Zircaloy Thermal Expansion

Preliminary Recommendation

α - Phase for 300 K < T < 1083 K Single Crystal

The preliminary recommendation for the thermal expansion of Zircaloy-2 and Zircaloy-4 in the α-phase are equations given in MATPRO [1] that were determined by fitting the data from measurements of Bunnell et al. [2] converted to the orientation of the single crystals. Because Zircaloy is anisotropic, thermal expansions in three orthogonal directions are required. In development of the thermal expansion model, basal plane symmetry was assumed for the single crystal so that the thermal expansions along two of the crystal axis are equal ($\varepsilon_{11} = \varepsilon_{22}$). The MATPRO equations for Zircaloy single crystal thermal expansion are:

$$\varepsilon_{11} = 4.95 \times 10^{-6} T - 1.485 \times 10^{-3} \quad (1)$$

$$\varepsilon_{33} = 1.26 \times 10^{-5} T - 3.78 \times 10^{-3} \quad (2)$$

where $\varepsilon_{11} = (\Delta L/L)_{11}$ is the circumferential single crystal thermal expansion in m/m,

$\varepsilon_{22} = (\Delta L/L)_{22}$ is the radial single crystal thermal expansion in m/m = ε_{11},

$\varepsilon_{33} = (\Delta L/L)_{3}$ is the axial single crystal thermal expansion in m/m, and

T is the temperature in K.

To calculate the cladding thermal expansion from these single crystal thermal expansions, the orientation of the crystal structure of the cladding is required so that a volume weighted average over the cladding can be done. This averaging requires a pole figure. The thermal expansion of the cladding in the laboratory system (Lab) is calculated from the thermal expansions for single crystals from the relations:

$$\varepsilon_{11}(Lab) = \varepsilon_{11}(\sin^2\varphi) + \varepsilon_{22}(\cos^2\theta\cos^2\varphi) + \varepsilon_{33}(\sin^2\theta\cos^2\varphi)$$
$$\varepsilon_{22}(Lab) = \varepsilon_{11}(\cos^2\varphi) + \varepsilon_{22}(\cos^2\theta\sin^2\varphi) + \varepsilon_{33}(\sin^2\theta\sin^2\varphi) \qquad \textbf{(3)}$$
$$\varepsilon_{33}(Lab) = \varepsilon_{22}(\sin^2\theta) + \varepsilon_{33}(\cos^2\theta)$$

where (*Lab)* designates the cladding or laboratory system,

ϕ is the angle between the circumferential direction of the cladding and the projection of the c-axis of the single crystal onto the circumferential-axial plane of the cladding, and

θ is the angle between the radial direction of the cladding and the c-axis of the single crystals.

In the MATPRO manual, ε_{11} (Lab) is defined as thermal expansion of the cladding in the circumferential direction, ε_{22} (Lab) as the thermal expansion of the cladding in the axial direction, and ε_{33} (Lab) as the thermal expansion of the cladding in the radial direction.

α - Phase for 300 K < T < 1083 K (Cladding when orientation not known)

Very often, the orientation of the crystalline c-axis in the cladding is not known. In this case, the equations reported in the MATPRO manual [1] for the data of Bunnell et al. [2,3] are recommended because these equations provide reasonable agreement with other cladding thermal expansions in the same directions. However, the directions for ε_{11} (Lab) and ε_{33} (Lab) given in the MATPRO manual are not consistent with the cladding direction labels reported by Bunnell et al.[2,3] In fact, the Bunnell et al. [2] "diametral" data are tabulated in the MATPRO manual as "circumferential" thermal expansions. Bunnell et al. [2,3] report no thermal expansion data for the cirumferential direction. The direction labels given below are the directions for the experimental data reported by Bunnell et al.[2,3] and are consistent with other experimental data. The recommended equations for the thermal expansion of Zircaloy cladding are:

$$\left(\frac{\Delta L}{L}\right)_{Diam} = \varepsilon_{11}(Lab) = -2.128 \times 10^{-3} + 7.092 \times 10^{-6}\, T \qquad (4)$$

$$\left(\frac{\Delta L}{L}\right)_{Axial} = \varepsilon_{22}(Lab) = -1.623 \times 10^{-3} + 5.458 \times 10^{-6}\, T \qquad (5)$$

$$\left(\frac{\Delta L}{L}\right)_{Circum} = \varepsilon_{33}(Lab) = -2.998 \times 10^{-3} + 9.999 \times 10^{-6}\, T \qquad (6)$$

Figure 1 shows the Zircaloy-4 diametral thermal expansion data of Bunnell et al. adjusted to give zero at 300 K and Eq. (4), which is the MATPRO equation for ε_{11} (Lab) for the data of Bunnell et al. {labeled “MATPRO 11 Lab B” in Figure 1}. The α-phase Zircaloy-4 axial thermal expansion data of Bunnell et al. adjusted to zero at 300 K and the MATPRO equation for the axial thermal expansion for cladding with the orientation of Bunnell’s data, Eq.(5), are shown in Figure 2.

Transition region between the α- and β-phases, 1035K < T < 1144 K

The recommended equation for Zicaloy-4 thermal expansion in the axial direction is:

$$\left(\frac{\Delta L}{L}\right)_{Axial} = -6.528 \times 10^{-3} + 9.796 \times 10^{-6}\, T + 6.187 \times 10^{-4}\, e^{\frac{(T-1063)^2}{2130}} \qquad (7)$$

Insufficient data are available to recommend an equation in the transition region for other directions of the cladding in the laboratory (cladding) frame or for the single crystals.

β-phase, T > 1144 K

The recommended equation for Zircaloy-4 cladding thermal expansion in the axial direction in the β-phase is

$$\left(\frac{\Delta L}{L}\right)_{Axial} = -6.394 \text{ x } 10^{-3} + 9.7 \text{ x } 10^{-6}\, T \qquad (8)$$

Although insufficient data are available to recommend equations for the β-phase for the other two orthogonal directions, the temperature behavior, ie. the slope, may be assumed to be the same as for the axial direction. This slope, 9.7 x 10^{-6}, is the slope for the average thermal expansion of zirconium in the β-phase. It is also the recommended slope for the thermal expansion of Zircaloy-4 single crystals in the β-phase.

Recommended values, calculated with the recommended equations, Eq.(4) through Eq.(8) are given in Table 1. Figure 3 shows the recommended values for the linear thermal expansion of Zircaloy-4 in the axial direction for the α-phase, transition region and β-phase. Uncertainies have been included in the figure.

Uncertainty

The uncertainties given in the MATPRO manual for the Zircaloy-4 single-crystal thermal expansion in the α-phase are respectively, 8% for the axial direction, ε_{33}, and 12% for the circumferential direction, ε_{11}. The uncertainty for the diametral thermal expansion of Zircaloy-4 cladding in the α-phase, as defined in Eq.(4), is 15%. The uncertainty for the axial thermal expansion of Zircaloy cladding in the α-phase, as given in Eq.(5), is 12%. These uncertainties are based on the scatter in the available α-phase data. The β-phase uncertainty is 20%. This uncertainty has been chosen large enough so that the uncertainty bands include the β-phase data for soft Zircaloy-2 and the data for zirconium. In the transition region, the uncertainty is assumed to increase linearly from 12% to 20%

Discussion

α - Phase, 300 K < T < 1083 K

The equations for the single crystal linear thermal expansion of Zircaloy-4 in the α-phase are from a model developed for the MATPRO database of RELAP [1]. The model was developed using the the Zircaloy-4 thermal expansion data and equations obtained by Bunnell et al.[2,3] Because the data and fitting equations of Bunnell et al. did not give zero expansion at 300 K, they were first adjusted to give zero at 300 K. The data were fit and the equations converted from the laboratory frame of the cladding to the single crystal frame using Eq.(3) and the orientation of the single crystals in the cladding. The angles θ and φ that define this orientation for the data of Bunnell et al.[2,3] are: $\theta = 35.67^o$ and $\varphi = 25.10^o$. The resulting equations that relate the laboratory frame thermal expansion data of Bunnell et al. to the single crystal thermal expansions ε_{11} and ε_{33} are:

$$\left(\frac{\Delta L}{L}\right)_{Diam} = \varepsilon_{11}(Lab) = 0.72\varepsilon_{11} + 0.28\varepsilon_{33} \tag{9}$$

$$\left(\frac{\Delta L}{L}\right)_{Axial} = \varepsilon_{22}(Lab) = 0.94\varepsilon_{11} + 0.06\varepsilon_{33} \tag{10}$$

$$\left(\frac{\Delta L}{L}\right)_{Circum} = \varepsilon_{33}(Lab) = 0.34\varepsilon_{11} + 0.66\varepsilon_{33} \tag{11}$$

where the single crystal expansions in the radial and circumferential directions are equal, ie., $\varepsilon_{11} = \varepsilon_{22,}$ and ε_{33} is the single crystal expansion in the axial direction. Substituting the MATPRO single crystal thermal expansions for ε_{11} and ε_{33} from Eq.(1-2) in Eq.(9-10) gives Eqs. (4-6). In Eqs.(4-6) and Eqs.(9-10), the labels, *Diam, Axial, Circum,* refer to the diametral, axial, and circumferential thermal expansions for the cladding and are consistent with the directions reported by Bunnell et al. [2,3]. In MATPRO, ε_{11} (*Lab*) is termed the "circumferential" expansion in the laboratory frame. However, Figure 4, which shows the MATPRO values for ε_{11}*(Lab)*, ε_{22}*(Lab)*, and ε_{33}*(Lab)* for cladding with Bunnell's orientation and the axial and diametral Zircaloy-4 data of Bunnell et al. adjusted to zero at 300K, indicates that ε_{11} (*Lab*) as defined in Eq.(9) and Eq.(4) is consistent with the diametral thermal expansion values of Bunnell et al. In addition, Figure 4 shows that ε_{33}*(Lab)* is not consistent with either the diametral or axial data and ε_{22}.

As a check on the reliability of the single crystal equations, Eq.(1) and Eq.(2), developed from the data of Bunnell et al.[2,3], these equations were compared with old data of Douglas [4], Scott [5], and Kearns [6]. The MATPRO manual reports that the data in the axial direction gave agreement within 10% and that most of the data in the circumferential direction agreed within 20%. Comparisons were also made with plate thermal expansions in the longitudinal and transverse directions obtained by Mehan and Wiesinger [7]. The percent difference between the MATPRO model slope and that of the data of Mehan and Wiesinger in the longitudinal direction is 15%. The percent difference for the transverse direction is 7%. Thus, the MATPRO single-crystal equations may be used to provide a reasonable representation of Zircaloy thermal expansion when the angle of orientation of the single crystals in the cladding are known.

If the orientation is not known, equations (4) through (6), which were derived to represent the data of Bunnell et al., are recommended. Although the MATPRO manual gives different equations for a "typical" LWR cladding tube of Zircaloy-4, the equations based on Bunnell's data are preferred because they give better agreement with other available data. For example, Figure 5 shows that the axial thermal expansion data of Bunnell et al.[2,3] for soft and hard samples of Zircaloy-2 cladding agree better with Eq.(5) than with the MATPRO equation for axial thermal expansion for a "typical" LWR Zircaloy-4 cladding tube. However, care must be taken in the use of Eqs.(4) through (6) when nothing is known with respect to the orientation, oxygen content, and heat treatment. In Figure 6, the Zircaloy-2 and Zircaloy-4 axial thermal expansion data of Bunnell et al. and the MATPRO axial equation that fits the Zircaloy-4 axial expansion data of Bunnell et al. are compared with the axial thermal expansion data of Peggs et al. [8] for a Zircaloy-2 pressure tube, a Zircaloy-4 fuel sheath, and a Zircaloy-2 calandria tube. Although the axial thermal expansion data for the Zircaloy-2 pressure tube is within the scatter of the data of Bunnell et al. for Zircaloy-4, the axial thermal expansion data of Peggs et al. for a Zircaloy-2 pressure tube and a Zircaloy-4 fuel sheath have a different slope and deviate from a linear dependence at around 800 K. Bunnell et al.[2,3] showed that the oxygen content has a significant effect on the thermal expansion and developed equations for the axial and diametral thermal expansion of Zircaloy-4 for oxygen content from 0.7 at% to 22.4 at% and of Zircaloy-2 for oxygen contents of 0.7 at% and 5.2 at%.

β-phase and Transition region between the α- and β-phases

The MATPRO single crystal thermal expansion equations for the β-phase have been based on the thermal expansion of zirconium in the β-phase obtained by Skinner and Johnston [9] because there was insufficient data on Zircaloy thermal expansion in the β-phase to construct a detailed model. The MATPRO equations for single crystal thermal expansion are

For T > 1244 K,

$$\varepsilon_{11} = 9.7 \times 10^{-6}\, T - 1.04 \times 10^{-2} \qquad (12)$$

$$\varepsilon_{33} = 9.7 \times 10^{-6}\, T - 4.4 \times 10^{-3} \qquad (13)$$

where ε_{11} is the single crystal circumferential thermal expansion in m/m,

ε_{33} is the single crystal axial thermal expansion in m/m,

$\varepsilon_{22} = \varepsilon_{11}$, and

T is the temperature in K.

The slope of these equations, 9.7×10^{-6}, is the slope of the linear equation that fits the linear thermal expansion data of Skinner and Johnston.

The MATPRO equations for the single crystal thermal expansion in the transition region are correlations developed using the constraint that at 1035 K, the thermal expansion must be equal to the thermal expansion in the α-phase at 1035K and at 1144 K, the thermal expansion must be equal to the thermal expansion in the β-phase. These two values are linked by a cosine function to give the expected curvature from the α-phase to the β-phase. The MATPRO single crystal equations for the circumferential thermal expansion, ε_{11}, and the axial thermal expansion, ε_{33}, in the transition region are

For $1083 \text{ K} \leq T \leq 1244 \text{ K}$,

$$\varepsilon_{11} = \left[2.77763 = 1.09822 \cos\left(\frac{T - 1083}{161} \pi \right) \right] \text{x}\, 10^{-3} \tag{14}$$

$$\varepsilon_{33} = \left[8.76758 = 1.09822 \cos\left(\frac{T - 1083}{161} \pi \right) \right] \text{x}\, 10^{-3} \tag{15}$$

where the arguments for the cosines are in radians. The large number of significant figures in these equations are to prevent discontinuities. Figure 7 shows the MATPRO single crystal thermal expansion equations for the β-phase and the phase transition region, Eqs.(12-15), the data of Skinner and Johnston for the average linear thermal expansion of zirconium in the β-phase, transition region, and upper temperature region of the α-phase, and the linear fit to the β-phase thermal expansion data of Skinner and Johnston.

Although Bunnell et al.[2,3] do not report thermal expansion for the circumferential or diametral directions in the β-phase, they report axial thermal expansions at temperatures in the transition region and the β-phase for Zircaloy-4 and hard and soft Zircaloy-2. In order to assess the reliability of the single crystal Zircaloy thermal expansion equations for the β-phase and transition region, Eq.(10) has been used to calculated the axial thermal expansion for the cladding from the single crystal values for ε_{11}, and ε_{33} given in Eqs.(12-15). In Figure 8, the cladding axial thermal expansion calculated from the MATPRO single-crystal equations for the orientation of the Zircaloy-4 cladding of Bunnell et al. (labeled "MATPRO 22 Lab B") are compared with the Zircaloy-4 and Zircaloy-2 axial thermal expansion data of Bunnell et al. for the α-phase, β-phase, and transition region. Although there is excellent agreement for the α-phase, the MATPRO values for the transition region and β-phase do not agree with the data. Because the axial thermal expansion values for the β-phase, and transition region obtained from the MATPRO single crystal equations using the orientation for the Zircaloy-4 sample of Bunnell et al. do not agree with the available data of Bunnell et al., the MATPRO single crystal equations for the β-phase and transition region are not recommended.

Figure 9 shows the three orthogonal thermal expansions for the α- and β-phases and transition region for Zircaloy-4, which were calculated from the MATPRO single crystal thermal expansions using Eqs.(9-11). The curves for the single crystal thermal expansions in each region have been included in Figure 9. The available Zircaloy thermal expansion data in the diametral direction and axial direction from the measurements by Bunnell et al. [2,3] have been included in Figure 9 for comparison with the laboratory thermal expansion equations determined from the MATPRO relations. For completeness, Figure 9 also shows the data of Skinner and Johnston for the average linear thermal expansion of zirconium and the equation which fits their data in the β-phase. This equation for the average thermal expansion of zirconium in the β-phase is

$$\left(\frac{\Delta L}{L}\right)_{Axial} = -7.200 \times 10^{-3} + 9.7 \times 10^{-6} T \qquad (16)$$

Figure 9 shows that the thermal expansions for the Zircaloy cladding in the β-phase have a slope that is similar to that of the zirconium data. Note that the transition from the α-phase to the β-phase for the available Zircaloy axial thermal expansion data does not have the large decrease predicted by the MATPRO model. In fact, even the zirconium data for the transtion from the α-phase to the β-phase do not show the same curvature that exists for the MATPTRO single crystals because the data of Skinner and Johnston for the average linear thermal expansion of zirconium in the α-phase show reasonable agreement with the data of Bunnell et al. for the α-phase thermal expansion of Zircaloy-4 in the diametral direction. The data for the β-phase axial thermal expansions of Zircaloy-4 and hard Zircaloy-2 are closer to the MATPRO Zircaloy laboratory-frame curve labeled “MATPRO 11 Lab B” than for the curve “MATPRO 22 Lab B” that gives the expansion in the axial direction in the laboratory frame for the orientation of the cladding of Bunnell et al. The data for the thermal expansion of soft Zircaloy-2 in the axial direction in the β-phase are closer to the zirconium data than the other axial Zircaloy data and indicates the effects of sample variation on the magnitude of the change in thermal expansion at the phase transition.

Figure 10 shows a linear regression fit to the Zircaloy-4 and hard Zircaloy-2 axial thermal expansion data of Bunnell et al. in the β-phase. These data are fit by the equation

$$\left(\frac{\Delta L}{L}\right)_{Axial} = -5.674\,\mathrm{x}\,10^{-3} + 9.2\,\mathrm{x}\,10^{-6}\,T \qquad (17)$$

Equation (8), which has the same slope as that of the fit to the zirconium data of Skinner and Johnston, has also been included in Figure 10. The root mean square standard deviation of the data from Eq.(17) is 0.091, whereas the root mean square standard deviation of the data from Eq.(8) is 0.095. Equation(8) has been recommended for the axial thermal expansion of Zircaloy cladding in the β-phase because all the available data for Zircaloy and zirconium in the β-phase are consistent with the slope of the zirconium data and the difference in the standard deviations from the two equations is small.

Examination of the axial data for Zircaloy-4 and hard Zircaloy-2 in the transition region between the α- and β-phases showed that these data are not consistent with the cosine function suggested in the MATPRO manual. A sine function also does not provide an adequate representation of these data. A nonlinear least squares technique was used to fit these data to Eq.(7), which has a linear and Gaussian temperature dependence. This equation was constrained to give the α- and β-phase values at the end points of the transition region. Figure 11 shows the recommended equations for the axial thermal expansion for Zircaloy-4 for the α-phase, transition region, and β-phase and the uncertainties for each phase. The uncertainty chosen for the β-phase, 20%, has been selected so that it is large enough to include the data for the axial thermal expansion of soft Zircaloy-2 in the β-phase.

References

1. D. T. Hagrman, ed., "SCDAP/RELAP5/MOD 3.1 Code Manual: MATPRO - A Library of Materials Properties for Light-Water-Reactor Accident Analysis, **NUREG/CR-6150, EGG-2720 Vol. 4** (1995).

2. L. R. Bunnell, G. B. Mellinger, J. L. Bates, and C. R. Hann, *High-Temperature Properties of Zircaloy-Oxygen Alloys*, Electric Power Research Institute Report **EPRI NP-524** (March 1977).

3. L. R. Bunnell, J. L. Bates, and G. B. Mellinger, *Some High-Temperature Properties of Zircaloy-Oxygen Alloys*, J. Nucl. Mater. **116**, 219-232 (1983).

4. D. L. Douglass, *The Physical Metallurgy of Zirconium,"* Atomic Energy Review **1**, (4), 73-74, (December 1962).

5. P. B. Scott, *Physical and Mechanical Properties of Zircaloy-2 and -4,* **WCAP-3269-41** (May 1965), as referenced by D. T. Hagrman in the MATPRO Manual (ref. 1).

6. J. J. Kearns, *Thermal Expansion and Preferred Orientation in Zircaloy,* **WAPD-TM-472**, (November 1965), pp. 17-18, as referenced by D. T. Hagrman in the MATPRO Manual (ref. 1).

7. R. L. Mehan and F. W. Wiesinger, "Mechanical Properties of Zircaloy-2," General Electric, Knolls Atomic Power Laboratory Report **KAPL-2110**, pp. 8-11 (February 1961).

8. I. D. Peggs, A. M. Stadnyk, and D. P. Godin, *Thermophysical Properties of Zirconium-Alloy Fuel-Channel Components*, High Temp.-High Pressures **8**, 441-450 (1976).

9. G. B. Skinner and H. L. Johnston, *Thermal Expansion of Zirconium between 298 K and 1600 K*, J. Chemical Physics **21** (8), 1383-1384 (August 1953).

Table 1 Recommended Values for the Linear Thermal Expansion of Zircaloy

Temperature (K)	Linear Thermal Expansion $\Delta L/L \times 10^3$ (m/m)	
	Axial	**Diametral**
300	0.00	0.00
350	0.29	0.35
400	0.56	0.71
450	0.83	1.06
500	1.11	1.42
550	1.38	1.77
600	1.65	2.13
650	1.92	2.48
700	2.20	2.84
750	2.47	3.19
800	2.74	3.55
850	3.02	3.90
900	3.29	4.25
950	3.56	4.61
1000	3.84	4.96
1050	4.33	5.32
1100	4.57	
1150	4.76	
1200	5.25	
1250	5.73	
1300	6.22	
1350	6.70	
1400	7.19	
1450	7.67	
1500	8.16	

Figure 1 MATPRO Fit to Zircaloy-4 Diametral Linear Thermal Expansion

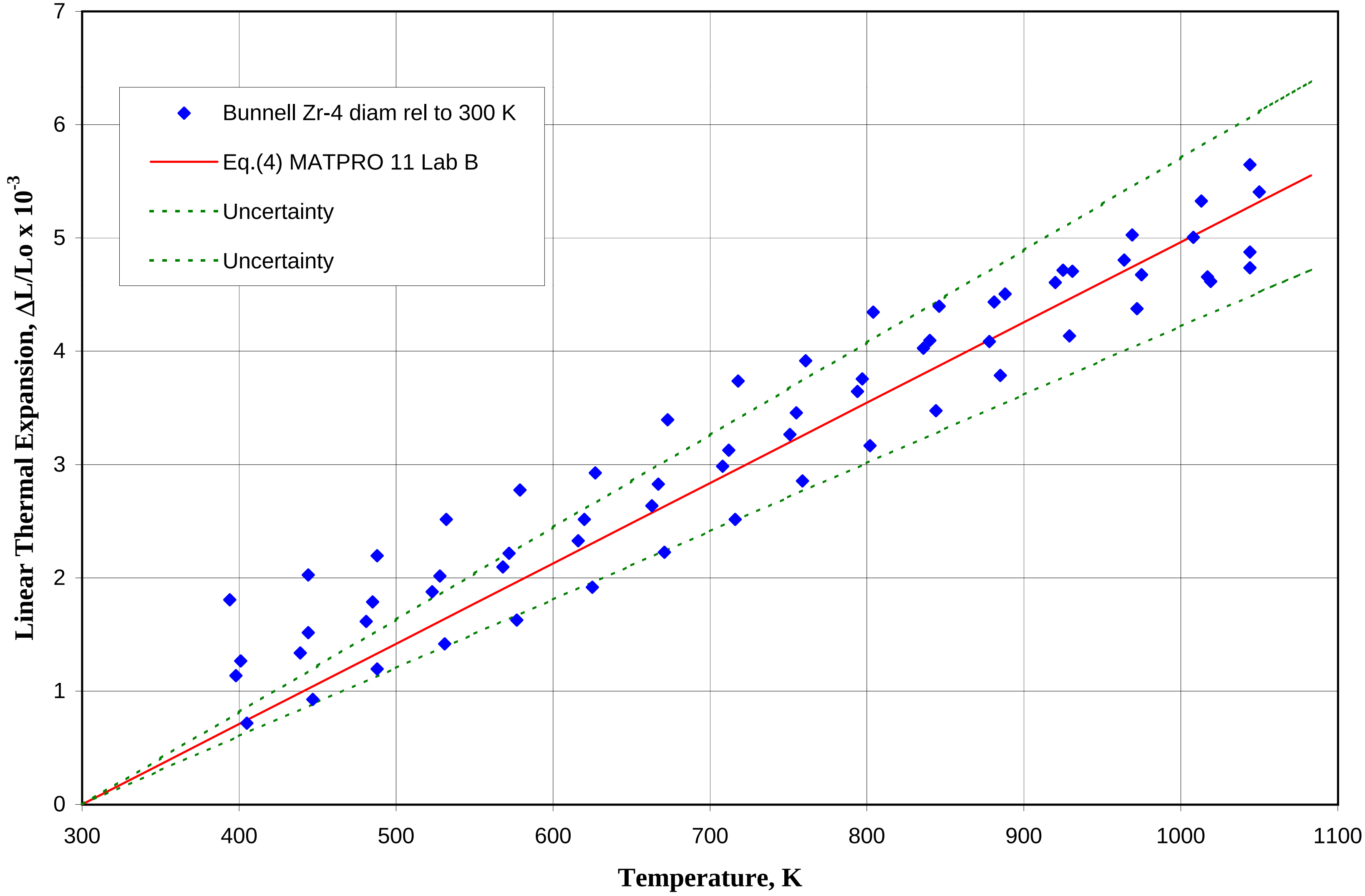

Figure 2 MATPRO Fit to Bunnell's Zircaloy-4 Axial Linear Thermal Expansion

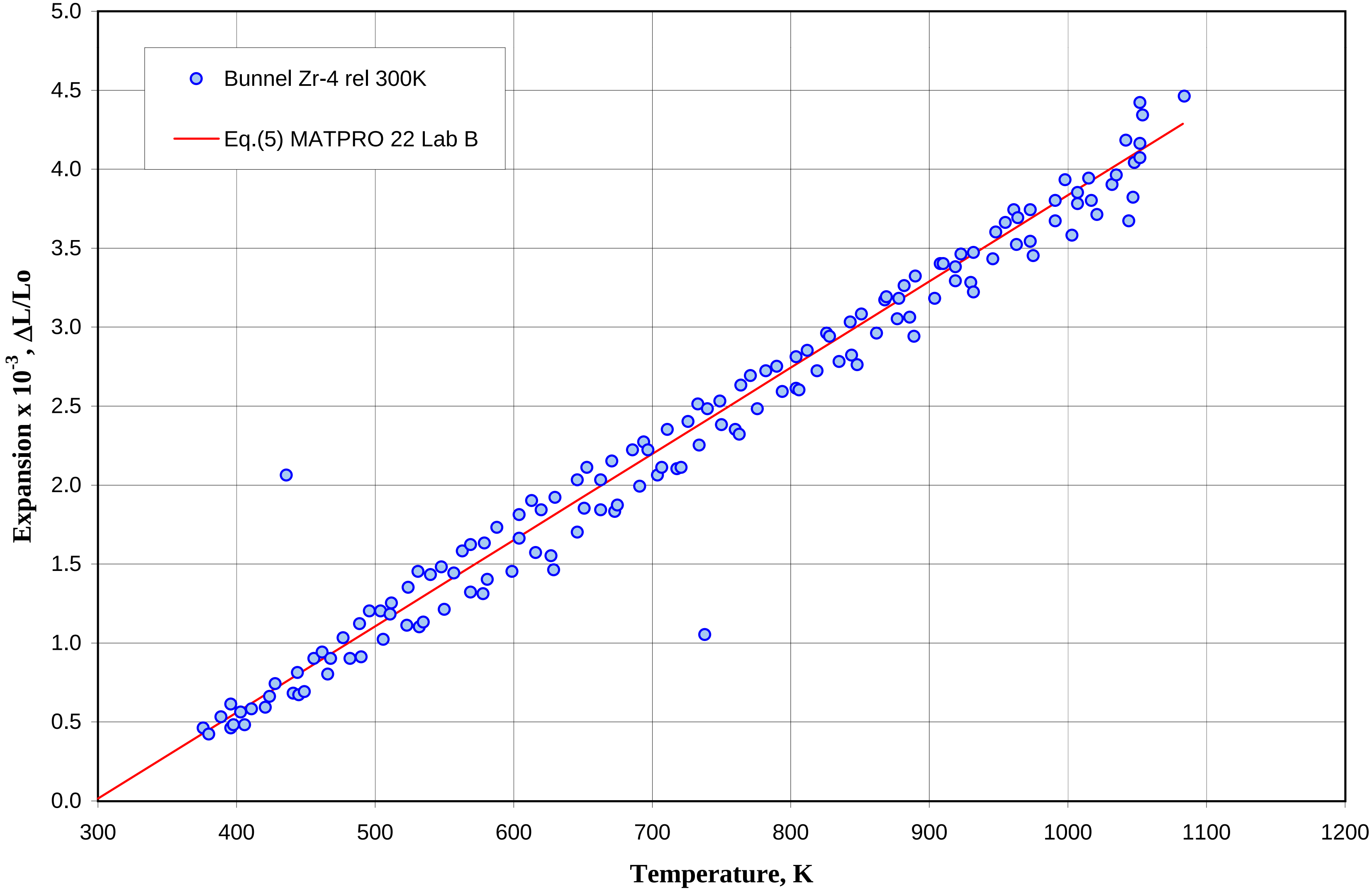

Figure 3 Zircaloy Linear Thermal Expansion in α , Transition and β Phases

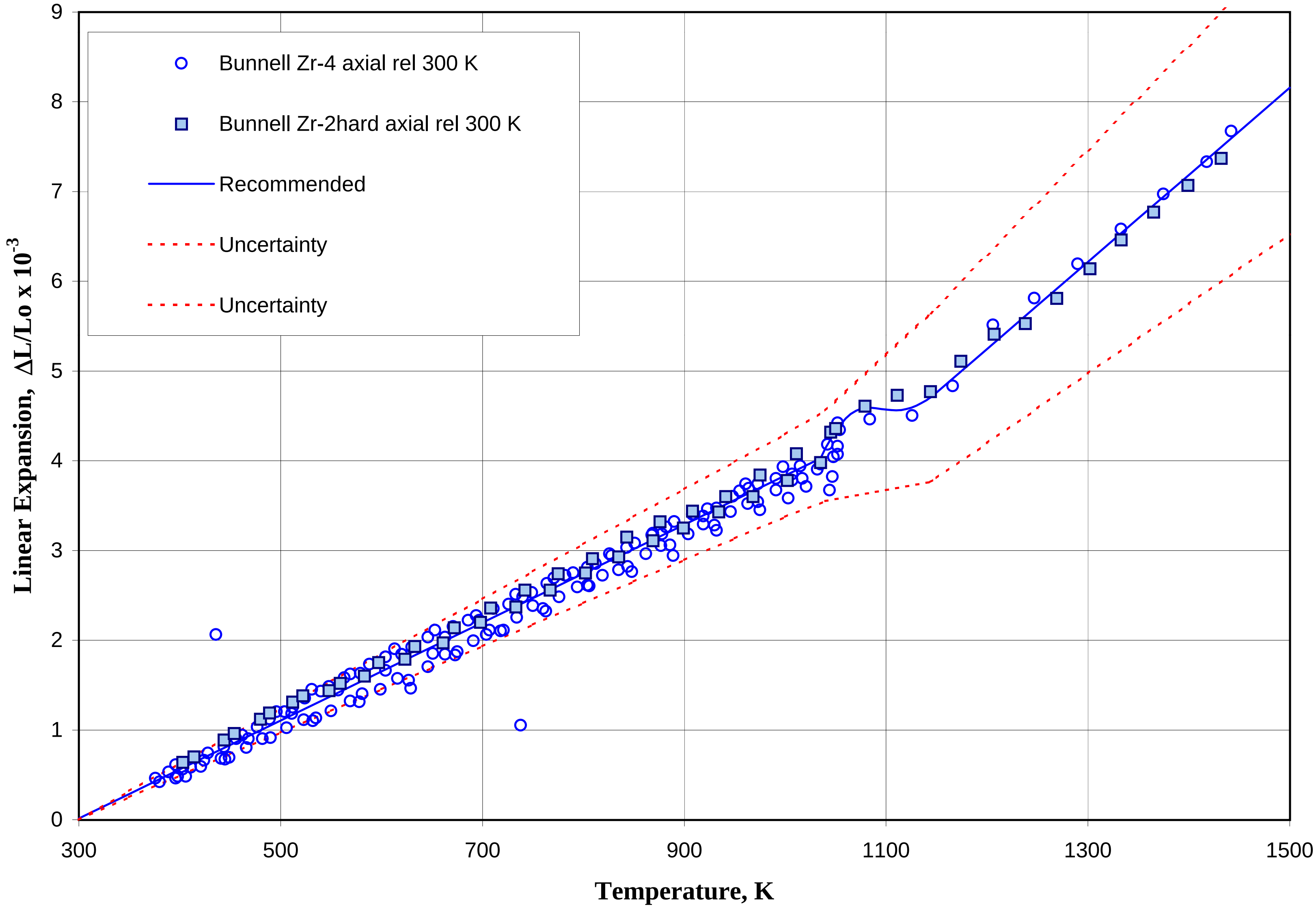

Figure 4 Comparison of MATPRO Values with Zircaloy-4 Data of Bunnell et al.

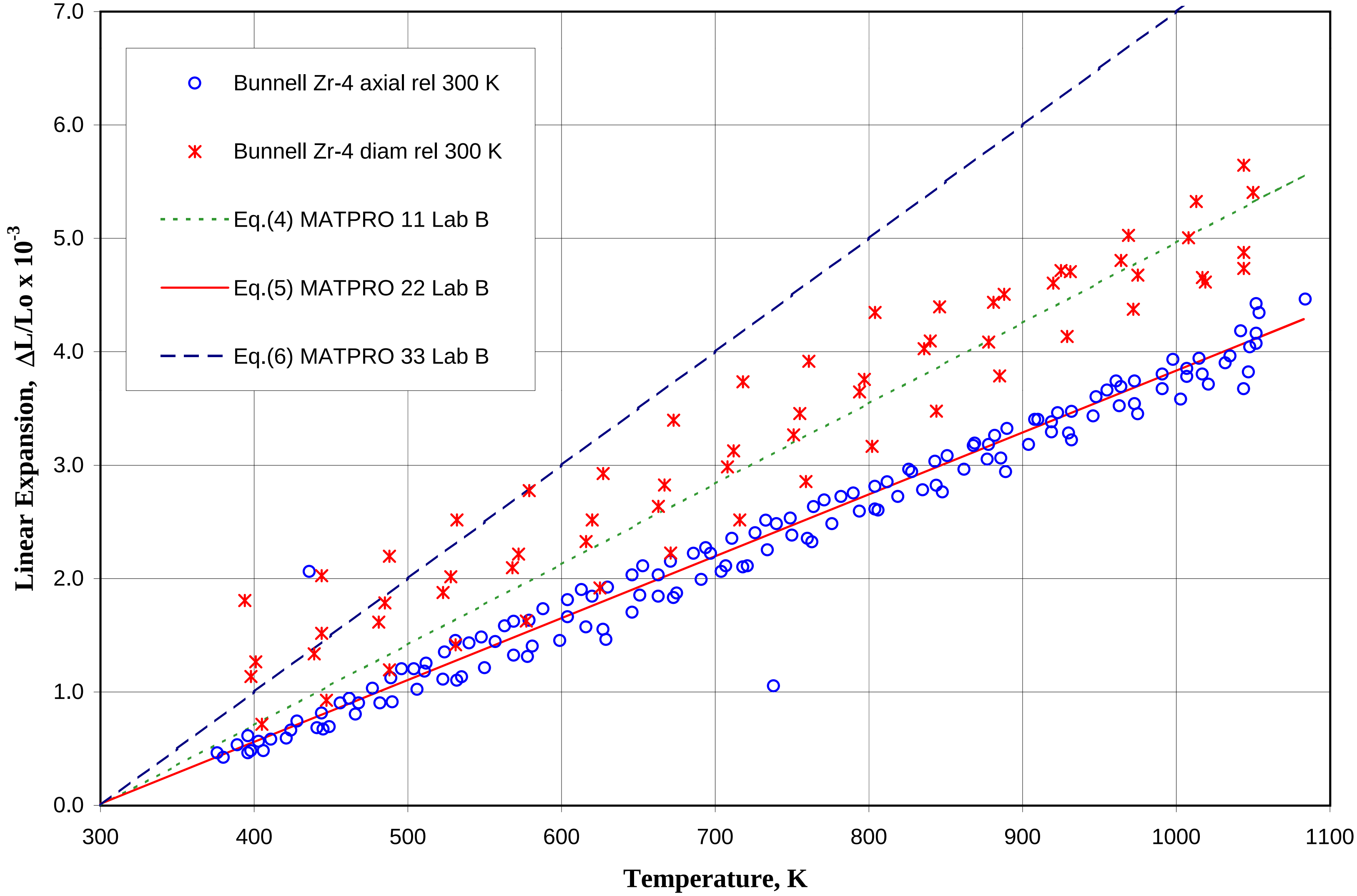

Figure 5 Zircaloy-2 Cladding Axial Linear Thermal Expansion

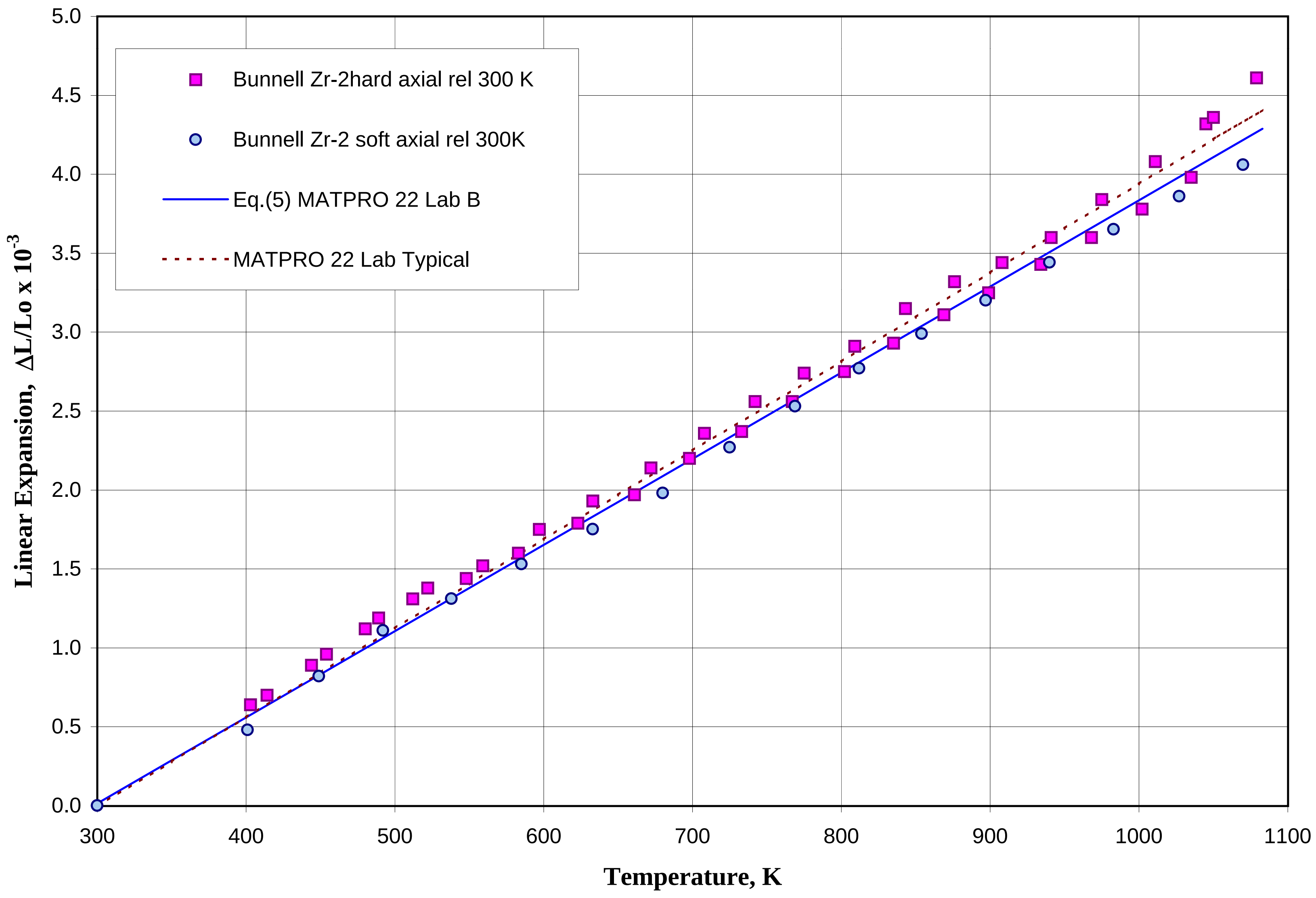

Figure 6 Zircaloy-2 and Zircaloy-4 Axial Linear Thermal Expansion

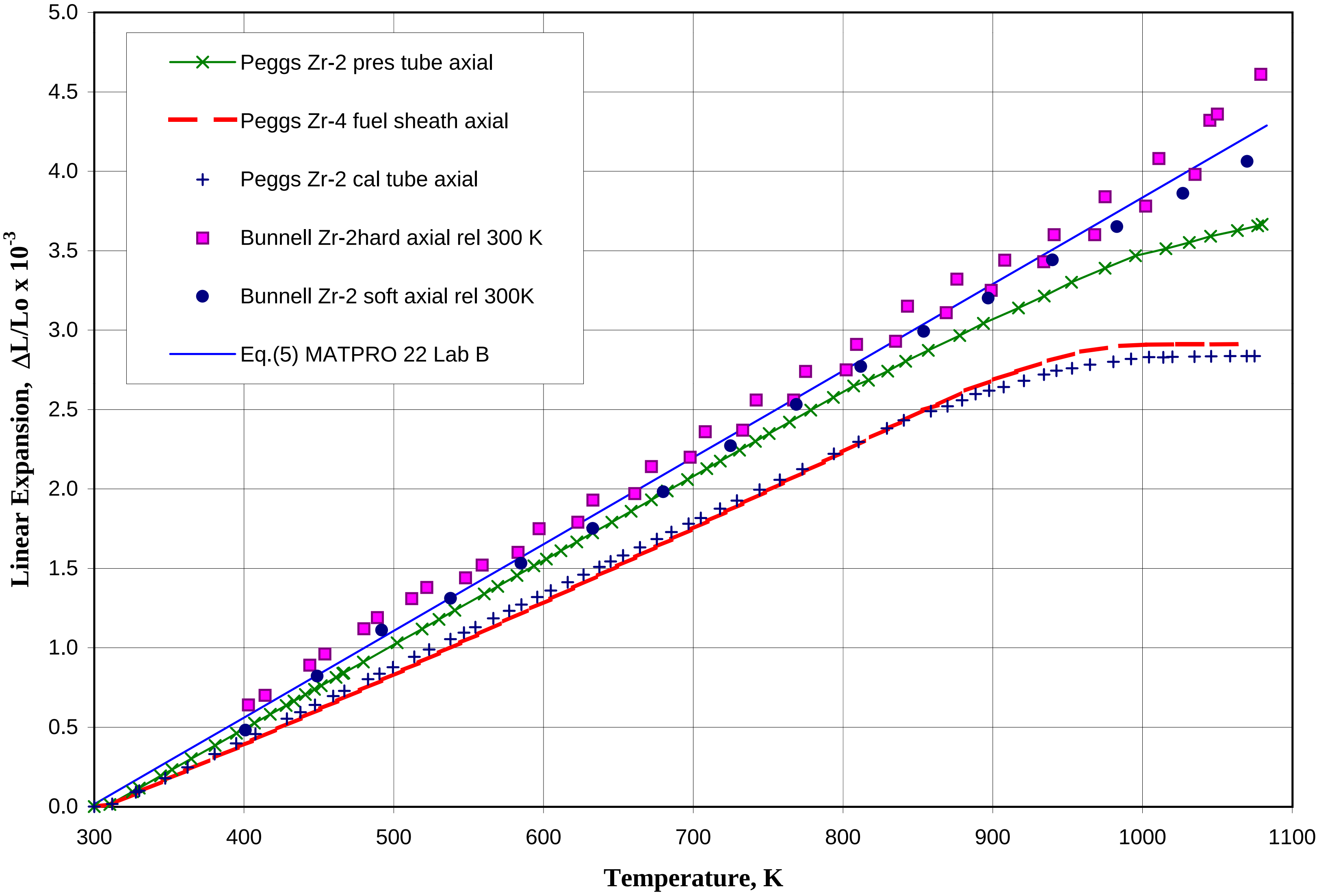

Figure 7 Zircaloy Single Crystal Linear Thermal Expansion

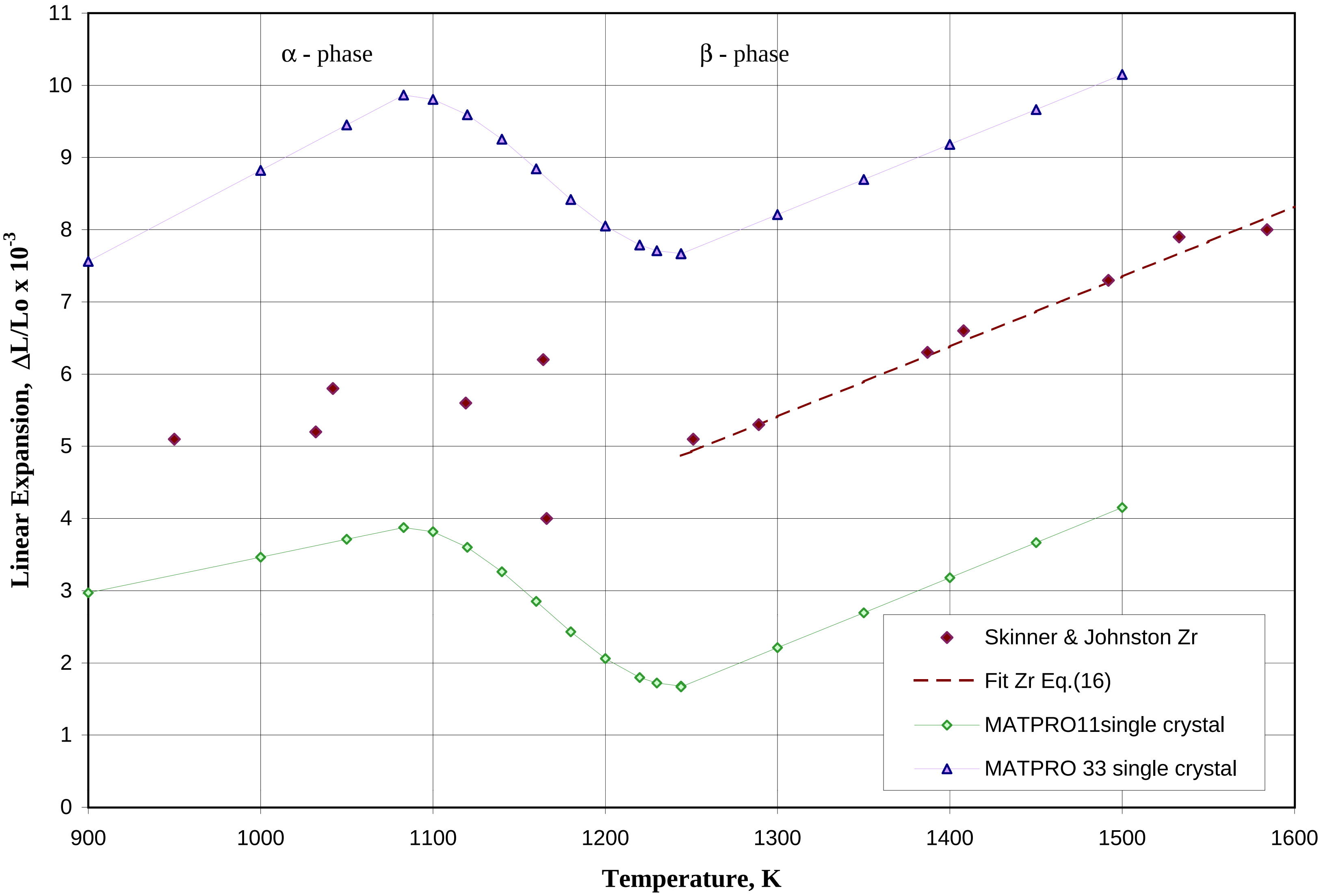

Figure 8 Zircaloy Linear Thermal Expansion

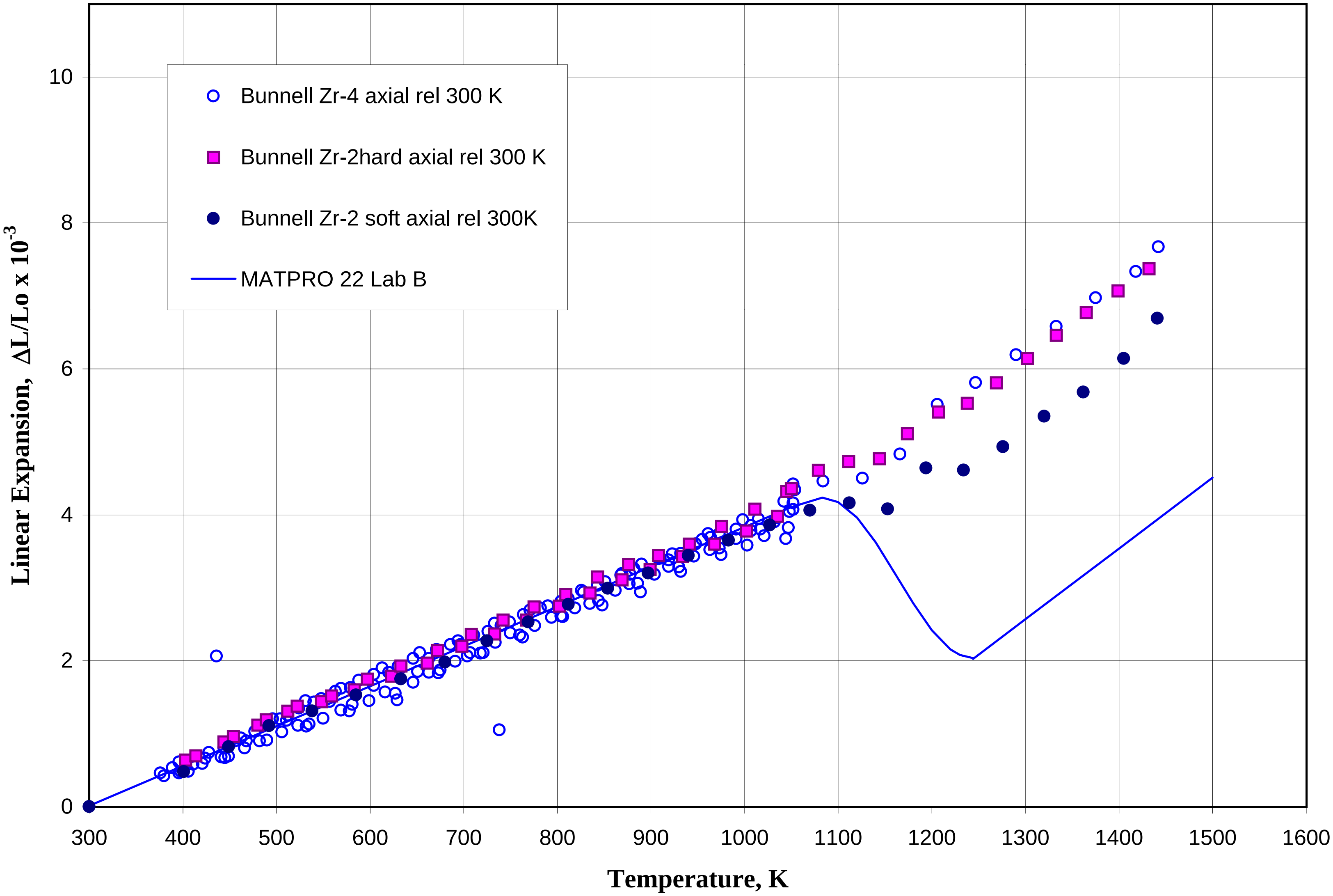

Figure 9 Zircaloy Linear Thermal Expansion

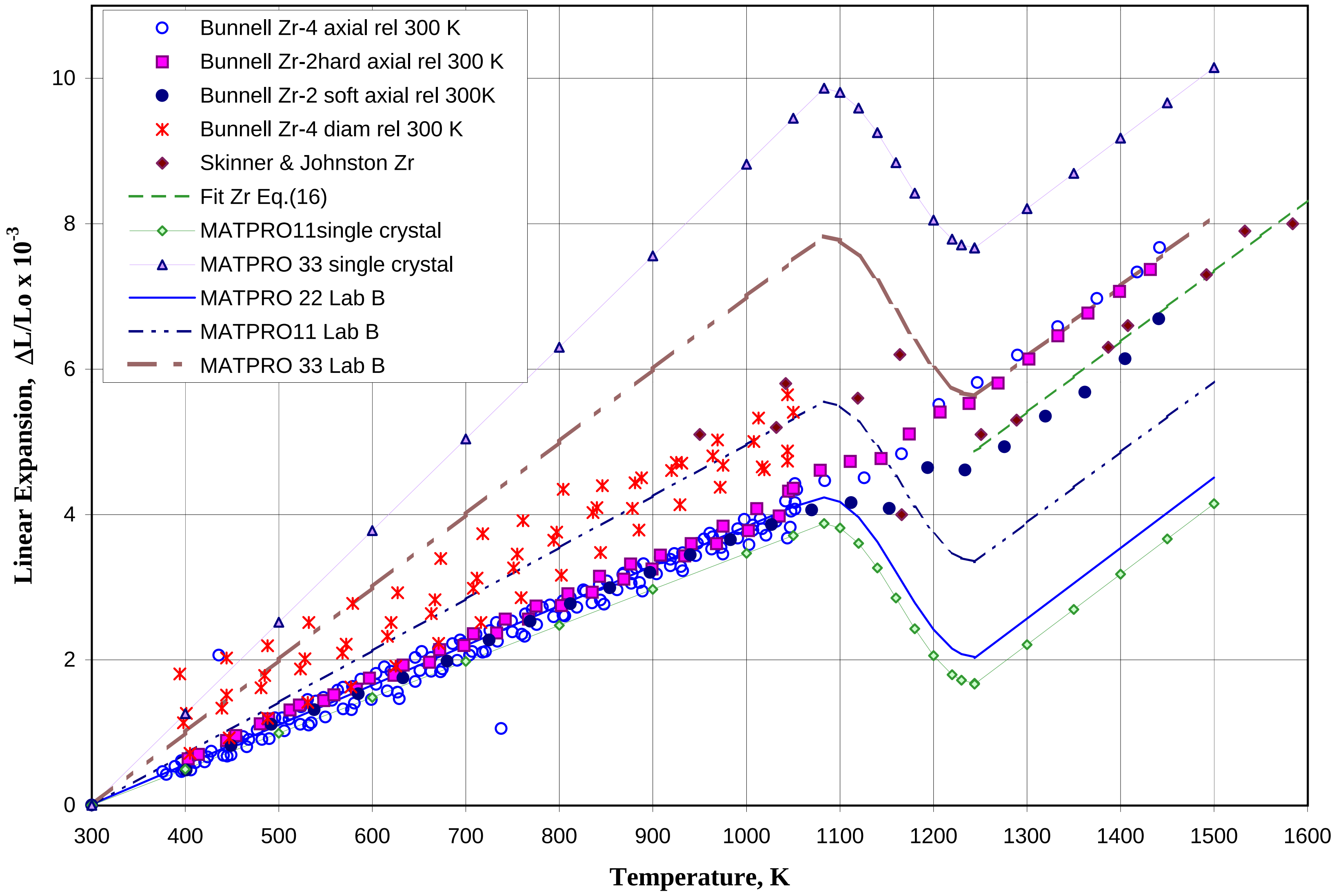

Figure 10 Zircaloy Linear Thermal Expansion in Beta Phase

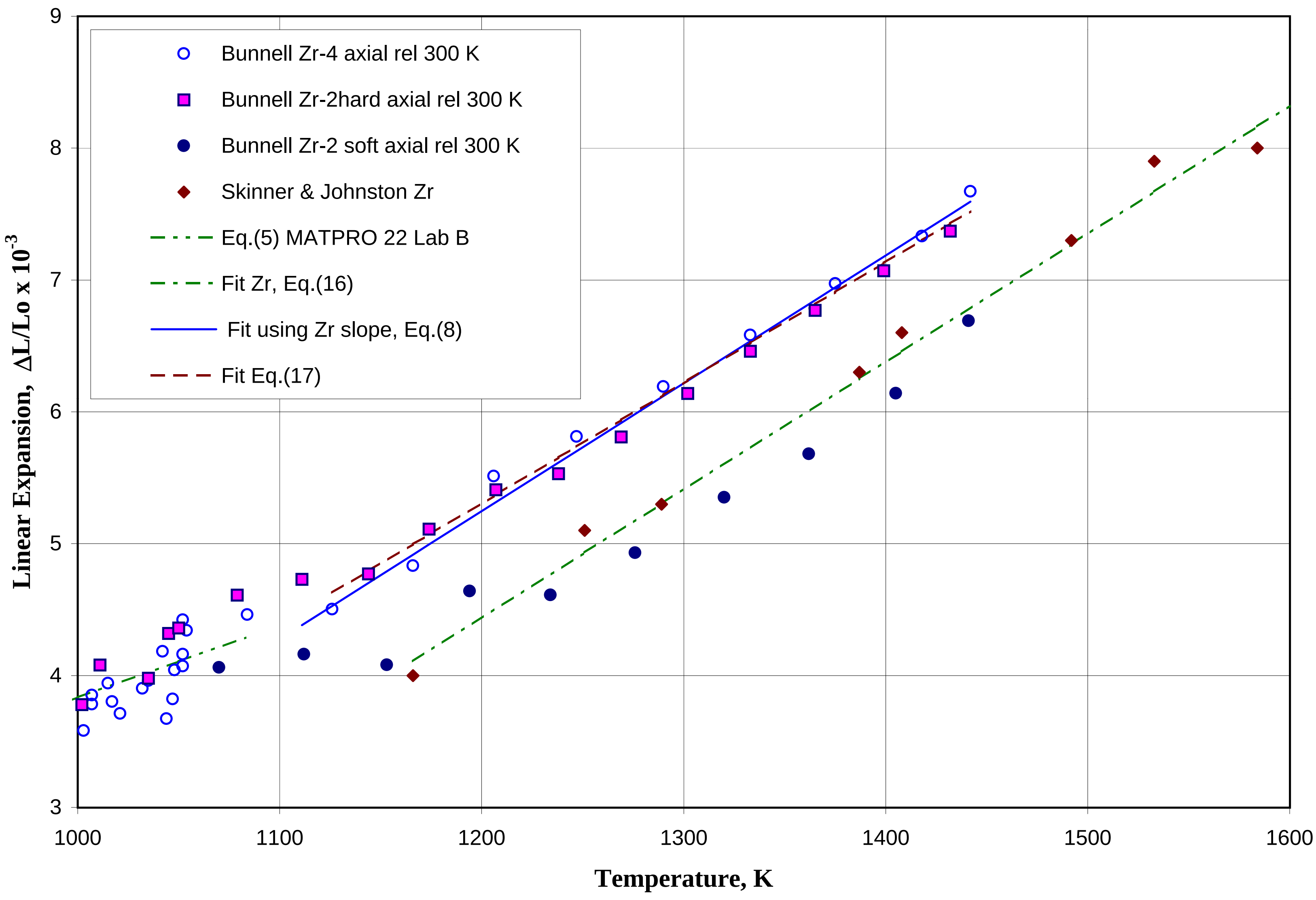

Figure 11 Zircaloy Linear Thermal Expansion in α , Transition and β Phases