A Short Review on Tungsten Expansivity Measurements, Assessments and Modeling

H. H. Hassan*, S. A. Khairy, H. S. Ayoub

Department of Physics, Faculty of Science, Cairo University, Egypt
*hussamhhassan49@yahoo.com

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:

$$\Delta L/L = \alpha\, \Delta T \qquad (1)$$

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

$$\Delta L/L = c_o + c_1 T + c_2 T^2 + ... + c_n T^n \qquad (2)$$

Where c_o, c_1, $c_{2,...}$,c_n 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 c_o, c_1, c_2,…, c_n. And hence the CTE can be obtained by differentiating the previous equation with respect to T yielding the expression:

$$\alpha = c_1 + 2c_2 T + 3c_3 T^2 + \ldots + nc_n T^{n-1} \qquad (3)$$

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 WWI (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:

$$\Delta L/L_o = 4.4\times10^{-6}\ (T-300) + 4.5\times10^{-11}\ (T-300)^2 + 2.2\times10^{-13}\ (T-300)^3 \qquad (4)$$

By the end of WWI, 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:

$$\alpha = 3.5\times 10^{-6} + 1.4\times 10^{-9}\,T + 2.74\times10^{6}\,T^{-2}e^{-36540/T} \quad (5)$$

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:

$$\Delta L/L_o = 4.266\times10^{-4}(T-293)+8.479\times10^{-8}(T-293)^2-1.974\times10^{-11}(T-293)^3$$
$$293\ K \le T < 1395\ K \quad (6)$$

$$\Delta L/L_o = 0.548+5.416\times10^{-4}(T-1395)+1.952\times10^{-8}(T-1395)^2+4.422\times10^{-11}(T-1395)^3$$
$$1395K \le T < 2495K. \quad (7)$$

$$\Delta L/L_o = 1.226+7.451\times10^{-4}(T-2495)+1.654\times10^{-7}(T-2495)^2+7.568\times10^{-12}(T-2495)^3$$
$$2495\ K \le T < 3600\ K \quad (8)$$

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:

$$\Delta L/L_o = 1.3896\times10^{-3} - 8.2797\times10^{-7}\,T + 4.0557\times10^{-9}\,T^2 - 1.2164\times10^{-12}\,T^3 + 1.7034\times10^{-9}\,T^4 \quad (9)$$

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:

$$\alpha = 7.862\times 10^{-6} + 6.392\times 10^{-9} \quad (10)$$

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:

$$\alpha = 3.872\times10^{-6} + 2.562\times 10^{-9}\, T - 2.8613\times 10^{-12}\, T^2 + 1.9862\times 10^{-15}\, T^3 + 0.58608\times10^{-18}\, T^4 + 0.070586\times10^{-21}\, T^5 \quad (11)$$

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.

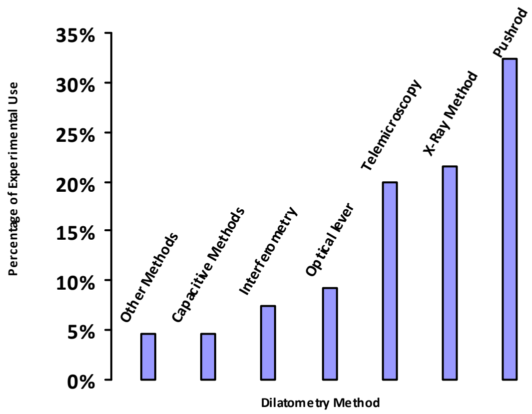

Fig 1. Dilatometry Methods Used in Studying Tungsten CTE

Corresponding Author:
Prof. Dr. H.H.Hasssn
Department of Physics
Faculty of Science
Cairo University, Egypt
E-mail: *hussamhhassan49@yahoo.com

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