**A Short Review on Tungsten Expansivity Measurements, Assessments and Modeling**

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**Abstract:** A short review on tungsten expansivity measurements and assessments, sorted in their historical order since the beginning of the last century. This work include the physical form of the tested tungsten sample, the used dilatometry technique, the temperature range of the measurement and the most important equations for linear expansion, deduced form the regression of experimental data at various era. Most important theoretical models are also listed.

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**1. Introduction**

Tungsten [1] is a refractory metal of bcc symmetry, widely used in many important industries, such as aerospace, electronics, lighting, mining, tooling and nuclear reactors. The efforts of physicists continue to enhance the accuracy of which the coefficient of thermal expansion CTE is being measured specially at high temperatures, in order to control the mechanical properties of functional parts made of this metal. Some low temperature measurements were intended to test the Grüneisen theory and some recent theoretical models. This work highlight these efforts, referring to their historical order.

**2. The Theory**

The thermal expansion [2,3] can be defined as the tendency of matter to change in volume in response to a change in temperature. Different substances expand by different amounts over small temperature ranges. The thermal expansion of uniform linear objects of length L is proportional to temperature change ΔT according to the relation:

ΔL/L = α ΔT (1)

Where α is the thermal coefficient of linear expansion. The thermal expansion can be expressed as a polynomial function of temperature of the form:

ΔL/L = co+ c1 T + c2 T2 + ...+ cn Tn (2)

Where co, c1, c2,…,cn are constants. To obtain ΔL/L this form, the experimental data of linear expansion versus temperature measured by means of dilatometers can be refitted and smoothed by regression techniques to get the value of the constants co, c1, c2,…, cn. And hence the CTE can be obtained by differentiating the previous equation with respect to T yielding the expression:

α = c1 + 2c2 T + 3c3 T2 + … + ncnTn-1  (3)

At most cases, the CTE of a metal is described by a polynomial of degree n ≥ 3, in order to comprise the non linearity of the expansion behavior over most of the solid range to melting point.

**3. Literature Review**

The chronology of thermal expansion measurements for tungsten was historically related to the major technological advances achieved at the last century. The story begins a hundred years ago, directly after the first successful powder metallurgy production of the pure metal, when tungsten replaces osmium and carbon filaments [4] in the early incandescent lamps industry at 1909. By this time the efforts of scientists were focused on increasing the lifetime of such lamps and precisely measuring the CTE of tungsten to prevent harmful thermal stresses on the filaments, and being able to design high power military lamps during WWΙ (1914-1918). First attempt was made by Fink [5,6] (1910), Langmuir [7] (1918), Gray [8] followed by the valuable work of Worthing [9,10] (1917) who conducted the first accurate CTE measurement at incandescence temperatures leading to the best fit equation in the range from 300 to 2700K:

∆L/Lo= 4.4×10−6 (T-300)+ 4.5×10−11 (T-300)² + 2.2×10−13 (T-300)³ (4)

By the end of WWΙ, tungsten was considered as strategic metal, for its important role in manufacturing high speed steel cutting tools, vacuum tubes, x-ray tubes at the early electronics era, the efforts of scientists continue to achieve accurate CTE measurements by the work of Dischs [11] (1921), Goucher [12], Benedicks and Berlin [13] (1925), Hidnert [8] (1925), Becker [14] (1927), Shinoda [15], and Burger [16] (1934).

As WWΠ (1938-1945) started, tungsten was a key constituent in more than 15000 military product including early propulsion systems, the tungsten CTE measurements stopped except for the work of Nix & MacNair [17] (1942), and that of Demarquay [18] (1945) who discovered the tungsten CTE hysteresis behavior during heating /cooling cycle.

At the end of the war it was obvious the era of nuclear energy has began the importance of tungsten as neutron deviator and low expansion refractory material reveals for the first generation of nuclear fission reactors. In addition to the heavy demands placed on tungsten for manufacturing cathode ray tubes at the beginning of commercial TV broadcasting, and as super alloys used in the early supersonic aviation, scientists continue studying the CTE of different tungsten grads, as in the work of Apblett [19] (1952), Mauer [20] (1955), Brand [21] (1956), White [22] (1958) who published the first valuable CTE data assessment, Baun [23], Fulkerson [24] (1959) who directed the first multinational project AGARD to study tungsten CTE for military purposes. In conjunction to the early advances in aerospace experiments and hypersonic aviation, the production of high purity, poly-crystalline and mono-crystalline tungsten methods has become more advanced, CTE measurements also continue to achieve wider temperature limits and better accuracy, as in the work of Anthony [25] (1960), Levingstein [26], Andrea [27] (1961), Deman [28], Neels [29] (1962), Houska [30], Dutta [31], Andres [32], Ross [33] (1963) Amoneko [34], Totskii [35], Matyushenko [36], Andres [37] (1964), Clark [38], Takamori [39], Yaggee [40], V’yugov [41] (1965), Conway [42], Rausch [43] (1966), Frantsevich [44], Conway [45] (1967), Brizes [46] (1968), Valentich [47], Knibbs [48], Nasekovskii [49] (1969), Shah [50, 51] (1971), Petukhov [52], Kirby [53], Lisovskii [54], Kraftmakher [55], Fitzer [56, 57] (1972), and Roberts [58] (1975). In his work on tungsten wire, Kraftmakher was able to use modulation calorimetry to determine the CTE of the metal at temperature above 2000 K given by the approximated formula:

α = 3.5× 10−6 + 1.4× 10−9 T +2.74×106 T-2e-36540/ T(5)

At the middle of the seventies, and due to diversity of the CTE measurements, Touloukian [59] (1975) summarizes all thermal expansion data for Tungsten and recommended the following equations:

∆L/Lo= 4.266×10−4(T-293)+8.479×10−8(T-293)²-1.974×10−11(T-293)³

293 K ≤ T <1395 K (6)

∆L/Lo= 0.548+5.416×10−4(T-1395)+1.952×10−8(T-1395)²+4.422×10−11(T-1395)³

1395K ≤ T <2495K. (7)

∆L/Lo= 1.226+7.451×10−4(T-2495)+1.654×10−7(T-2495)²+7.568×10−12(T-2495)³

 2495 K ≤ T <3600 K (8)

These equations were considered, the most accurate and best fitted for tungsten CTE data ever published at this time. However this approximation was not quit accurate near tungsten melting point, because of the absence of enough data at this limit. Later, several works were published by Waseda [60] (1975), Kirby [61] (1976), White [62] (1978), Rodriguez [63] (1981), and In Kook Suh [64] (1988).

By the end of the eighties, Miiller & Cezairliyan [65] (1990) had measured the linear thermal expansion of tungsten in the temperature range 1500–3600 K by means of a transient interferometry technique. The basic method involved rapid heating of the specimen from room temperature up to and through the temperature range of interest in less than 1 s by passing an electrical current pulse through it and simultaneously measuring the specimen temperature by means of a high-speed photoelectric pyrometer and the spacing of fringe pattern produced by a Michelson-type interferometer. The results for tungsten were expressed by the relation:

∆L/Lo= 1.3896×10−3 - 8.2797×10−7 T + 4.0557×10−9 T² - 1.2164×10−12 T3 + 1.7034×10−9 T4 (9)

Later, a number of experimental studies were published as, the work of Lahav [66] (1990) on tungsten thin film, Dubrovinsky & Saxena [67] (1997), and IAEA [68] (2006) on the SRM737. In the distinctive work of Dubrovinsky & Saxena, the *In situ* x-ray data on molar volumes of tungsten over the temperature range from 300 K to melting, was combined to the technique of spectro-radiometry and electrical resistance wire heating, hence the thermal expansion of Tungsten between 300 and 3600 K was approximated and given by:

α = 7.862× 10−6 + 6.392× 10−9  (10)

Since the achievement of Miiller & Cezairliyan, studies were mostly theoretical assessments of CTE data, and verifications of Gruneizen theory. as in the work of Guillermet & Grimvall [69] (1991),White & Minges [70, 71] (1994, 1996), Wang & Reeber [72] (1998), Dorogokupets [73] (2012), Zhang [74] (2013), Westinghouse Company [75] (2013) work on fission reactor materials, and Litasov [76] (2013). Perhaps the most valuable of these work to our experimental study, is that of White & Minges, where they included the data of transient interferometry to Touloukian’s equations in a refitted polynomial covering the range from 300 to 3500k on the form:

α =3.872×10-6 +2.562× 10−9 T-2.8613× 10−12 T2+1.9862× 10−15 T3+ 0.58608×10−18 T4+0.070586×10−21 T5 (11)

Lately, as a consequence of the advances in the field of nano technology, and plasma facing materials PFM used in the future fusion reactors, Ritz [77] (2013) carried out a study on different grades of nano-structured tungsten, which revealed that the presence of doping materials has a minor effect on the CTE behavior of different tungsten alloys. Yanwei [78] (2015), also investigated the CTE of ultra high purity and fully dense tungsten, prepared by chemical vapor deposition and found the same result.

Finally, a chronological survey (since 1910 to 2015) on tungsten CTE measurements, techniques, sample dimensions, and temperature range are summarized at Table (1).

Table 1. A Survey on Tungsten CTE Measurements

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Researcher** | **Year** | **Used Method** | **Sample Form**  | **Temp. Range K** |
| Fink [5]  | 1910 | Telemicroscopy | Wire 0.005 inch  | 293 - 373 |
| Fink [6] | 1913 | Telemicroscopy | Wire | 293 - 373 |
| Langmuir [7] | 1916 | Telemicroscopy | Filament | 1000 - 2100 |
| Worthing [9] | 1916 | Telemicroscopy | Filament of large cross section  | 1000 - 2000 |
| Worthing [10] | 1917 | Telemicroscopy | Filament 18 cm long  | 563 - 2670 |
| Gray [8] | 1917 | Pushrod  | Rod of 5.6 mm diameter | 173 - 473 |
| Dish [11] | 1921 | Pushrod  | rod | 83 - 673 |
| Goucher [12]  | 1924 | Telemicroscopy | Wires 1 mm  | 283 - 1197 |
| Berlin [13] | 1924 | Telemicroscopy | wire 2.3×140 mm  | 288 -1973 |
| Hidnert [8]  | 1925 | Pushrod  | Rod 4.5×300 mm  | 173 - 773 |
| Becker [14] | 1926 | X-ray diffractometry | Powder | 800 - 2450 |
| Shinoda [15] | 1934 | X-ray diffractometry | Powder | 288 - 1328 |
| Burger [16] | 1934 | Pushrod  | Rod | 298 - 823 |
| Nix [17] | 1942 | Interferometry | Ring | 102 - 301 |
| Demarquay [18] | 1945 | Telemicroscopy | Rod | 945 - 2350 |
| Apbett [19] | 1952 | Recording Dilatometry | Rod | 550 -2850 |
| Mauer [20] | 1955 | X-ray diffractometry | Powder | 273 - 1613 |
| Brand [21] | 1956 | X-ray diffractometry | Powder | 273 - 1573 |
| White [22] | 1958 | Assessment | 83 - 2700 |
| Baun [23] | 1959 | X-ray diffractometry | Powder | 291 - 1246 |
| Fulkerson [24] | 1959 | Pushrod  | Rod 6.7 ×25.4 mm | 293 - 1573 |
| Anthony [25] | 1960 | Pushrod  | Rod 9.4 ×76.2 mm  | 300 - 1616 |
| Levinstein [26] | 1961 | Pushrod  | Rod | 297 - 1422 |
| Anders [27] | 1961 | Optical lever | - | 4 – 10  |
| Deman [28] | 1962 | Pushrod  | Rod | 297 - 1366 |
| Neels [29]  | 1962 | Pushrod  | Rod 6.772 mm long | 2783 - 294 |
| Neels [29]  | 1962 | Pushrod  | Rod 6.772 mm long | 294 - 3025 |
| Houska [30] | 1963 | X-ray diffractometry | Powder | 298 – 2050 |
| Dutta [31] | 1963 | X-ray diffractometry | Powder | 298 - 1151 |
| Anders [32] | 1963 | Optical lever | - | 4 - 10 |
| Ross [33] | 1963 | X-ray Camera | - | 298 - 3373 |
| Amoneko [34] | 1964 | Pushrod  | Rod | 293 - 2273 |
| Totskii [35] | 1964 | Pushrod  | Rod | 273 - 1373 |
| Matyushenko [36] | 1964 | X-ray diffractometry | Disilicides | 300 - 1000 |
| Anders [37] | 1964 | Optical lever | - | 6.6 - 14 |
| Tietz [79] | 1965 | Citation | 83 - 2700 |
| Clark [38] | 1965 | X-ray diffractometry | Powder | 300 - 1499 |
| Takamori [39] | 1965 | Pushrod  | Rod | 848 - 293 |
| Yaggee [40] | 1965 | Pushrod  | Rod | 298 - 1263 |
| V’yugov [41] | 1965 | Telemicroscopy | Wire 2×240 mm | 1937 - 3322 |
| Conway [42] | 1966 | Telemicroscopy | Rod 6.35 × 63.5 mm  | 490 - 2772 |
| Rausch 43] | 1966 | - | Coating | 290 - 2255 |
| Conway [45] | 1967 | Telemicroscopy | - | 533 - 2748 |
| Frantsevich [44]  | 1967 | - | Powder | 293 - 1473 |
| Brizes [46] | 1968 | Telemicroscopy | Rod 6.4×76.2 mm | 782 - 2321 |
| Valentich [47] | 1969 | Pushrod  | Rod | 293 - 2489 |
| Knibbs [48] | 1969 | Optical method | Rod 6.4×76.2 mm  | 1942 - 2558 |
| Nasekovskii [49] | 1969 | Capacitive dilatometry  | - | 77 - 1200 |
| Shah [50, 51] | 1971 | X-ray diffractometry | Powder | 40 - 180 |
| Fitzer [56] | 1972 | Pushrod  | Rod 3.3-6.8 mm diameters | 293 - 1973 |
| Petukhov [52]  | 1972 | Optical method | - | 293 - 3335 |
| Kirby [53]  | 1972 | Telemicroscopy | Rod | 293 - 1800 |
| Kraftmakher [55] | 1972 | Modulation Calorimetry  | Wire of 0.05 mm diameter | 2050 - 2897 |
| Lisovskii [54]  | 1972 | Capacitive dilatometry  | Cylinder 100 mm  | 55 - 300 |
| Novikova [80] | 1974 | Assessment | 173 - 3400 |
| Roberts [58] | 1975 | Interferometry | - | 300 - 1300 |
| Touloukian [59] | 1975 | Assessment | 293 - 3495 |
| Waseda[60]  | 1975 | X-ray diffractometry | Powder |  |
| Slack [81] | 1975 | Assessment | 77-1300 |
| Kirby [61] | 1976 | Interferometry | - | 300 - 1300 |
| White [62]  | 1978 | Capacitive dilatometry  | - | 20 - 90 |
| West [82]  | 1978 | Assessment |  |
| Rodriguez [63]  | 1981 | Pushrod  | Rod | 20 - 300 |
| White [83] | 1983 | Assessment | 250 - 3400 |
| Shevchenko [84] | 1986 | Interferometry | Rod | 373 - 673 |
| In Kook Suh [64]  | 1988 | Pushrod / X-ray diffractometry | Rod 5 × 20mm  | 400 - 1700 |
| Miiller [65] | 1990 | Transient Interferometry | Tube 76 mm long 5.3-6.4 mm diameter | 1500 - 3600 |
| Lahav [66] | 1990 | in-situ stress | Thin film | 293 - 723 |
| Guillermet [69] | 1991 | Assessment / Theoretical Modeling | 300 - 3600 |
| White [70] | 1994 | Assessment | 10 - 3500 |
| White [71] | 1996 | Assessment | 10 - 3500 |
| Dubrovin-sky [67] | 1997 | X-ray / Radiometry | wire | 300 - 3100 |
| Wang [72] | 1998 | Assessment / Theoretical Modeling | 20 - 3500 |
| IAEA [68] | 2006 | Pushrod  | Rod | 300 - 1773  |
| Westengh-ouse [75]  | 2013 | Assessment | 297 - 2773 |
| Dorogoku-pets [73] | 2012 | Assessment / Theoretical Modeling | 100 - 3600 |
| Zhang [74] | 2013 | Assessment / Theoretical Modeling | 300 - 5000 |
| Litasov [76] | 2013 | Assessment / Theoretical Modeling  | 300 -1673 |
| Ritz [77] | 2013 | Pushrod  | Rod | 573 - 1173 |
| Yanwei [78] | 2015 | Pushrod | Rod | 473 - 1273 |

**4. Discussion**

The previous review showed that the many methods are being used to measure the thermal expansion of tungsten namely Pushrod, X-ray, Telemicroscopy, optical levers, laser interferometry, capactive methods and some miscellaneous techniques. The following figure shows their percentage of use (according to Table 1).

**5. Conclusion**

From the preceding survey, one can conclude that most of the tungsten CTE measurements at high temperatures were basically relying on special techniques namely telemicroscopy, pushrod, x-ray, interferometry, modulation calorimetry and transient interferometry which is the most accurate method known till the time of writing this paper.

Fig 1. Dilatometry Methods Used in Studying Tungsten CTE

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