEW METAL-CERAMIC JOINTS

A better understanding of the science of metal-ceramic bonding, such as the dynamic matching approach given in this paper, has enabled the development of many difficult-to-make metal-ceramic joints. For example, "unmatched" graphite-carbon steel (SAE 1010) joints have been repeatedly made that withstand rapid air quenches from 950°C and ice-water quenches from 800°C .¹⁶ These joints are almost indestructible mechanically when repeatedly pounded with a 340 g hammer or other heavy-metal objects, confirming their excellent flexure-test results.¹⁶ Forced fractures of the joints occur away from the bonding interfaces, indicating defect-free bonding regions.

In thermomechanical shock resistance,

these metal-ceramic joints compare favorably relative to ceramic materials for the U.S. Department of Energy (DOE) automotive-engine program.¹⁷ Even the best Japanese metal-ceramic joints made with expensive, "matching" high-alloy steels have maximum practical useful temperatures of only 600°C .² The DOE ceramic engine materials, developed during the past ten years at high cost, are only specified to withstand air quenching from unspecified high temperatures to 204°C in 30 s, not the 10,000 times more severe water quenching to 0°C in less than 1 s.¹⁷

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