Online Supplementary Information

for the paper:

The ITS-90 after definition of neon isotopic reference composition: extent of the isotopic effect tested on previous inter-comparison results

Franco Pavese[1], Anna Szmyrka-Grzebyk[1], Peter P.M. Steur[2]

A. Chemical impurities effect

The *chemical impurities*, reported in the column of chemical corrections of Table 3 of the paper, show that: [1]

- the corrections for these impurities may be even more critical that the ones for the isotopic composition. The experience of the International Project on isotopic neon [13] [2] has shown that the availability from KRISS of excellent assays also on the chemical impurities was a basic asset to obtain an outstanding top-accuracy *overall* correction of the results on the measured samples in that Project;
- The lack of data on some critical impurities, such as hydrogen in neon, can impair the validity of the correction for chemical impurities—and the usefulness of the isotopic correction. [15]

It is the authors' opinion that the CCT should consider the need, for top accuracy, of the application of corrections, by using the SIE or similar methods [S1–3].

Supplementary references

S1. Fellmuth B., Hill K.D., Pearce J.V., Peruzzi A., Steur P.P.M., Zhang J., Guide to the Realization of the ITS-90. Fixed Points: Influence of the Impurities. BIPM, CCT, update of 21 October 2015 (2015)
S2. Pavese F., Metrologia **46**, 47 (2009)
S3. Pavese F., Metrologia **48**, 268 (2011)

B. Specific conditions for each laboratories concerning the K2.(x) comparisons

BNM-INM (now **LNE-LCM**)—The sample BCMH2O of neon used in K2 is *not* traceable to isotopic assays. However, from the STAR inter-comparison [12] it can be inferred that the bottle was the same as for INM sample 99/2, for which an isotopic assay is available [15].

[1] Effect on $T_{tp,Ne}$: for H_2 (-7 ± 3) μK/10^{-6}; for N_2 (-8 ± 3) μK/10^{-6}.

[2] The reference number refers to the main paper References.

Later another measurement was available in the frame of the comparison K2.4 in 2005-2006, using cell Ne02/1 (see Table 2 in the paper also for the next references to samples), for which an isotopic assay exist: this result is also reported.
Finally, the BNM-INM datum in K2 for set #2 was a clear outlier, though within its very large uncertainty.

CNR-IMGC (now **INRIM**)—The sample of neon used in K2 was sealed in cell 3Ne, later outsourced by the IMGC to NIM, and the IMGC reference was moved to cell 1Ne, using a different bottle of gas (see Table 3 in the paper). However, as explained in [4] and its Online Supplementary Information, the thermal results obtained with cells 1Ne and 3Ne are not consistent with the different measured compositions: the difference in the isotopic correction according to the assays amounts to (1Ne – 3Ne) = 195(30) μK. The difference ($T_{1Ne} - T_{3Ne}$) was –44(110) μK in INRIM measurements [4] and –29(76) μK in [12]—the latter will be used in the following. On the other hand, the isotopic assays have some reasons for being less accurate than claimed because the sample available for the assays was quite small: the bottles only contained 2 mmol and 3 mmol, respectively, of gas, decades old. In addition, in the set of observed T_L *vs* ^{22}x values, the T_L(3Ne) looks as an outlier being too low by 100-150 μK, consistent with the above discrepancy. In Table 3 of the paper the values for sample 1Ne are also provided.
INRIM performed a comparison of some key cells in 2008-09 [4]. This is referred in Table 3 as "INRIM" comparison, having INRIM cell Ec2Ne as the reference, now INRIM reference.

KRISS—This laboratory was not considered with respect to the CCT-K2 comparison until recently [S4], when the isotopic composition of the sample of neon used for the (open cell) realisation during CCT-K2 became available. In addition, in [S4] the realisation of the triple point of neon is also reported using samples from three different gas bottles whose isotopic composition is known from a *calibrated* mass spectrometer.

NBS (now **NIST**)—The sample of neon used for the (open cell) realisation during CCT-K2 was taken from a known bottle, whose isotopic assay was made available in 2003. For NIST, it is known that the same gas was used also for the data of CCT-K1 by direct realisations of the ITS-90 using an interpolating constant-volume gas thermometer.

NPL—The sample of neon used for the sealed cell realization during CCT-K2 for thermometer 1728839 was sealed in the NPL cell Ne2 and Ne1: NPL assumed the two cells having been filled from the same bottle, for which isotopic assays became available in 2003. The data for thermometer 213865 came instead from calibration by comparison with the NPL reference thermometer.

NRC—The sample of neon used for the sealed cell realization during CCT-K2 was sealed in the NRC cell F15, taken from a bottle whose isotopic assays became available only in 2003. The same bottle is said by NRC to have been used for sealing cell F17 [S5]. See hereinafter and the main text. [24–26]

PTB— The sample of neon used for the sealed cell realization during CCT-K2 was sealed in the PTB cell Ne-7, taken from a bottle for which isotopic assays became available in 2003. The PTB cell Ne-12, sealed later from the same bottle, is known to be basically identical to cell Ne-7, ΔT = 8(66) μK. Only thermometer 1842379 was used for the K2 equivalence table.
PTB was the co-ordinator of a subsequent comparison [12], reported in Table 3 as Star inter-comparison. INRIM cell Ec2Ne is used as the reference cell also for this exercise.

PRMI (later **VNIIFTRI**)—The sample of neon used for participation to the CCT-K2 is unknown. In all instances, the PRMI withdrew from the K2. Later, a bilateral comparison, K2.1, was performed with NRC. The pilot still used a thermometer calibrated on cell F15, so that no calculation was necessary to refer to the CCT-K2 KCRV. The VNIIFTRI thermometers were calibrated against an *average* realisation of the ITS-90, so that no data on the sample of neon used are available. No correction for isotopic composition can be performed.
In 1978-84, the PRMI participated in the first International Intercomparison of fixed points in sealed cells [34] with sealed cell MC-897, whose relationship with other still-existing sealed cells exist, and quite recently it was re-measured in the frame of the Star inter-comparison [12], where it was found (MC-897 – 3Ne) = 90(92) μK. The isotopic composition of the gas used to fill this cell was provided by the VNIIFTRI with low resolution, corresponding to an uncertainty component of 80 μK.

NMI (then **NMI-VSL**, now **VSL**)— NMI withdrew from its initial participation to the CCT-K2. Later a bilateral comparison, K2.3, was performed with NRC. VSL used a sealed neon cell produced by IMGC, 12Ne. NRC was unable to perform a calibration on the cell F15 used at the time of CCT-K2, and had to use a chain of calibrations to relate the last cell used, Cu-M-1, to cell F15, through cell F17 (53 μK colder than F15). The resulting difference resulted to be (Cu-M-1 – F15) = –165(200) μK. Instead, the same difference obtained from the isotopic composition corrections is –342(95) μK (see more below under INTiBS for NRC). On the Report of comparison K2.3, VSL performed the isotopic corrections for *e*-H2 and H_2O using the official equations available in 2006. On the contrary for Neon, VSL used a home-made evaluation, –157(7) μK; for the latter now the official correction for the neon sample sealed in cell 12Ne is available and is: –123(20) μK. The difference with the VSL estimate is irrelevant considering the uncertainty of the overall temperature deviations.

INTiBS—This Institute participated in a trilateral comparison, K2.4, with BNM and NRC (as the pilot). INTiBS used a sealed neon cell produced by the IMGC (E3Ne), with known isotopic composition. As in the case of K2.3, NRC was unable to perform a calibration based on the cell F15 used at the time of CCT-K2, and had to use a chain of calibrations to relate the last cell used, Cu-M-1, to cell F15, through cell F17. Assuming that cells F15 and F17 were effectively filled from the same neon bottle, the resulting difference resulted to be (Cu-M-1 – F15) = –165(200) μK.

NMIJ-AIST— This Institute participated in a trilateral comparison, K2.5, with INRIM and NRC, where the supplied temperature values are corrected for isotopic composition. Two thermometers were calibrated for this exercise, and the values are supplied for the neon isotopic composition corrected to the reference one [36], one sample being almost coincident with the latter. Only cell Ne-5 is the NMIJ-AIST reference, cell Ne-2 (see INRIM inter-comparison) being known to be hotter by 31(50) μK.

Supplementary references

S4. Yang I., Gam K.S., Joung W., Kim Y.G., Int. J. Thermophysics **36**, 2072 (2015)
S5. K. Hill, Private Communication. The statement could not be verified.

C. Results of other direct-cell comparisons

In a *direct* cell comparison under **European Project MULTICELLS** [38], differences were found as follows, with respect to cell 7Ne, supplied by INRIM to PTB—sealed in 1986 and of the batch 6Ne - 11Ne, the same of cell #20 in this paper): INRIM Eb1Ne 0.024(18) mK; [3] INRIM Ec2Ne –0.065(17); LNE-INM Ne99/2 = 0.31(20) mK. Under the same Project, at VSL the difference (LNE-INM Ne02-1 – INRIM 3Ne) was found to be –0.07(45) mK.

From previous inter-comparisons, the bilateral DoE of the **Comparison Int84**, as reported in [34], are the following with respect to IMGC-CNR reference cell—where the difference between cells 1Ne and 3Ne was then set to 0.00 mK: ASMW (later PTB) 0.34 mK, INM –0.03 mK, NRC –0.04 mK, NRLM = –0.13 mK and PRMI (VNIIFTRI) 0.06 mK; *U* was estimated to be 0.3 mK. INM and VNIIFTRI cells are traceable to present data.

[3] Here the uncertainty in parenthesis is the expanded one, *U*, which is the overall measurement uncertainty budget, so including the isotopic effect.

Good results were also found in the *direct* inter-comparison of sealed cells performed at INRIM [4], more restricted than the STAR inter-comparison of Fig. 4:

(PTB Ne-7 – INRIM Ec2Ne) = 0.057(52) mK; [3]

(NMIJ Ne-2 –INRIM Ec2Ne) = 0.006(57)—whence (NMIJ Ne-5 –INRIM Ec2Ne) = 0.043(57);

(NPL Ne2 – INRIM Ec2Ne) = 0.154(53) mK;

(INTiBS E3Ne – INRIM Ec2Ne) = 0.029(54) mK (from Fig. 2);

(VSL 12Ne – INRIM Ec2Ne) = 0.049(54) mK (from Fig. 2).

Further, (15Ne – Ec2Ne) = 0.054(58) mK; (1Ne –Ec2Ne) = 0.131(66).

Furthermore, using cell INRIM Ec2Ne as reference, via PTB Ne-12 by knowing that it is 0.008 mK hotter than PTB cell Ne-7 as measured in [12] where the latter is the reference, the differences to PTB Ne-7 ar found to be:

(3Ne – Ne-7) = –0.066(45) mK and (12Ne –Ne-7) = 0.062(48) mK, respectively. [3]

In addition, (E1Ne –Ne7) = –0.023(45) mK. Thus, being PTB Ne-12 hotter than INRIM Ec2Ne by 0.054(33) mK —so (Ne-7 – Ec2Ne) = 0.046 mK— one finds (E3Ne – Ec2Ne) = –0.031 mK and (12Ne – Ec2Ne) = 0.108 mK, respectively, and (E1Ne –Ec2Ne) = 0.069 mK.